nature portfolio

Peer Review File

Enhanced aerosol-jet printing using annular acoustic field for high resolution and minimal overspray

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

This paper combines a novel nozzle geometry with a practical and easy to implement acoustic focusing (AF) technique to reduce the printed line width by approximately 60%, generating 5.7 micrometer-wide lines. The AF approach results in a more compact structure, improved conductance and sharper line shapes. Moreover, the printing speed is not compromised. All of these are highly beneficial.

It would be useful to provide more details on the AF mechanism, as well as additional experimental details as outlined below:

1) One of the main claims of the paper is a "real-time, stable and low cost optimization method". The line profiles are first characterised with AF OFF and then the percentage change is reported.

- How does the ink viscosity and/ or particle size change the effectiveness? The necessary ARF will depend on viscosity and particle diameter.

- Standoff distance: Does the standoff distance need to be small for the actuation to be effective? What is the threshold beyond which the ink will start spreading?

- To answer these types of questions, can the authors present an empirical theory linking the line profile (width and/or thickness as well as overspray) with a combination of the following parameters?

o Fluid properties (Viscosity/particle size & density etc.)
o Acoustic enhancement
o Standoff distance
o Sheath flow conditions

This would be extremely useful to evaluate suitability of the technique for different types of inks and process conditions. It would also highlight how the AF is contributing to the line resolution.

As a side note combining the AF with varying flow rates could also be potentially useful to generate line profiles with varying widths, which cannot be easily achieved through existing techniques.

2) Reducing the overspray is another key claim of the paper. The authors should provide numerical values of the overspray in the presence of AF for all of the considered cases. For the ~6 micron-wide lines overspray is approximately 5% (as opposed to 7% overspray reported when AF is OFF to generate 10-micron-wide lines)

3) The stability of the process was tested by printing 20/50 micrometer-wide lines over an 8 hour period. The claimed resolution is around 5.7 microns. It would be good to see how the stability changes for the smallest line width as well. Can the 8-hour stability be maintained or does the approach have different levels of stability corresponding to different resolutions? This may be an important point for readers who may wish to use this approach

4) What is the rms roughness of the printed area when AF is OFF and ON? The authors use the AFM to provide line scans of the widths, but do not share any information on how the surface roughness changes as a function of AF. This may be an important factor for many applications and will be of interest to readers.

5) As the authors claim in the introduction section, aerosol printing is advantageous/preferrable to form lines on arbitrary substrates.

o Would the acoustic enhancement work with different topographies?
o Can the authors comment on the adhesion of the ink to the substrate? Does the adhesion get any better after the AF is tuned on?

Reviewer #2 (Remarks to the Author):

Review for manuscript NCOMMS-23-48632:

Overall, the paper is well written and presented. The figures (especially Fig. 1) are well done and show some work was put into them to follow the standards of Nature Communications. The results being documented are noteworthy both academically and for industry. The paper explores the use of ultrasound radiation forces on particles, annular acoustic focusing (AF), in order to focus the powder stream used in Aerosol jet printing. This was demonstrated to decrease the line width of printed traces by about 60%. At this stage of the review process, I would like to see the following mayor revisions addressed to increase the quality of the manuscript.

1. (Section 1) In line 33, change the words 'the sharp corner' to 'sharp corners'.

2. For the paragraph starting in line 44, include not only what was investigated in previous work, but also what their conclusions were. This is also an opportunity to link previous work to the current work. For example, in reference 31, one can describe how the ratio of sheath gas flow rate to carrier gas flow rate affected the final resolution, and then mention excessive sheath gas flow can only focus particles only up to a certain limit, due to the flow field, turbulence, etc.

3. Reword paragraph starting at line 60 to link it better to the article; wasn't AF presented previously? Maybe move the sentence starting with 'One promising approach to micro-manipulate particles of different sizes in a fluid is AF' to an earlier part of the article?

4. In line 65, it is not clear what 'three-dimensional flow form' means. Does this mean 'the shape of the 3D flow field'?

5. In line 71, briefly mention sound will be used to focus the particles, hence the annular piezoelectric tube is used.

6. The introduction section needs a lot of work to increase its comprehensiveness, in particular tying the current work in the article with the following fields: other fields that have used ultrasound for sorting particles and/or focusing a powder stream, other ways particles have been focused besides acoustic radiation forces, and work done within additive manufacturing to focus a powder stream using ultrasound. To address these gaps, the authors should consider incorporating the following recent research:

Cicek, Ahmet, et al. "Acoustophoretic separation of airborne millimeter-size particles by a Fresnel lens." Scientific reports 7.1 (2017): 43374.

Choi, Hoseop, et al. "Controlled electrostatic focusing of charged aerosol nanoparticles via an electrified mask." Journal of Aerosol Science 88 (2015): 90-97.

Pan, Yong-Le, et al. "Opto-aerodynamic focusing of aerosol particles." Aerosol Science and Technology 52.1 (2018): 13-18.

Huang, Yuze, Mir Behrad Khamesee, and Ehsan Toyserkani. "Electrodynamic concentration of non-ferrous metallic particles in the moving gas-powder stream: Mathematical modeling and analysis." International Journal of Magnetics and Electromagnetism 5 (2019): 019.

Smith, Peter H., et al. "Magnetic manipulation in directed energy deposition using a programmable solenoid." Journal of Materials Processing Technology 299 (2022): 117342.

Martinez-Marchese, A., et al. "On the application of sound radiation force for focusing of powder stream in directed energy deposition." Ultrasonics 127 (2023): 106830.

Martinez-Marchese, A., et al. "Ultrasound particle lensing: Powder stream control in directed energy deposition using acoustic radiation force fields." Additive Manufacturing 73 (2023): 103698.

7. (Section 2) In line 91, clarify or correct the statement 'the ink undergoes turbulent dynamics, giving rise to Stokes drag force'. Does turbulence cause Stokes drag? Expand on the effect on turbulence and particle size on the powder stream characteristics; one thing to look for in the literature that is used to quantify this is the Stokes number. Is this drag referring to the particle's acceleration mechanism? If so, mention it.

8. In line 125, what is it meant by 'forms the propagation of acoustic waves'? Instead of this statement, one could use 'produces the acoustic waves'.

9. Include references for Eqs. 1, 2, 4 and 5. Unless the equations are simple and self-evident such as Eqs. 7 and 8, go over the article to make sure the equations being used are either derived or properly referenced.

10. In line 188, '30 m/s from the published result', what published result? In this article? If so, use 'reported result'.

11. In line 204, how is the over-spray determined? Please expand on what is meant by over-spray in the article.

12. (Section 3) Regarding the model and COMSOL simulation, one thing that is not specified in a lot of the published literature is the boundary pressure amplitude used (in Pa or dB or both). How was this determined? Could the pressure amplitude used be linked back to the piezoelectric tube manufacturing specifications or other measurements?

13. Include more information on the Lagrangian particle tracking used in the simulation; how many particles where used? How are they initially distributed in position and velocity? What was the integration time step? Drag models, etc.? How was the width decrease determined from the final particle positions? This is essential to reproduce the simulation results.

14. (General remarks) One thing that would greatly improve this article would be the addition of summed images from a high speed camera or even from a single photograph of the powder stream in order to know the particle density distribution (PCD) in the powder stream. This has been done numerous times in the additive manufacturing literature, for example in:

Zeng, Minxiang, et al. "High-throughput printing of combinatorial materials from aerosols." Nature 617.7960 (2023): 292-298.

Ansari, M., et al. "A mathematical model of laser directed energy deposition for process mapping and geometry prediction of Ti-5553 single-tracks." Materialia 12 (2020): 100710.

The rationale for why a photograph indicates the PCD can be found in:

Pinkerton, Andrew J., and Lin Li. "Modelling powder concentration distribution from a coaxial deposition nozzle for laser-based rapid tooling." J. Manuf. Sci. Eng. 126.1 (2004): 33-41.

If the assumptions used in the above article do not apply, and/or the equipment is not available, another option would be obtaining an estimate from the COMSOL model. This can be done by running the simulation and recording the final particle positions at a common plane normal to the nozzle. Then counting the number of particles (making sure the number is statistically significant, see Monte Carlo Methods by Kalos) at different radial widths from the center of the nozzle and normalizing using the mass flow-rate to give a concentration value.

15. Another thing that could be commented on is the mass balance of this process; does the track cross-sectional area match with the mass flow-rate from the nozzle? Does the sound affect this by changing over-spray for example?

16. The reason for obtaining the PCD above the traces is that it could be used to describe the change in width (as well as the geometry of the transition region shown in Fig. 2) when the sound is being applied. This could be done by considering the track geometry to be related via a

convolution with the PCD. This has been done in the context of cold spray additive manufacturing:

Venturi, F., N. Gilfillan, and T. Hussain. "A convolution-based approach to cold spray additive manufacturing." Additive Manufacturing Letters 1 (2021): 100014.

17. (Videos) For the videos, instead of using using the phrase 'Acoustic ON/OFF', the phrase 'Acoustic Focusing ON/OFF' could be used.

Reviewer #3 (Remarks to the Author):

The authors demonstrate a novel and readily applicable advancement to improve the overall performance of aerosol jet printing technology. This approach is conceptually defined through simulation and calculations, and experimentally demonstrated by analysis of printed traces for line width, overspray, and conductivity. The results presented justify the claims well and the annular acoustic focusing is shown to be a robust and practical method. Although no device applications are shown to further highlight the advantages of the improved printing method, this work is very well done and should be accepted with only minor revisions according to the points made below:
1) Line 39: "Conversely…" This sentence is worded in a somewhat confusing way. It seems to be saying the presence of overspray reduces the material density of a given trace, but the wording is hard to follow.
2) In Figure 1g, Stokes drag force is shown to be pointing in the same direction as the particle is being jetted but drag force should actually point in the opposite direction from particle motion. Also "flow filed" seems to be a type for "flow field".
3) Line 181: "tapper" seems to be a typo.
4) Line 198: It would be good to share the optical image results of the printed traces at frequencies from 5 – 7 MHz so it can be understood why 5.8 MHz was chosen as optimal. This could also provide some insight into what printed traces look like when there are undesired pressure nodes present.
5) Line 199: It is briefly mentioned that the optimal frequency of 5.8 MHz would be almost identical regardless of ink, but this isn't fully substantiated. Is this because of the nozzle geometry or are the material properties not as significant in determining the optimal frequency?
6) In Figure 5a, sample A and sample B should have their own colors that are consistent between the thickness measurements and line width measurements.
7) Line 356: "acoustic acoustic-focused" seems to be a typo.
8) More detail on the methods used to manufacture the custom nozzle should be provided given the uniquity of a custom-made aerosol jet nozzle.

Reviewer #4 (Remarks to the Author):

The concept presented in this paper to use annular acoustic focusing to improve the resolution of aerosol jet printing is novel and of interest to the manufacturing community. I have a number of serious concerns regarding the methodology and results claimed in this study as well as several questions about the modeling. I will also point out that the authors use a number of variables without defining them throughout the document.

In section 1, I have the following concerns/questions:
• There are a number of ambitious and weighty claims made about existing literature without appropriate citations. In lines 35 & 56, claims of 'little success' are made without any definitive, quantitative statement about the current state of the field. Some concrete examples of what has been achieved will greatly add to this discussion.
• How significant must overspray be before seeing the issues mentioned in lines 41-43? Overspray is typically a small portion of the printed volume and should not have a large effect on conductivity.

• The first two paragraphs of the introduction should be combined and reorganized.

In section 2.1 (and relevant methods section), I have the following concerns/questions:
• The claim made in line 94-96 about the reason for ink flow diameter blames 'environmental disturbances'. Given that this is determined in the nozzle itself, what disturbances do the authors mean? In addition, 'limited pneumatic focusing' should include an explanation about why there is a limit to pneumatic focusing.
• The model appears to be built on previous work studying the acoustic forces on microparticles in a liquid-filled microchannel (the author's reference 48). There is no discussion on the potential applicability of this model in aerosol jet printing, which utilizes nanoparticles suspended in microdroplets (hence aerosol). This is a large assumption which the authors must thoroughly justify in order for the modeling in this work to be plausible.
• The work described in lines 174-178 needs much more description.

In section 2.2 (and relevant methods section), I have the following concerns/questions:
• Given that aerodynamic properties dominate aerosol jet, and a new nozzle is used here, it is imperative that that design is included in the main text. The authors do not even mention the nozzle diameter until much later in the work. In addition, the work shown here should be compared to typical commercially-available nozzles. Without this, it is impossible to evaluate the effectiveness of this new nozzle alone, which from what is here seems to be impressive in its own right.
• This entire section has severely inadequate details about the work, including the details in the later methods section. Details about the piezoelectric actuator, for example, are vital to evaluating and reproducing the outcomes achieved here and must be included.
• There is a mention in line 194-195 that differences between simulations and reality could be the result of material property variation. Nowhere does this work mention what material properties (density, specific heat, viscosities, thermal conductivity, etc.) were used in the simulations.
• In lines 195-200, the authors claim to have found an optimum frequency but do not show any data to demonstrate that claim. In addition, the range of frequencies considered for experimentation include frequencies higher than those included in the modeling. Why were higher frequencies not considered in the modeling section?
• The mention of the necessity of a power amplifier in lines 200-202 does not include details about what model or specifications were used here.
• In line 204, values for line width and overspray (presumably width although the manuscript is not clear) include no details about the statistical significance of this data. How were these values measured. Given the nearly Gaussian curve the authors claim, what was the criteria used to determine the edge of the line and the edge of the overspray? This is not a trivial question and will have a significant effect on the results of this study. Without these details, this number provides little meaning.
• 'minimal overspray' in line 207 has no meaning. This type of result must have some quantification of the detection limits of the measurement and analysis technique. Moreover, this reviewer disputes this claim based on Fig. 2a-b. The overspray size seems nearly identical in both cases.
• Differences before and after the AF region of a line are attributed to process drift in line 210. This reviewer would like to see an analysis on how often line dimensions change positively or negatively. Without such analysis of a large sample size, the effect of AF (or lack thereof) cannot be stated with any degree of confidence.
• Line 214 – what is meant by 'channel'?
• Line 218 – can the jet velocity be compared to inlet gas flows or some reference to ensure its reasonableness? Again, all of these values should come from multiple measurements and be presented with some measure of data spread.
• What substrate material was used here? And what platen temperature? Both of these will have a large effect on line width.

In section 2.3 (and relevant methods section), I have the following concerns/questions:
• Why for 'repeatability' tests does there only seem to be one cross-section measurement taken for each data set? Without multiple sections across multiple prints, the repeatability cannot be assessed. In addition, there is a high precision given on measurements here – this again begs the question of how the 'edge' of such a curve was determined.

• The authors define line thickness using the largest measurable – this is an acceptable choice, but I must again question how this can be a statistically significant, meaningful value based on a single measurement.
• The definition of 'change rate' is not given.
• It is again unclear to this reviewer how the data in Fig. 3e can be considered proof of repeatability without some quantitative measure of line dimensions, particularly when the authors admit there are variations. There is no evidence on convincing reference that these variations can be 'primarily attributable to the fluctuation in gas flow'. I would also dispute that these results can be combined with others to prove the techniques applicability to multiple directions.
• Lines 256-263 give the only true measure of repeatability in this section, but vital details are again omitted here. How many data points are used in these values? What statistical measure is used in the +/-? Analysis is again qualitative in describing overspray as 'tiny' and of questionable veracity. Fig. 3f has visible overspray, particularly in the inset. This again brings the question of how is overspray being measured? As an average deviation from the line? Given that electrical shorting is the concern, it seems more useful to measure maximum distance at which overspray is found or some measure of density vs. distance from the line's edge. Much more detail is required to describe the work.
• Fig. 2b – why the apparent lateral shift before and after the AF?
• Fig. 2c & d – the is an apparent shoulder in the AF ON lines – is this not evidence of measurable overspray that could be quantified?

In section 2.4 (and relevant methods section), I have the following concerns/questions:
• The data in Fig. 3a-d and Supplementary Table S2 do not seem to agree on quantities. Given the definition of line thickness, line width, and apparent Gaussian cross-section, the values given for cross-sectional area seem to be quite large. It seems warranted to check these values again for any calculation error or provide further details on how this area was calculated.
• In line 283, it is suddenly mentioned that the traces were 'annealed'. None of the conductivity data has any meaning whatsoever without details about the heating process. Was this merely drying the traces? Given the ink used, the manufacturer gives a range of resistivities equal to about 2.5 orders of magnitude from 120C to 200C. These values also cover the entire range of conductivities shown in this work. Given this information, the method of heating the samples and is vitally important to evaluating the degree of conductivity as well as how likely any differences can actually be attributed to the effects claimed here.
• The definition of 'change rate' is not given.
• In Fig. 4a & b, what do the error bars represent?
• Line 293 – was particle size checked before or after the annealing process? Small differences could be obscured by the onset of sintering.
• How were the samples cross-sectioned? The images appear to be different in geometry. With poor adhesion between particles, the method of sectioning can have a large effect on the observed morphology.
• Without more details the porosity measurement is not meaningful. Algorithms to measure porosity on a cross-section which is not flat and polished can have high error. Images showing the detected pores on top of the relevant image would be one way of demonstrating this.
• Density is also limited by the stacking of circular particles. A comparison of the relative contribution is warranted. That is to say – how close or far to the theoretical maximum packing density is either case discussed here?
• The statements in lines 304-313 seem quite out of place. This information is relevant during the modeling portion of the work more than in this location (as mentioned above in previous comments).
• In Fig. 4i, you show the coalescing of microdroplets as a result of nanoparticle focusing. To this reviewer's best judgement, none of the work here proves this mechanism. A smaller material stream with consistent volumetric flow will have a higher velocity. It is, therefore, possible that droplets in the aerosol stay the same size and merely their arrangement changes. Unless there is compelling evidence of droplet size increasing (already expected to be 1-5 um in diameter, close to the minimum line width shown here), this schematic should be reconsidered or given with sufficient disclaimer that it is merely a hypothesis of the flow state.

In section 2.5 (and relevant methods section), I have the following concerns/questions:
• Why was printing stability not measured for the 6 um line width? Without this information, the

value of this work is significantly diminished. Its lack also makes this reviewer concerned about the actual viability of the claims in this work.

- Line widths seem always above the planned values in Fig. 5a. Why?
- All of the discussion in line 330-338 is speculation without data from smaller nozzles. In addition, clogging issue in aerosol jet are a matter of statistics rather than a yes/no scenario. Actual tests with multiple nozzle sizes would give statistical meaning to this analysis. Otherwise, it should be removed from this section entirely.
- Fig. 5b aggregates data but adds little without additional discussion on the methods used in the cited works. Also, how was overspray compared between these works? It is highly material and process parameter dependent.

As a general comment, the authors attribute multiple changes to process drift without any evidence and between claims of efforts made to mitigate process drift. Please reconsider the veracity of these claims. In addition, authors claim this work is 'material agnostic' in line 349, while only demonstrating one material. Although AJ printing has little dependency on the target material, the work shown here depends on the fluid properties of solvent and additives. As such, this phrase should be removed or proven.

Overall, it is unclear to this reviewer whether the claimed improved resolution of 6 um can actually be achieved readily using this method. The data shown here seems to be the result of one or two successful prints without any measure of statistical significance. Without much more detail, likely several pages of methodology, and many repetitions in experimental data, it is impossible to have any confidence in the repeatability or impact of this work. As such, I must recommend rejection of this manuscript at this time, despite the novelty of the idea presented here.

Response to Reviewers of NCOMMS-23-48632

We express our sincere gratitude to the reviewers for their insightful comments and to the Editor for granting us the opportunity to refine our manuscript (NCOMMS-23-48632) entitled "Enhanced aerosol-jet printing using annular acoustic field for ultrahigh resolution and minimal overspray with improved conductivity" submitted for publication on Nature Communications. After thoroughly considering the comments provided carefully, we concur that the manuscript has undergone significant improvement through the incorporation of the suggested amendments. We have highlighted all revisions in red in the revised version. In the following sections, the reviewer's comments are in black, our responses to the comments are in blue and red indicates the text added/modified in the manuscript.

Reviewer #1 (Remarks to the Author):

This paper combines a novel nozzle geometry with a practical and easy to implement acoustic focusing (AF) technique to reduce the printed line width by approximately 60%, generating 5.7 micrometer-wide lines. The AF approach results in a more compact structure, improved conductance and sharper line shapes. Moreover, the printing speed is not compromised. All of these are highly beneficial.

It would be useful to provide more details on the AF mechanism, as well as additional experimental details as outlined below:

Response

We express our gratitude to the reviewer for the astute observations and affirmative evaluation of our manuscript. It is gratifying to learn that our innovative integration of a novel nozzle geometry with AF was perceived as both original and advantageous. We appreciate his/her constructive feedback immensely and offer a detailed response to each comment provided by the reviewer in the following.

Comment #1

One of the main claims of the paper is a "real-time, stable and low cost optimization method". The line profiles are first characterized with AF OFF and then the percentage change is reported.

- How does the ink viscosity and/ or particle size change the effectiveness? The necessary ARF will depend on viscosity and particle diameter.
- Standoff distance: Does the standoff distance need to be small for the actuation to be effective? What is the threshold beyond which the ink will start spreading?
- To answer these types of questions, can the authors present an empirical theory linking the line profile (width and/or thickness as well as overspray) with a combination of the following parameters?

o Fluid properties (Viscosity/particle size & density etc.)

o Acoustic enhancement

o Standoff distance

o Sheath flow conditions

This would be extremely useful to evaluate suitability of the technique for different types of inks and process conditions. It would also highlight how the AF is contributing to the line resolution.

As a side note combining the AF with varying flow rates could also be potentially useful to generate line profiles with varying widths, which cannot be easily achieved through existing techniques.

Response

We are grateful that the reviewer appreciated that the AF strategy is of availability and proposed such systematic suggestions for empirical theory construction. We have analyzed and concluded on the influencing mechanism and planning methods of these four types of parameters separately. Among them, for fluid properties, we conducted theoretical analysis based on force-motion relationships; for acoustic enhancement, we have provided a complete explanation in Section 2.2 and briefly summarize it here; we carried out experiments to detect the influence of AF for its standoff distance; Summarily, the relationship between printing resolution and these four types of printing parameters under AF would be complicate and different from ordinary AJ printing condition. We think it needs a more systematic and comprehensive work and plan to focus on this interesting work in our future studies. To clarify this issue, we have added the following statement in the revised manuscript.

"*It should be noted that the radially focusing behavior of AF is primarily influenced by the acoustic radiation force (Eq. 5) which is determined by the ink properties including the viscosity, particle size and solid concentration. For different types of ink, preliminary calculations can be conducted using simulation models developed in this study to evaluate the achievable focus ratio. Here, the primary determinant of acoustic enhancement effectiveness lies in the optimal focusing frequency, which is determined by the nozzle geometry. For optimal focusing effect in this study, the acoustic wave's pressure node should be positioned at the neutral axis of nozzle. This positioning requires an optimal distribution of acoustic pressure that is independent of the material properties. However, the optimal frequency may experience adjustments as needed when connected to a potentially superior nozzle with a distinct channel configuration for different inks. To explore the influence of AF for the standoff distance, AF-assisted AJ printing experiments were conducted with different standoff distance ranging from 1 mm to 6.5 mm. As depicted in Fig. S10, the LW demonstrates a trend of initial decline followed by an increase as the standoff distance increases for both situations with the AF ON and OFF. This pattern aligns with previous findings on the AJ printing performed without AF, indicating an optimal standoff distance of 2.5 mm that results in the minimal LW for both scenarios. It has been observed that the maximum standoff distance required to maintain effective print quality in AJ printing with AF is 6 mm, whereas without AF, it is limited to 5 mm. This variation is attributed to the superior radial particle concentration achieved by AF, which exhibits an enhanced shape-preserving capability after exiting the nozzle. Regarding to the printing parameters such as carrier gas flow, sheath gas flow, and printing speed, they are still following the established guidelines from our previous observations. In future studies, a more systematic and comprehensive work should be done to fully explore the relationship between printing parameters and LW with AF ON.*" (please see page 19 in the revised manuscript)

"***12 Standoff distance test***

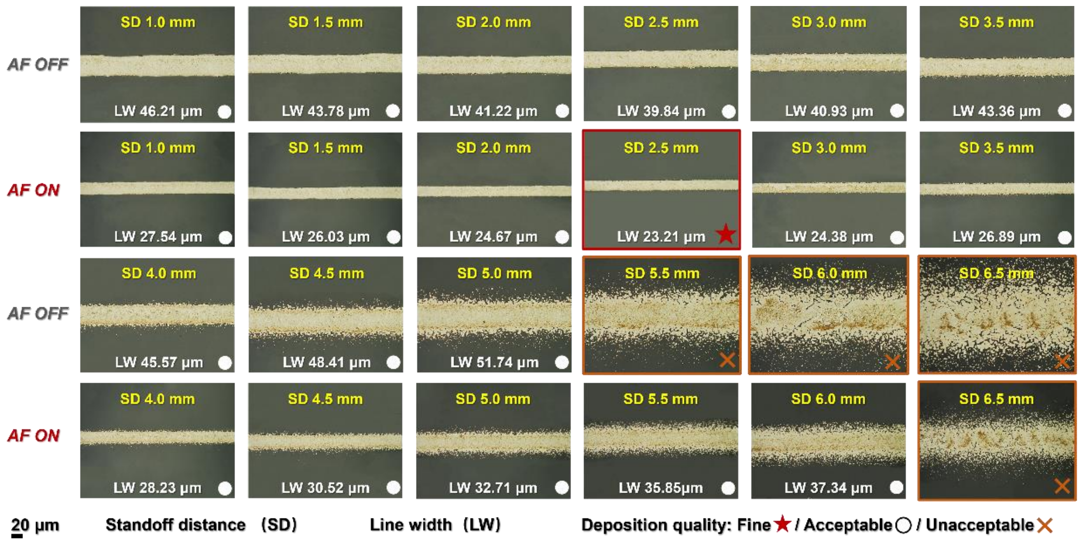


Fig. S10 Images of AF-assisted AJ printed traces with different standoff distance (ranging from 1.0 mm to 6.5 mm)" (please see page 10 in the revised supplementary information)

Comment #2

Reducing the overspray is another key claim of the paper. The authors should provide numerical values of the overspray in the presence of AF for all of the considered cases. For the ~6 micron-wide lines overspray is approximately 5% (as opposed to 7% overspray reported when AF is OFF to generate 10-micron-wide lines).

Response

We are grateful for this perceptive observation concerning the reduction of overspray in our research. This comment will substantially improve the clarity and rigor of our work. First, we have defined the LW in this work and especially the overspray, both using the concept of average LW. Then, we have used this method to comprehensively quantify and analyze the overspray of all printing results in this work.

"*We also proposed a new print nozzle geometry, resulting in ultrafine traces (line width* <*6 μm, overspray* <*0.1 μm)*." (please see page 1 in the revised manuscript)

"*Experimentally, an ultrafine-printed Ag ink stream with a line width (LW) of 5.71 μm and overspray of 0.08 μm was achieved under AF, the corresponding focusing and defocusing process were observed which indicates the focusing behavior is fully reversible and omnidirectional.*" (please see page 4 in the revised manuscript)

"*As illustrated in Fig. 2b, an ink stream with a LW of 5.71 μm and a minimal overspray of 0.08 μm was observed without altering any printing parameters.*" (please see page 10 in the revised manuscript)

"*The overspray for the ~6 μm traces is estimated to be ~2%, which is lower than the reported 7% overspray when the AF is OFF to generate 10 μm traces.*" (please see page 11 in the revised manuscript)

“

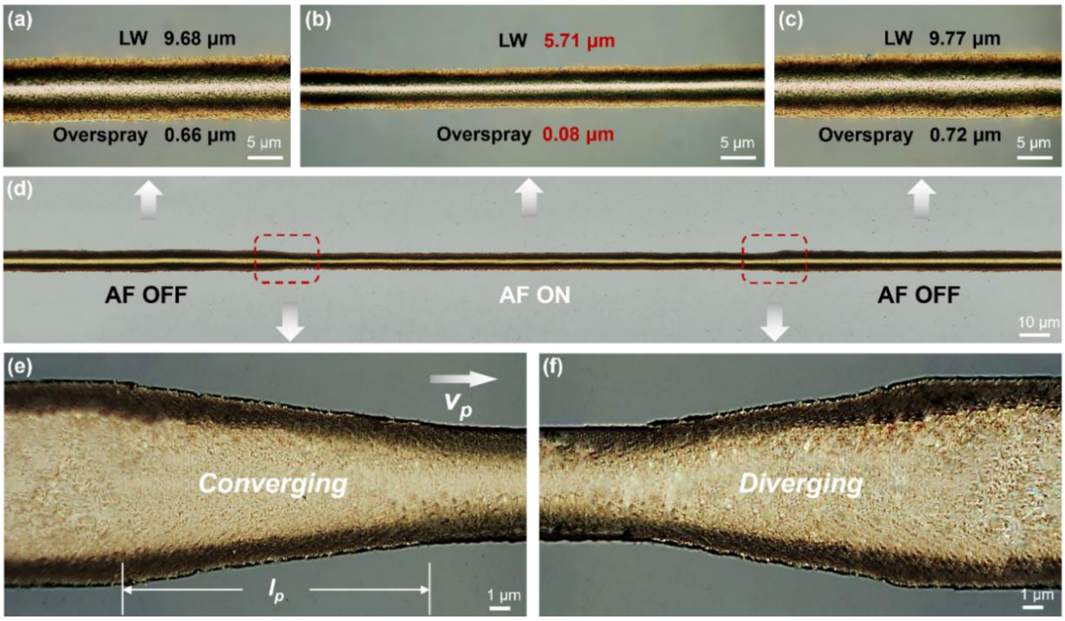


Fig. 2 The impact of AF on the geometric morphology of AJ printed line. ……” (please see page 12 in the revised manuscript)

“*As a result, it achieves an ultra-high resolution in AJ printing, up to 5.7 μm, while minimal overspray occurred to less than 0.1 μm, which completely breaks through the results of previous optimization studies, as shown in Fig. 5c.*” (please see page 18 in the revised manuscript)

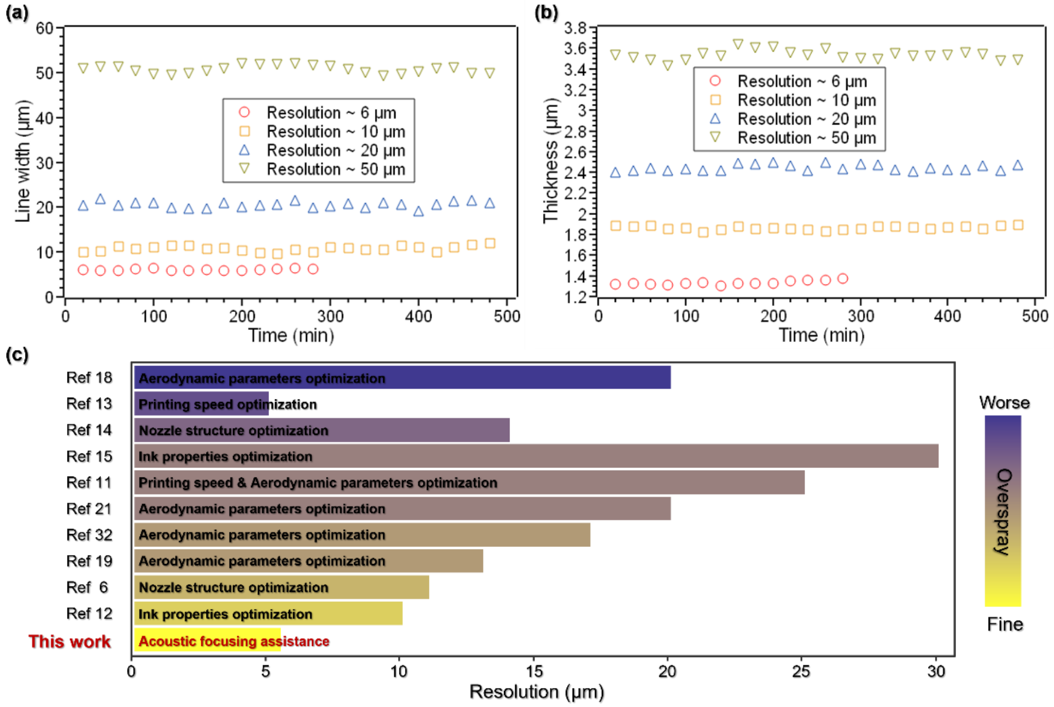
Comment #3

The stability of the process was tested by printing 20/50 micrometer-wide lines over an 8 hour period. The claimed resolution is around 5.7 microns. It would be good to see how the stability changes for the smallest line width as well. Can the 8-hour stability be maintained or does the approach have different levels of stability corresponding to different resolutions? This may be an important point for readers who may wish to use this approach.

Response

We agree with the reviewer’s comment that the stability for the smallest line width would be a very important point for readers who may wish to use this approach. Therefore, this has now been discussed in the revised manuscript.

“*To assess AJ printing stability with AF, four printing stability tests were conducted under ambient conditions, which involved printing lines with the resolution of 6, 10, 20, and 50 μm respectively. Among these, the stability of 6 μm resolution lasted for approximately 4.5 hours. This duration was notably shorter compared to the other three resolutions with 10 μm, 20 μm, and 50 μm, which all maintained stability for almost 8 hours. The discrepancy can be attributed to the instability observed in the original optimal resolution of 10 μm without AF, but still has greatly improved stability under ultrahigh printing resolution. An optical image was captured every 20 minutes as shown in Supplementary Fig. S9, the LW, and thickness of the trace at various time points can be viewed in Fig. 5a, b. Although a small variation of LW was observed during the long-term printing, the average LW (6.0±0.3 μm, 10.8±1.2 μm, 20.6±1.4 μm, 50.7±1.4 μm) of the AF-assisted AJ printed traces indicates that the excellent printing stability compared to traditional AJ printing method. Compared to the stability shown by the LW approach, there was a significant fluctuation in trace thickness which suggests that the deposition rate varies during* *printing, a common occurrence in practical applications. Specifically, there is a variation in deposition volume with consistent width but noticeable difference in height. However, this highlights the optimizing impact of AF in AJ printing to maintain the quality and stability of printed LW despite fluctuations in deposition rate.*" (please see page 18 in the revised manuscript)



"

Fig. 5 Stability quantification and performance comparison of AF-assisted AJ printing. a *For 8-hour continuous AJ printing with resolution ~6 μm, ~10 μm, ~20 μm, and ~50 μm,* ***a*** *LWs and* ***b*** *thickness were recorded at intervals of 20 mins.* ***c*** *Comparison of different optimization methods on printing resolution and overspray of AJ printing with other reported results.*" (please see page 19 in the revised manuscript)

"***11 Printing stability test***

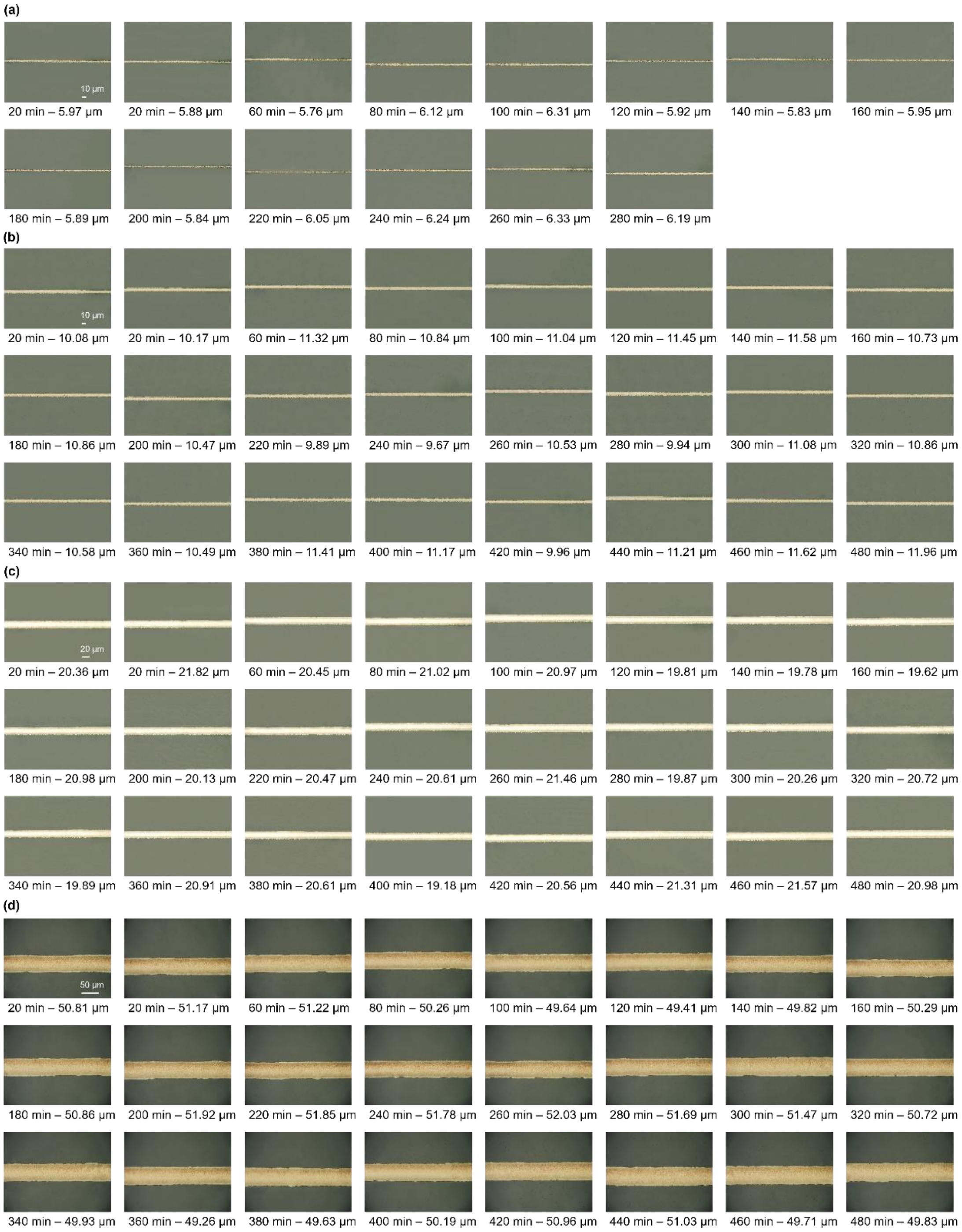


***Fig. S9** Continuous AJ printing assisted with AF, during which the morphology and geometric values of the printed line were measured in steps of 20 minutes. **a** LW ~6 μm for 4.5 hours. **b** LW ~10 μm for 8 hours. **c** LW ~20 μm for 8 hours. **d** LW ~50 μm for 8 hours.*" (please see page 9 in the revised supplementary information)

<u>Comment #4</u>

What is the rms roughness of the printed area when AF is OFF and ON? The authors use the AFM to provide line scans of the widths, but do not share any information on how the surface roughness changes as a function of AF. This may be an important factor for many applications and will be of interest to readers.

Response

We agree with the reviewer's comment and understand that this is an important issue. Therefore, this has now been discussed in the revised manuscript.

"*In addition, the surface RMS roughness of AJ printed traces before and after AF at different printing resolutions was quantified based on the AFM measurement results, as shown in Fig. S11a-h. Overall, AF has a certain effect (≤10%) on reducing the surface roughness of depositions, as shown in Fig. S11i.*" (please see page 13 in the revised manuscript)

"***13 RMS roughness analysis***

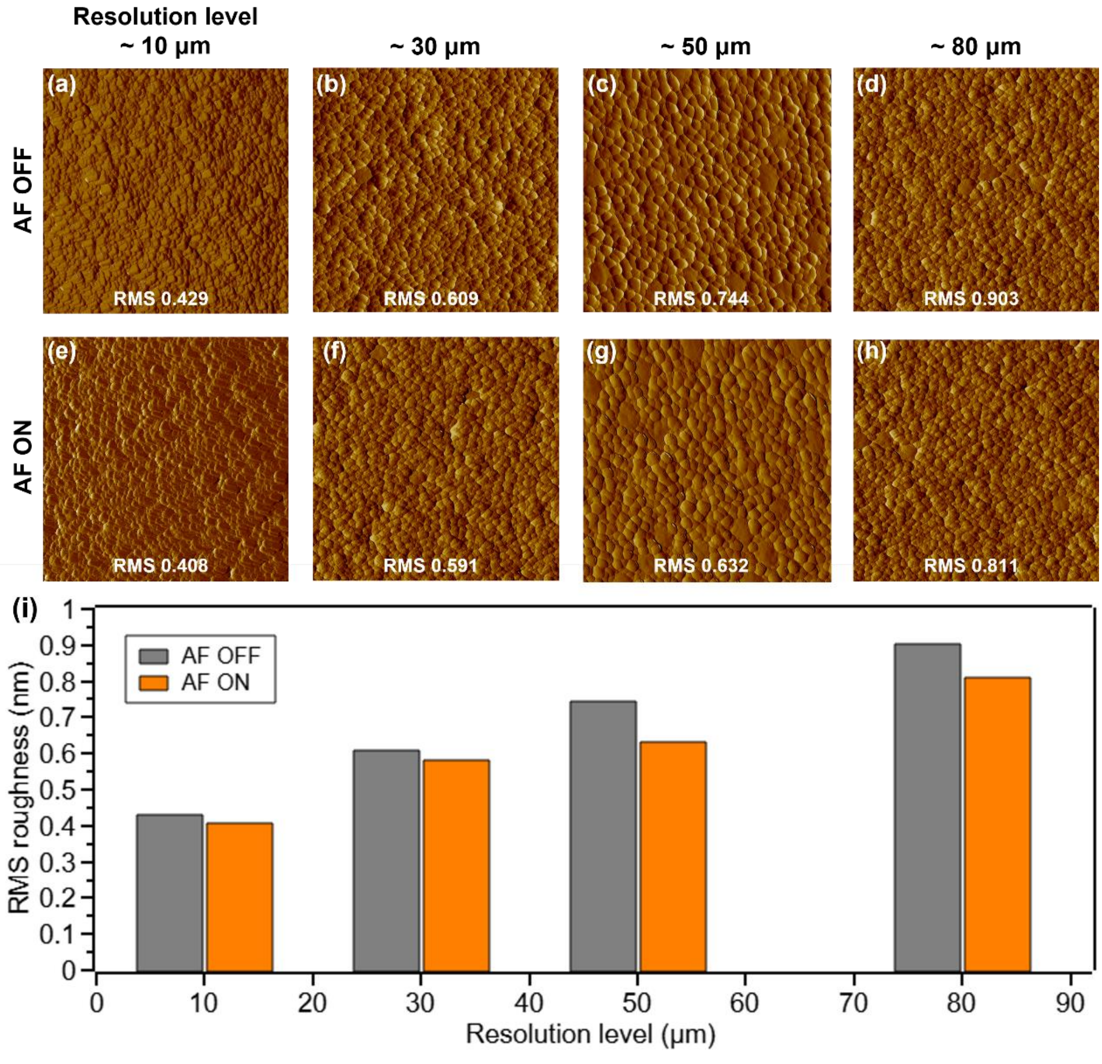


***Fig. S11 a-h** Surface morphology with corresponding RMS roughness of the AJ printed deposition at different printing resolutions (~10, ~30, ~50, ~80 µm) before (a, b, c, d) and after (e, f, g, h) AF measured by AFM. **i** Analysis of the change in RMS roughness before and after AF.*" (please see page 10 in the revised supplementary information)

Comment #5

As the authors claim in the introduction section, aerosol printing is advantageous/preferable to form lines on arbitrary substrates.

o Would the acoustic enhancement work with different topographies?

o Can the authors comment on the adhesion of the ink to the substrate? Does the adhesion get any better after the AF is turned on?

Response

We express our gratitude to the reviewers for their insightful suggestions, proposing novel innovative aspects and potential directions for application derived from AF strategy to AJ printing. We have now addressed it in the revised manuscript by adding the following sentences.

(1) Based on AJ printing inherently possesses conformal printing capabilities adaptable to substrates of any shape or material, and considering the supplementary acoustic enhancement that applies radial focusing to ink particles only throughout the aerosol ink flow's jetting process rather than at the substrate, this technique is not limited by the nature of the substrates. Consequently, it can be employed on substrates of any shape or material.

"*It is noted that AJ Printing's conformal printing technique is capable of adapting to various substrate shapes and materials, due to the utilization of AF on ink particles during the jetting process rather than on the substrate. This method is still applicable to all types of substrates.*" (please see page 20 in the revised manuscript)

(2) We have investigated the effect of AF on the interfacial adhesion between ink and substrate. However, the experimental outcomes did not indicate any variation in the adhesion properties of the ink to the substrate, irrespective of the presence of AF. Additionally, the scanning electron microscopy analysis, as depicted in Fig. 4c-h, revealed few substantial alterations in the surface morphology of the AJ printed deposition, both pre- and post-application of AF. This observation leads us to infer that the morphology of the surface in contact with the substrate remains consistent, thereby not affecting the adhesion of the ink to the substrate. We deduce that the interfacial adhesion is a manifestation of the forces acting perpendicularly to the interface between the ink and the substrate. However, the acoustic radiation force, induced by the acoustic field, acts radially concerning the ink stream and tangentially to the substrate interface. Consequently, the direction of the acoustic radiation force is orthogonal to that of the interfacial adhesion forces. This orientation suggests that the influence of AF has not yet produced a measurable effect on interfacial adhesion. In future research work, we will continue to pay attention to the issue of interfacial adhesion of the ink to the substrate of AJ printing based on your insightful concern thank you again.

"*In addition, we have verified the impact of AF on interfacial adhesion. The results in Fig. S12 showed that AF still maintained the interfacial adhesion within the classification of 5B specified by the standard*[64]." (please see page 20 in the revised manuscript)

"*According to the standard test methods for measuring adhesion by tape test*[64]*, both pads printed with/without AF were subjected to grid processing and tape peel test.*" (please see page 23 in the revised manuscript)

"***12 Adhesion test***

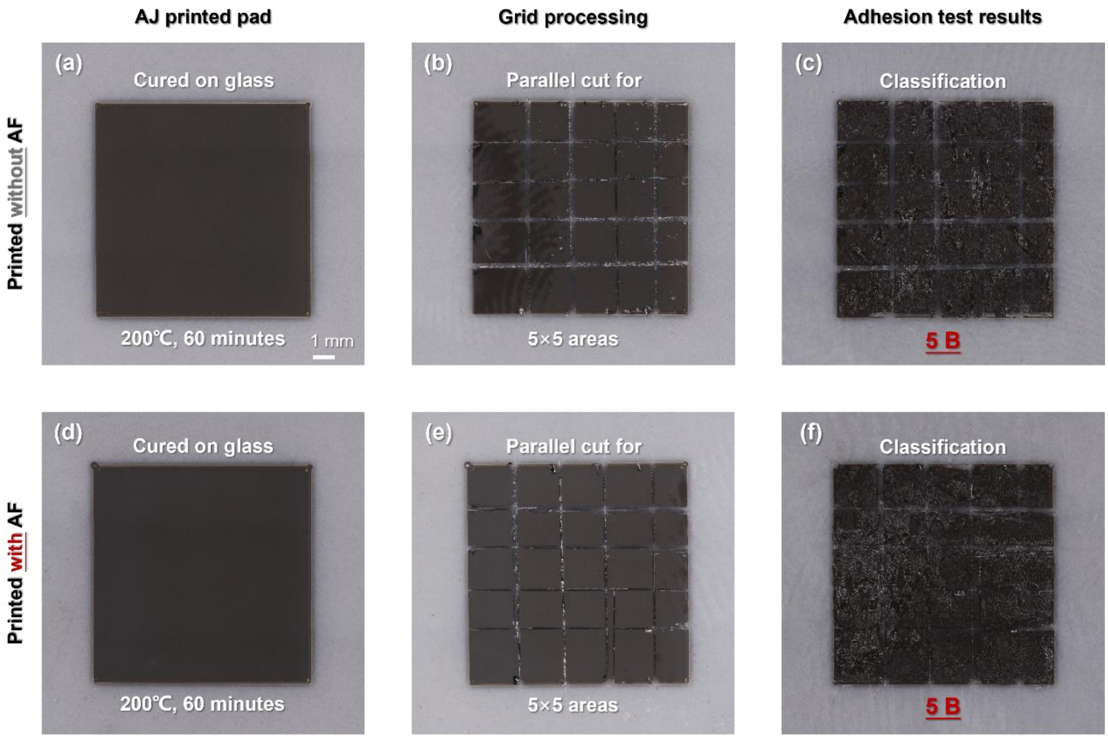


***Fig. S12 a, d** AJ printed pads cured on glass at 200 ℃ for 60 minutes without AF (a) / with AF (d). **b, e** 5×5 areas obtained by parallel cutting of the pad printed without AF (b) / with AF (e). Adhesion test results with classification of AJ printed pads without AF (c) / with AF (f)*" (please see page 11 in the revised supplementary information)

Reviewer #2 (Remarks to the Author):

Review for manuscript NCOMMS-23-48632:

Overall, the paper is well written and presented. The figures (especially Fig. 1) are well done and show some work was put into them to follow the standards of Nature Communications. The results being documented are noteworthy both academically and for industry. The paper explores the use of ultrasound radiation forces on particles, annular acoustic focusing (AF), in order to focus the powder stream used in Aerosol jet printing. This was demonstrated to decrease the line width of printed traces by about 60%. At this stage of the review process, I would like to see the following mayor revisions addressed to increase the quality of the manuscript.

Response

We express our gratitude to the reviewer for the affirmative feedback regarding the quality of our manuscript and the diligence applied in presenting our findings. It is gratifying to learn that the results were perceived as both academically significant and pertinent to industry applications. We recognize the necessity for major revisions as suggested to augment the manuscript's quality. Accordingly, we hereby pledge to rigorously address the identified domains necessitating improvement, with a particular focus on refining the articulation of the introduction, methodology, and results. Additionally, we will enhance the description, characterization and analysis of the printed particle specifics, thus guaranteeing the lucidity and comprehensibility of our scholarly work.

Comment #1

(Section 1) In line 33, change the words 'the sharp corner' to 'sharp corners'.

Response

We thank the reviewer for the careful checking, and we have corrected this typo and checked other possible errors throughout the manuscript.

"*The large working distance provides a distinct advantage for printing on surfaces with significant roughness or topographical features, especially along sharp corners over a complex surface.*" (please see page 2 in the revised manuscript)

Comment #2

For the paragraph starting in line 44, include not only what was investigated in previous work, but also what their conclusions were. This is also an opportunity to link previous work to the current work. For example, in reference 31, one can describe how the ratio of sheath gas flow rate to carrier gas flow rate affected the final resolution, and then mention excessive sheath gas flow can only focus particles only up to a certain limit, due to the flow field, turbulence, etc.

Response

We thank the reviewer for the comprehensive review and in-depth exploration of the research status of this article with the detailed comment for refinement. We have fully followed your suggestion to modify the conclusion of the research status, which highlights the significance and key role of this work.

"*The focusing ratio, defined as the ratio of sheath gas flow rate to carrier gas flow rate, was considered as a key parameter to determine the geometry of printed traces*[31]*, which has an optimal value corresponding to the highest resolution obtained, while both the final resolution and its stability is constrained by the unstable turbulent and printing equipment. Das et al. established a detailed and accurate three-dimensional computational fluid dynamics model for AJ printing to reveal the interplay between the monodisperse droplet size and gas flow*[20] *and pointed out that higher print resolution requires a higher sheath gas velocity, however, overspray increases after decreasing to its minimum value with the increasing sheath gas velocity. The fundamental fluid mechanics principles that govern the deposition process in AJ printing were also explored*[32]*, which proposed that those aerosols deposited along the center of the line are distorted much more significantly than the edge aerosols. The ink composition variation and the interaction between drying induced by the sheath gas and impaction also have a dramatic influence on the line quality*[33,34]*, and it is also necessary to consider whether they meet the physicochemical conditions of AJ printing when formulating inks to improve printing quality and conductivity.*" (please see page 2 in the revised manuscript)

Comment #3

Reword paragraph starting at line 60 to link it better to the article; wasn't AF presented previously? Maybe move the sentence starting with 'One promising approach to micro-manipulate particles of different sizes in a fluid is AF' to an earlier part of the article?

Response

We thank the reviewer for the thorough consideration of the explanation of the research status. With your rigorous and meticulous editing suggestions, the logic and coherence of the research status have been greatly improved.

"*Due to excellent directionality*[37,38]*, strong penetration*[39,40]*, high sensitivity*[41,42]*, and stability*[43]*, acoustic focusing (AF) is another promising approach to kinds of micro-manipulate particles in a fluid, which has been extensively applied in many particles focusing research works as follows.*" (please see page 3 in the revised manuscript)

Comment #4

In line 65, it is not clear what 'three-dimensional flow form' means. Does this mean 'the shape of the 3D flow field'?

Response

We thank the reviewer for reviewing the article carefully and asking the detailed question. We apologize for not clearly explaining this key technical requirement and will provide the more-clear description in the article.

"*Nonetheless, for aerosolized particles (APS) with smaller particle sizes (20~50 nm), faster jet speed (30~80 m/s), and the shape of the 3D flow field*[17] *as well as the requirement of radial focusing from all angles, this is still a challenge.*"(please page 3 in the revised manuscript)

Comment #5

In line 71, briefly mention sound will be used to focus the particles, hence the annular piezoelectric tube is used.

Response

We thank the reviewer for the comprehensive review and detailed revision suggestion for the article. We have accepted this comment and modified the manuscript to ensure the completeness and coherence of the article.

"*A new print nozzle geometry with two tapered focusing zones was proposed to fix the annular piezoelectric tube, in order to form an annular acoustic field radially focusing the APS from all angles.*" (please see page 4 in the revised manuscript)

Comment #6

The introduction section needs a lot of work to increase its comprehensiveness, in particular tying the current work in the article with the following fields: other fields that have used ultrasound for sorting particles and/or focusing a powder stream, other ways particles have been focused besides acoustic radiation forces, and work done within additive manufacturing to focus a powder stream using ultrasound. To address these gaps, the authors should consider incorporating the following recent research:

Cicek, Ahmet, et al. "Acoustophoretic separation of airborne millimeter-size particles by a Fresnel lens." Scientific reports 7.1 (2017): 43374.

Choi, Hoseop, et al. "Controlled electrostatic focusing of charged aerosol nanoparticles via an electrified mask." Journal of Aerosol Science 88 (2015): 90-97.

Pan, Yong-Le, et al. "Opto-aerodynamic focusing of aerosol particles." Aerosol Science and Technology

52.1 (2018): 13-18.

Huang, Yuze, Mir Behrad Khamesee, and Ehsan Toyserkani. "Electrodynamic concentration of non-ferrous metallic particles in the moving gas-powder stream: Mathematical modeling and analysis." International Journal of Magnetics and Electromagnetism 5 (2019): 019.

Smith, Peter H., et al. "Magnetic manipulation in directed energy deposition using a programmable solenoid." Journal of Materials Processing Technology 299 (2022): 117342.

Martinez-Marchese, A., et al. "On the application of sound radiation force for focusing of powder stream in directed energy deposition." Ultrasonics 127 (2023): 106830.

Martinez-Marchese, A., et al. " Ultrasound particle lensing: Powder stream control in directed energy deposition using acoustic radiation force fields." Additive Manufacturing 73 (2023): 103698.

Response

We express our gratitude for the reviewer's thorough examination of the research status and the detailed, extensive recommendations provided for advancing acoustic particle manipulation studies. Our sincere appreciation extends to the reviewer for dedicating considerable effort to compile an extensive list of references for our examination and consideration. Upon meticulous review of the cited literature, we have identified and incorporated pertinent findings as follows that significantly augment the research status of our article. This incorporation not only amplifies the relevance of our study but also accentuates its innovative aspects.

"*Cicek et al. numerically proposed an acoustophoretic separation method of spherical solid particles in the air by means of an acoustic Fresnel lens*[50]*. Choi et al. demonstrated controlled printing of charged nanoparticles using a metal-coated stencil mask by applying an electric potential to engineer the electric field streamlines through the mask opening*[51]*. Pan et al. focused a stream of moving micron-sized aerosol particles in the air by the combined drag force and optical force that is generated by a double-layer co-axial nozzle and a focused doughnut-shaped hollow laser beam, respectively*[52]*. Huang et al. presented an electrodynamic concentration approach for submillimeter-sized conductive metal particles focusing on moving gas-powder streams to generate a tightly focused powder stream in powder-fed additive manufacturing*[53]*. Smith et al. demonstrated a methodology allowing magnetic fields to dynamically manipulate processing conditions as an adaptive control technique in cases where greater control of track dimensions or enhanced material feed control is required*[54]*. Martinez-Marchese et al. used an ultrasound phased array to produce a small volume of high-intensity ultrasound to acoustically focus and deflect particle streams exiting from a nozzle analog before it reaches the build plate with an increased catchment efficiency*[55,56]." (please see page 3 in the revised manuscript)

"*50. Cicek, A., Korozlu, N., Adem Kaya, O. & Ulug, B. Acoustophoretic separation of airborne millimeter-size particles by a Fresnel lens. Sci. Rep.* ***7****, 43374 (2017).*

51. Choi, H. et al. Controlled electrostatic focusing of charged aerosol nanoparticles via an electrified mask. J. Aerosol Sci. ***88****, 90–97 (2015).*

52. Pan, Y.-L., Kalume, A., Wang, C. & Santarpia, J. L. Opto-aerodynamic focusing of aerosol particles. Aerosol Sci. Technol. ***52****, 13–18 (2018).*

53. Huang, Y., Khamesee, M. B. & Toyserkani, E. Electrodynamic Concentration of Non-ferrous Metallic Particles in the Moving Gas-powder Stream: Mathematical Modeling and Analysis. Int. J. Magn. Electromagn. ***5****, 1–12 (2019).*

54. Smith, P. H., Murray, J. W., Jackson-Crisp, A., Segal, J. & Clare, A. T. Magnetic manipulation in directed energy deposition using a programmable solenoid. J. Mater. Process. Technol. ***299****, 117342*

(2022).
55. Martinez-Marchese, A., Ansari, M., Wang, M., Marzo, A. & Toyserkani, E. On the application of sound radiation force for focusing of powder stream in directed energy deposition. Ultrasonics ***127****, 106830 (2023).*
56. Martinez-Marchese, A. et al. Ultrasound particle lensing: Powder stream control in directed energy deposition using acoustic radiation force fields. Addit. Manuf. ***73****, 103698 (2023).*" (please see page 26 in the revised manuscript)

Comment #7

(Section 2) In line 91, clarify or correct the statement 'the ink undergoes turbulent dynamics, giving rise to Stokes drag force'. Does turbulence cause Stokes drag? Expand on the effect on turbulence and particle size on the powder stream characteristics; one thing to look for in the literature that is used to quantify this is the Stokes number. Is this drag referring to the particle's acceleration mechanism? If so, mention it.

Response

We thank the reviewer for the in-depth review of the motion mechanism of aerosol ink particles. Here, we correct and elaborate on this statement as follows.

"*The APS, propelled by the carrier/sheath gas, undergo laminar flow dynamics[20] characterized by a notable Stokes number[18]. This leads to the dominance of Stokes drag[27], as the primary acceleration mechanism for the particle, propelling the APS through the nozzle as depicted in Fig. 1b.*" (please see page 4 in the revised manuscript)

Comment #8

In line 125, what is it meant by 'forms the propagation of acoustic waves'? Instead of this statement, one could use 'produces the acoustic waves'.

Response

We are grateful for the reviewer's thorough review of the theoretical expression in the article. We have now made modifications to it according to your suggestions and carefully checked for any other potentially unreasonable expressions.

"The boundary input acoustic pressure $\boldsymbol{p}_t$ produces the acoustic waves in the acoustic medium..." (please page 6 in the revised manuscript)

Comment #9

Include references for Eqs. 1, 2, 4 and 5. Unless the equations are simple and self-evident such as Eqs. 7 and 8, go over the article to make sure the equations being used are either derived or properly referenced.

Response

We are grateful for the reviewer's thorough examination and rigorous suggestions on the equations in the text. We have added references to all the equations mentioned in this comment. In addition, we have gone over the article to make sure the equations being used are either derived or properly referenced.

"*The coupling equation of the acoustic-structure boundary[57] is*

$$\boldsymbol{n} \cdot \left(\frac{\nabla p_t}{\rho_0}\right) = \boldsymbol{n} \cdot \boldsymbol{a} \tag{1}$$

……*The acoustic pressure distribution in the entire acoustic space is obtained from the Helmholtz equation*[57], *and the simplified equation is*

$$\nabla^2 \boldsymbol{p} = -{k_{eq}}^2 \boldsymbol{p} \tag{2}$$

……*For the thermally conducting and viscous model*[50],

$${k_{eq}}^2 = \frac{\rho\omega^2}{\rho c^2 + i\omega\left[\frac{4}{3}\mu + \mu_B + (\gamma-1)\frac{k_{th}}{c_p}\right]} \tag{4}$$

……*The acoustic radiation force* $\boldsymbol{F}_{rad}$ *exerted on an individual particle within an acoustic field*[58] *can be expressed as*

$$\boldsymbol{F}_{rad} = -\pi r \nabla \left\{ \frac{2\kappa_f}{3} Re\left[\left(1 - \frac{\kappa_p}{\kappa_f}\right) \boldsymbol{p}\nabla\boldsymbol{p}\right] - \rho_f Re\left[\left(\frac{2(\rho_p-\rho_f)}{2\rho_p+\rho_f}\right) \boldsymbol{c}\nabla\boldsymbol{c}\right]\right\} \tag{5}$$

” (please see page 6 in the revised manuscript)

“*50. Cicek, A., Korozlu, N., Adem Kaya, O. & Ulug, B. Acoustophoretic separation of airborne millimeter-size particles by a Fresnel lens. Sci. Rep. 7, 43374 (2017).*

57. Fan, X. Acoustic radiation forces on spherical objects in a viscous fluid by Bessel beams. J. Appl. Phys. ***134****, 134701 (2023).*

58. Muller, P. B., Barnkob, R., Jensen, M. J. H. & Bruus, H. A numerical study of microparticle acoustophoresis driven by acoustic radiation forces and streaming-induced drag forces. Lab. Chip ***12****, 4617 (2012).*” (please see page 26 in the revised manuscript)

Comment #10

In line 188, ‘30 m/s from the published result’, what published result? In this article? If so, use ‘reported result’.

Response

We appreciate the reviewer for the careful review of the wording in our article. We apologize for any inaccuracies in our choice of words and have made revisions according to this suggestion.

“*…30 m/s from the reported result*[24]*…*” (please see page 9 in the manuscript)

Comment #11

In line 204, how is the over-spray determined? Please expand on what is meant by over-spray in the article.

Response

We thank the reviewer for the perceptive observations concerning the determination of overspray in our research. Your recommendation to expand on what is meant by over-spray in the article to substantiate our assertions is highly valued and it will substantially improve the clarity and rigor of our work. First, we have defined the LW in this work and especially the overspray, both using the concept of average line width, and the details are as follows. What’s more, we have used this method to comprehensively quantify and analyze the over-spraying of all printing results in this work.

“*Both the LW and the overspray in this work were defined through the utilization of the average LW. Along the printing direction, the maximum deposition width* L_{max_i} *was extracted on each pixel column in orthogonal directions. Next, the discontinuous pixel units along the printing direction were removed and a continuous deposition width* L_i *along the printing direction was obtained. Finally, the LW of the AJ printed traces was denoted as* $LW = \frac{1}{n}\sum L_i$. *The maximum printing range was defined as* $PW =$

$\frac{1}{n}\sum L_{max_i}$. *The width of overspray was denoted as* $OW = \frac{1}{2}(PW - LW)$." (please see page 22 in the revised manuscript)

Comment #12

(Section 3) Regarding the model and COMSOL simulation, one thing that is not specified in a lot of the published literature is the boundary pressure amplitude used (in Pa or dB or both). How was this determined? Could the pressure amplitude used be linked back to the piezoelectric tube manufacturing specifications or other measurements?

Response

We thank the reviewer's attention to and in-depth inquiries about the simulation design. In response, we provide the following answers. Piezoelectric tube specs detail electric field strength, piezoelectric coefficients, and mechanical properties, which aid in estimating acoustic pressure upon voltage application. Stress-pressure relationships are derived using piezoelectric equations and constants. Pressure amplitude is theoretically modeled based on electric potential, material properties, and geometry through wave equation solutions. COMSOL simulations use a piezoelectric module and material parameters with voltage to calculate boundary pressure amplitude automatically.

Comment #13

Include more information on the Lagrangian particle tracking used in the simulation; how many particles where used? How are they initially distributed in position and velocity? What was the integration time step? Drag models, etc.? How was the width decrease determined from the final particle positions? This is essential to reproduce the simulation results.

Response

We thank the reviewer for the attention and inquiries about the particulars of the particle tracing simulation. Its details are supplemented as follows.

"*Next, a particle beam is placed in the ink domain, and the acoustic radiation force (only set in AF-assisted AJ printing simulation) and drag force are applied to it for transient study. The quantity of particles emitted at the nozzle inlet is 100, positioned initially in accordance with a density distribution of order 5 at the inlet. The initial velocity aligns with the gas velocity distribution at the inlet based on the gas flow rate. Transient investigations were performed utilizing a time step set at 0.01 ms. Subsequently, the printed LW was defined as twice the maximum distance covered by the particles from the base surface to the model axis. The simulated focusing ratio is derived from the LW of the simulation with acoustic radiation force applied relative to one without such force.*" (please see page 22 in the revised manuscript)

Comment #14

(General remarks) One thing that would greatly improve this article would be the addition of summed images from a high speed camera or even from a single photograph of the powder stream in order to know the particle density distribution (PCD) in the powder stream. This has been done numerous times in the additive manufacturing literature, for example in:

Zeng, Minxiang, et al. "High-throughput printing of combinatorial materials from aerosols." Nature

617.7960 (2023): 292-298.

Ansari, M., et al. "A mathematical model of laser directed energy deposition for process mapping and geometry prediction of Ti-5553 single-tracks." Materialia 12 (2020): 100710.

The rationale for why a photograph indicates the PCD can be found in:

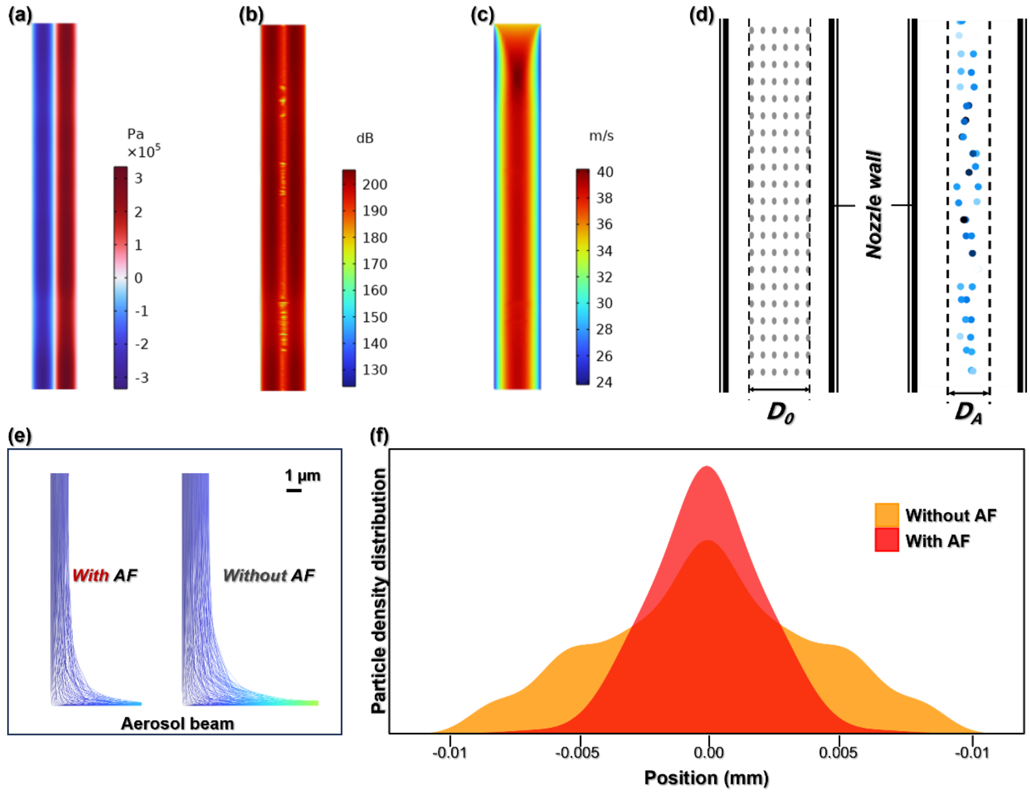
Pinkerton, Andrew J., and Lin Li. "Modelling powder concentration distribution from a coaxial deposition nozzle for laser-based rapid tooling." J. Manuf. Sci. Eng. 126.1 (2004): 33-41.

If the assumptions used in the above article do not apply, and/or the equipment is not available, another option would be obtaining an estimate from the COMSOL model. This can be done by running the simulation and recording the final particle positions at a common plane normal to the nozzle. Then counting the number of particles (making sure the number is statistically significant, see Monte Carlo Methods by Kalos) at different radial widths from the center of the nozzle and normalizing using the mass flow-rate to give a concentration value.

Response

We thank the reviewer for the attention to the simulation results and the proposed simulation data analysis method. We have processed the particle position information to obtain particle density distribution and conducted the following analysis according to your suggestions.

"*By tracking the trajectory of ink particles in the acoustic-flow coupled physical field model, the spatial arrangement of particle clusters deposited on the substrate is obtained (Fig. S2e). The particle density distribution, as shown in Fig. S2f, was derived from statistical analysis of the position information of all particles. The longitudinal trend of its distribution is closer to the topography profile of actual printed lines. The distance between the two ends of the distribution can be approximately determined as LW. Remarkably, our calculations reveal that the utilization of simulated AF has resulted in a reduction of LW to approximately 70% of its original magnitude.*" (please see page 8 in the revised manuscript)



"

Fig. S1 *Simulated process of AJ printing with/without AF of the optimal frequency (6.035 MHz).* ***a*** *Acoustic pressure distribution.* ***b*** *Acoustic intensity distribution.* ***c*** *Flow field distribution.* ***d*** *Particle motion distribution in aerosol ink flow before and after acoustic focusing (AF).* ***e*** *Simulated particle trajectories with/without AF.* ***f*** *Particle density distribution of the AJ printed trace with/without AF.*" (please see page 3 in the revised supplementary information)

Comment #15

Another thing that could be commented on is the mass balance of this process; does the track cross-sectional area match with the mass flow-rate from the nozzle? Does the sound affect this by changing over-spray for example?

Response

We thank the reviewer for proposing this effective evaluation method. Based on your suggestions, we have designed the specific implementation of this evaluation method as follows.

(1) If there is no loss of material during the jet, the mass flow rate at the output of the nozzle and the cross-section mass are the same, and the continuity equation applies $\rho_b A_b v_b = \rho_d A_d v_d$, where ρ_b and ρ_d are density of the jetting aerosol beam and the deposition material, respectively. A_b and v_b are the cross-sectional area and jet velocity of the aerosol beam at the nozzle output, respectively. A_d and v_d are the cross-sectional area of the printed trace and the printing speed, respectively. Evaluating this study through the mass balance of the process is completely effective. Based on our research equipment conditions, there is currently no ultrahigh-speed microscope camera, etc. available to obtain the geometry and motion parameters of the jetting aerosol beam, including ρ_b, A_b and v_b.

(2) In the research design of the mass balance mentioned above, it is expected that the cross-section mass of the printed trace will inevitably be less than the actual value due to overspray amount that cannot be measured or insufficiently measured. However, under the condition of reducing overspray by AF, such cross-section mass should be able to better match the mass flow rate from the nozzle.

Though, we believe that there will be an opportunity in the future to achieve this measurement and evaluate a series of studies on AJ printing based on the method you proposed.

Comment #16

The reason for obtaining the PCD above the traces is that it could be used to describe the change in width (as well as the geometry of the transition region shown in Fig. 2) when the sound is being applied. This could be done by considering the track geometry to be related via a convolution with the PCD. This has been done in the context of cold spray additive manufacturing:

Venturi, F., N. Gilfillan, and T. Hussain. "A convolution-based approach to cold spray additive manufacturing." Additive Manufacturing Letters 1 (2021): 100014.

Response

We thank the reviewer for the in-depth attention to the track geometry change through AF process and suggesting a convolution with the PCD to relate it, and we have supplemented it as follows.

"*Moreover, the entire track geometries of the trace printed before, during, and after a short-term AF process, incorporating both two-dimensional and three-dimensional*[60] *measurements, are supplemented in Fig. S6g, h.*" (please see page 11 in the revised manuscript)

"*60. Venturi, F., Gilfillan, N. & Hussain, T. A convolution-based approach to cold spray additive* *manufacturing. Addit. Manuf. Lett.* ***1****, 100014 (2021).*" (please see page 27 in the revised manuscript)

"

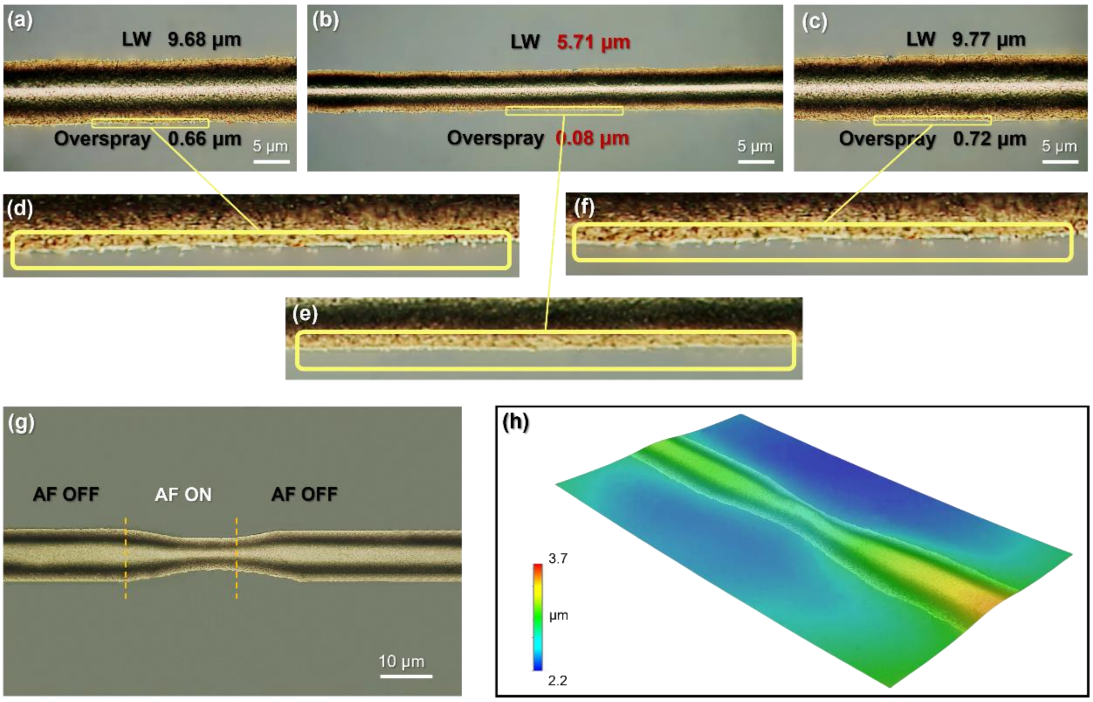


Fig. S6 a *Original geometric morphology of an AJ printed line before AF.* ***b*** *Geometric morphology of the line printed continuously under AF ON.* ***c*** *Geometric morphology of the line printed continuously with (b) after turning off AF.* ***d, e, f*** *Enlarged images of the edge morphology of the AJ printed line without/with/without AF of (a), (b), (c) respectively.* ***g*** *2D and* ***h*** *3D whole track geometry of the AJ printed trace before, during and after a short-term AF.*" (please see page 6 in the revised supplementary information)

Comment #17

(Videos) For the videos, instead of using the phrase 'Acoustic ON/OFF', the phrase 'Acoustic Focusing ON/OFF' could be used.

Response

We thank the reviewer's comments on the expression of the article. In the demonstrative video showcasing the continuous AJ printing with alternating activation and deactivation of AF, the terminology "*Acoustic Focusing ON/OFF*" has been employed rather than the simpler "*Acoustic ON/OFF*" (please see the revised Supplementary Movie 1).

Reviewer #3 (Remarks to the Author):

The authors demonstrate a novel and readily applicable advancement to improve the overall performance of aerosol jet printing technology. This approach is conceptually defined through simulation and calculations, and experimentally demonstrated by analysis of printed traces for line width, overspray, and conductivity. The results presented justify the claims well and the annular acoustic focusing is shown to be a robust and practical method. Although no device applications are shown to further highlight the advantages of the improved printing method, this work is very well done and should be accepted with only minor revisions according to the points made below:

Response

We thank the reviewer for acknowledging the novelty and practicality of our proposed AF strategy in improving AJ printing performance, and the recommendation with only minor revisions. We will ensure that all suggested points are carefully addressed in the revised version, mainly supplementing the method of choosing the optimal acoustic frequency, optical image results of the printed traces at different frequencies and the specific detail on the methods used to manufacture the custom nozzle. Thanks once again for your positive assessment and valuable recommendation to further highlight the advantages of our optimized printing method.

Comment #1

Line 39: "Conversely…" This sentence is worded in a somewhat confusing way. It seems to be saying the presence of overspray reduces the material density of a given trace, but the wording is hard to follow.

Response

We thank the reviewer for the careful examination and consideration of the wording in the article. We have rewritten the wording here, and carefully considered any other potentially confusing inappropriate wording.

"*Concurrently, the presence of overspray reduces the density of a given trace, leading to the actual effective deposition being insufficient compared to the intended amount*[22]." (please see page 2 in the revised manuscript)

Comment #2

In Figure 1g, Stokes drag force is shown to be pointing in the same direction as the particle is being jetted but drag force should actually point in the opposite direction from particle motion. Also "flow filed" seems to be a type for "flow field".

Response

We thank the reviewer for the careful review of the article and apologize for the lack of clarity in my description of the above two points, which may have caused inconvenience to your understanding.

(1) In AJ printing process, the flowing gas generates Stokes drag force on the ink particles that need to be jetted, which is a drag force for jetted particles to maintain inertia and a driving force for their jet motion in the direction of gas flow. Therefore, the direction of Stokes drag force is consistent with the direction of particles.

(2) We have corrected the typo in Fig. 1g. (please see page 5 in the revised manuscript)

"

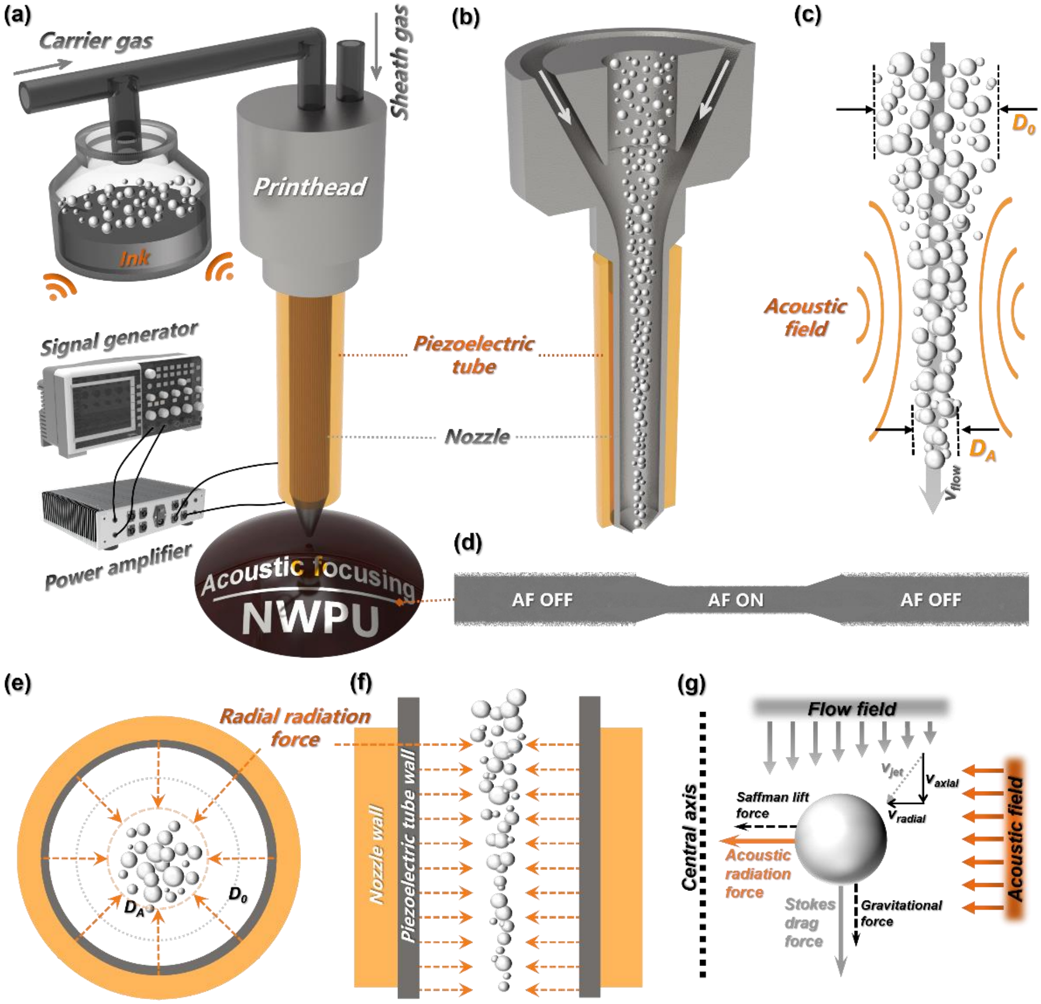


Fig. 1 Overall concept, design, and effects. ……" (please see page 5 in the revised manuscript)

<u>Comment #3</u>

Line 181: "tapper" seems to be a typo.

<u>Response</u>

We thank the reviewer for the careful checking, and we have corrected this typo and checked the possible errors throughout the manuscript.

"*To realize the annular acoustic field assisted AJ printing, the primary task is integrating the circular piezoelectric tube into the printing nozzle which is normally fabricated with a taper cylindrical geometry and maintaining the printing capability as well.*" (please see page 8 in the revised manuscript)

<u>Comment #4</u>

Line 198: It would be good to share the optical image results of the printed traces at frequencies from 5 – 7 MHz so it can be understood why 5.8 MHz was chosen as optimal. This could also provide some insight into what printed traces look like when there are undesired pressure nodes present.

<u>Response</u>

We are grateful for the reviewer's deep concern and inquiry about the results of AF-assisted AJ printing under different frequency. We apologize for not providing this complete experimental result for reviewers in the initial draft. Now, in response to this comment, we provide optical image results of all printed traces within the frequency range of 5-7 MHz with a step size of 0.2 MHz, and perform quantitative comparison of their LWs, enabling a comprehensive and sufficient explanation of the optimal acoustic frequency suitable for AF-assisted AJ printing.

"*The morphology variations of the AJ printed traces were recorded by an optical camera, as shown in Fig. S5, which enabled us to identify the optimal focusing frequency. By measuring and comparing their LWs, it can be seen that the LW under frequency changes does not vary monotonically, and the instantaneous values of LW at some single frequencies clearly cannot be stably maintained. This phenomenon can be associated with the simulated outcomes of acoustic field distribution established under different frequency inputs (Fig. S1b) and explained. When the acoustic pressure nodes are not at the neutral axis position, ink particles are not sufficiently focused to the central axis, so the ink flow is not constrained to be dense enough, resulting in a larger LW. When the distribution of pressure node is erratic, ink particles are driven to move in chaotic directions, causing noticeable unstable LW variations of the printed traces. Thereby, the optimal focusing frequency of Ag nanoparticle ink employed in this study has been determined to be 5.8 MHz.*" (please see page 9 in the revised manuscript)

"***5 Optimal AF frequency search experiment***

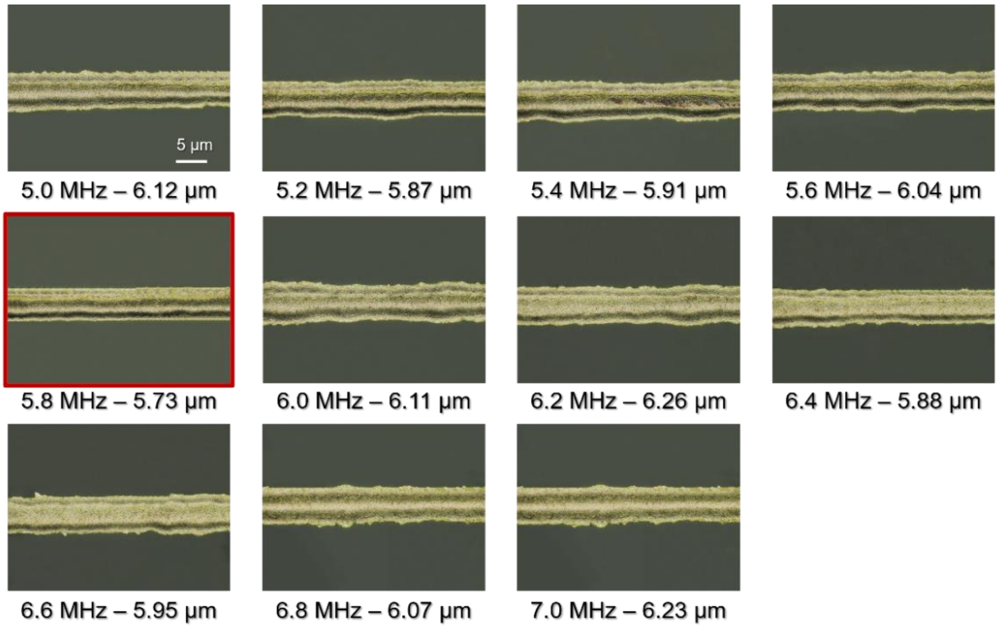


Fig. S5 *AF-assisted AJ printed trace images with a step size of 0.2 MHz in the acoustic frequency range of 5-7 MHz.*" (please see page 5 in the revised supplementary information)

Comment #5

Line 199: It is briefly mentioned that the optimal frequency of 5.8 MHz would be almost identical regardless of ink, but this isn't fully substantiated. Is this because of the nozzle geometry or are the material properties not as significant in determining the optimal frequency?

Response

We are grateful for the reviewer's reviewing and questioning the universality of the optimal frequency.

We apologize for not providing a thorough explanation when mentioning this claim, and supplement the statement as follows.

"*Based on that the acoustic frequency is directly determined by the wavelength, and the design of the wavelength is based on the nozzle geometry, which will be explained as follows, the optimal focusing frequency is uniquely determined by the nozzle geometry. For the purpose of focusing the particles, it is required that the pressure node of the acoustic wave be set at this axis position. There exists an optimal wavelength that satisfies this distribution, which enables the entire acoustic field to form an optimal acoustic pressure distribution and achieve an optimal focusing effect for printing ink particles. This design is not about the ink. For different inks printed using a nozzle of the type in this study, their optimal focusing frequencies are consistent, however, due to differences in ink particle properties, there may be variations in achievable focusing degree at this optimum frequency. What's more, although the nozzle geometry design in this study has achieved comparable printing results compared with ones using ordinary nozzle, there is still room for optimization in its geometry and corresponding printing effects. As a result, the corresponding optimum frequency will undergo corresponding changes accordingly when using a possible more excellent nozzle geometry.*" (please see page 10 in the revised manuscript)

Comment #6

In Figure 5a, sample A and sample B should have their own colors that are consistent between the thickness measurements and line width measurements.

Response

We thank the reviewer for the comments on the presentation of the article, and we have revised the description of Fig. 5a. In addition, we have supplemented the printing stability experiment and data characterization at the two limiting resolutions involving ~6 μm and ~10 μm) here.

"*To assess AJ printing stability with AF, four printing stability tests were conducted under ambient conditions, which involved printing lines with the resolution of 6, 10, 20, and 50 μm respectively. Among these, the stability of 6 μm resolution lasted for approximately 4.5 hours. This duration was notably shorter compared to the other three resolutions with 10 μm, 20 μm, and 50 μm, which all maintained stability for almost 8 hours. The discrepancy can be attributed to the instability observed in the original optimal resolution of 10 μm without AF, but still has greatly improved stability under ultrahigh printing resolution. An optical image was captured every 20 minutes as shown in Supplementary Fig. S9, the LW, and thickness of the trace at various time points can be viewed in Fig. 5a, b. Although a small variation of LW was observed during the long-term printing, the average LW (6.0±0.3 μm, 10.8±1.2 μm, 20.6±1.4 μm, 50.7±1.4 μm) of the AF-assisted AJ printed traces indicates that the excellent printing stability compared to traditional AJ printing method. Compared to the stability shown by the LW approach, there was a significant fluctuation in trace thickness which suggests that the deposition rate varies during printing, a common occurrence in practical applications. Specifically, there is a variation in deposition volume with consistent width but noticeable difference in height. However, this highlights the optimizing impact of AF in AJ printing to maintain the quality and stability of printed LW despite fluctuations in deposition rate. ……*

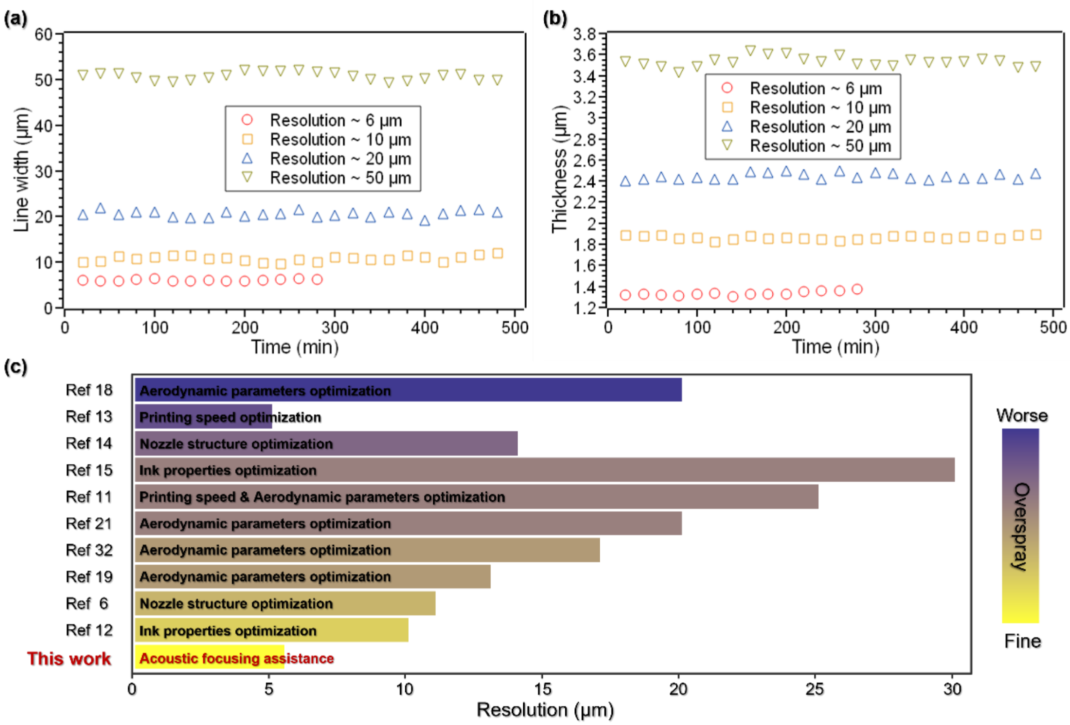


Fig. 5 Stability quantification and performance comparison of AF-assisted AJ printing. *For 8-hour continuous AJ printing with resolution ~6 μm, ~10 μm, ~20 μm, and ~50 μm,* ***a*** *LWs and* ***b*** *thickness were recorded at intervals of 20 mins.* ***c*** *Comparison of different optimization methods on printing resolution and overspray of AJ printing with other reported results.*" (please see page 18 in the revised manuscript)

Comment #7

Line 356: "acoustic acoustic-focused" seems to be a typo.

Response

We thank the reviewer for the careful checking, and we have corrected this typo and checked the possible errors throughout the manuscript."

"*Meanwhile, the conductivity of the acoustic-focused Ag ink stream has an increase to 180±5% compared to that without AF due to the more densely packed nanoparticles.*" (please see page 20 in the revised manuscript)

Comment #8

More detail on the methods used to manufacture the custom nozzle should be provided given the uniquity of a custom-made aerosol jet nozzle.

Response

We appreciate the reviewer's in-depth inquiries about our work. The newly designed nozzle for AF strategy is indeed an important component of this project, and we apologize for not fully explaining this part of the work in a comprehensive manner.

"*Based on the conical design of the original nozzle for pneumatic focusing, combined with the standard tubular geometry of the piezoelectric tube for AF and considering its assembly stability, a standard tubular channel has been integrated into the conical nozzle's structure for the AF mechanism in the AJ* *system, in order to ensure consistent radial transfer of acoustic radiation force. The modified nozzle structure is depicted in Fig. S3a. Its inlet maintains the original structure and parameters, connected to the printer. There are also five parameters to constrain this structure, defined as upper focusing length (UFL), lower focusing length (LFL), acoustic channel sizes (length and diameter), and outlet diameter, which jointly determine the printing effect of the corresponding nozzle. To ensure the AF quality, the acoustic channel length is calculated to be 30 mm which is determined by the focusing time and velocity of the APS. Here, the velocity is approximately 30 m/s from the reported result*[24] *and the focusing time is established to be 1 ms from the simulated result. The inner and outer diameters of the acoustic channel are determined by theoretical calculations using the available parameters of the piezoelectric tube. The outlet diameter is determined by the minimum aperture achievable through current processes. The other two parameters, namely UFL and LFL, jointly determine the pneumatic focusing printing effect without AF in this structure. A numerical combination method, as shown in Fig. S3a, was used to optimize these two parameters without AF. As shown in Fig. S3d, the investigation resulted in a LW of 10.13 μm and an overspray of 0.84 μm which met the high resolution requirement of the AJ printing manufacturers. Therefore, the nozzle of this type can be selected for AF-assisted AJ printing, as shown in Fig. S3b, c, and whose structural parameters are shown in Table. S2.*" (please see page 8 in the revised manuscript)

"***3.1 Device design and nozzle manufacturing***

……

The nozzles were manufactured by laser cladding deposition 3D printing (Sonic Mini 8K, Phrozen), as shown in Fig. S3b, c. Then, the nozzles underwent a meticulous cleaning regimen involving both acetone and isopropanol within an ultrasonic bath. Subsequently, compressed air was employed to meticulously expunge any contaminants and excess resin from the intricate internal configurations of the nozzles. This rigorous purification process is critical to maintaining the integrity of the internal structure, thereby precluding any potential functional discrepancies attributable to structural imperfections in the nozzles."
(please see page 21 in revised manuscript)

"*For the printed nozzles, we used red wax (Photosensitive resin 121, Funcrecol), selected for its superior rigidity and minimal deformation characteristics (3%), which contributed to exceptional mold precision and negligible discrepancies.*" (please see page 21 in the revised manuscript)

"***3 New nozzle design***

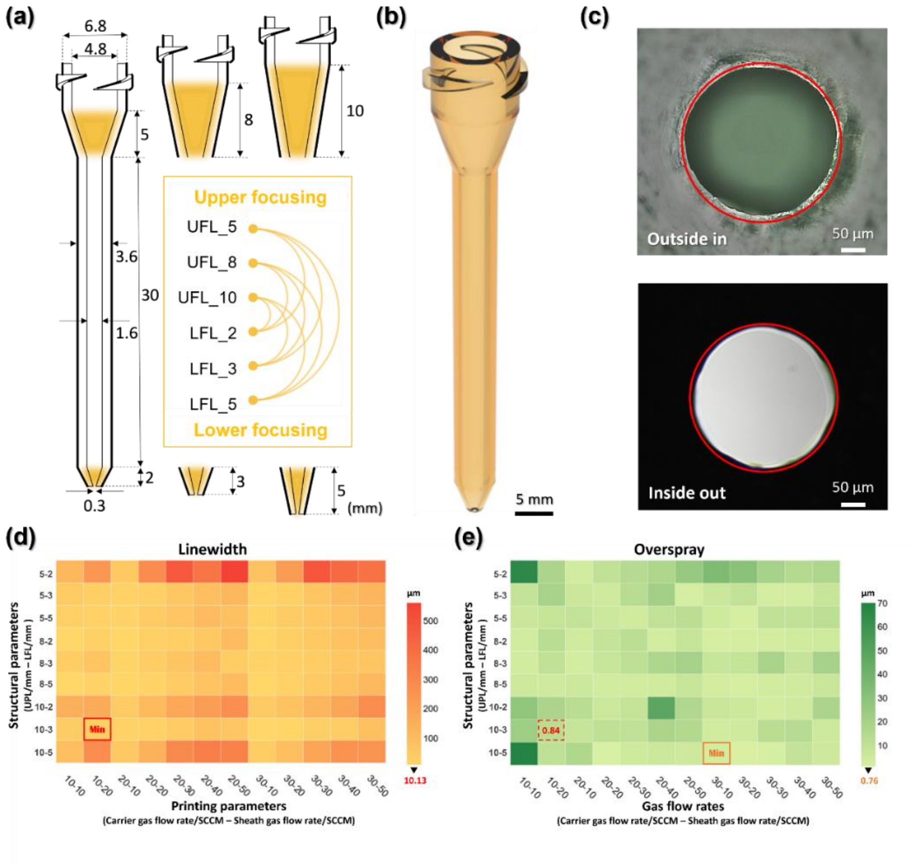


***Fig. S2 a** Geometric structure and design parameters of the nozzle. **b** Optical image of the nozzle. **c** Outlet of the stereolithography printed nozzle. **d, e** Quantitative comparisons of the printed LW (d) and overspray (e) under different printing parameters for nozzles with different structural parameters.*

***Table. S2** Structural parameters of the new nozzle*

Upper focusing length (mm)	*Lower focusing length (mm)*	**Acoustic channel** *inner diameter (mm)*	**Acoustic channel** *outer diameter (mm)*	*Outlet diameter (mm)*	*Inlet diameter (mm)*
10	*3*	*1.3*	*3.5*	*0.3*	*4.8*

” (please see page 4 in the revised supplementary information)

Reviewer #4 (Remarks to the Author):

The concept presented in this paper to use annular acoustic focusing to improve the resolution of aerosol jet printing is novel and of interest to the manufacturing community. I have a number of serious concerns regarding the methodology and results claimed in this study as well as several questions about the modeling. I will also point out that the authors use a number of variables without defining them throughout the document.

Response

We express our gratitude towards the reviewer for the insightful comments regarding the presentation of our article. In response to the reviewer’s feedback, we have undertaken a thorough revision of the article’s expression, ensuring that it now aligns with the highest academic standards. This revision process involved a careful examination of our initial expressions and the incorporation of the reviewer's suggestions, which has significantly enhanced the clarity and coherence of our work.

In section 1, I have the following concerns/questions:

Comment #1

There are a number of ambitious and weighty claims made about existing literature without appropriate citations. In lines 35 & 56, claims of ‘little success’ are made without any definitive, quantitative statement about the current state of the field. Some concrete examples of what has been achieved will greatly add to this discussion.

Response

We thank the reviewer’s in-depth attention and strict suggestions on our claim. In this regard, we acknowledge that the term “little success” was not appropriate, so we have revised the wording and introduced relevant references to supplement the discussion of the research status.

“*However, it remains a challenge when it comes to fabricating a pattern with multi-ultrafine lines and extremely small line gaps*.” (please see page 2 in the revised manuscript)

“*Despite the advancements in AJ printing technology, the micro-electronic devices produced exhibit suboptimal electrical performance due to the inferior geometric characteristics of the printed lines*[35]*. Specifically, the low controllability of the printed line width may induce overlap of narrowly spaced circuits or unnecessary intertrack voids, which will hinder the wide application of AJ printing in the advanced electronic manufacturing industry*[36]*. Therefore, ensuring high precision and low overspray for high-density is essential for AJ printing, which urgently needs an effective, real-time, stable, and low-constrained optimization approach.*” (please see page 3 in the revised manuscript)

Comment #2

How significant must overspray be before seeing the issues mentioned in lines 41-43? Overspray is typically a small portion of the printed volume and should not have a large effect on conductivity.

Response

We thank the reviewer for the in-depth inquiry about the statement of overspray’s impact on conductivity. An additional explanation on its impact on electrical performance and attached relevant references are supplemented as follows.

“*In addition, overspray is critical to the electrical properties of a printed device, as an excess of overspray may lead to short circuits and increased resistance*[21].” (please see page 2 in the revised manuscript)

Comment #3

The first two paragraphs of the introduction should be combined and reorganized.

Response

We thank the reviewer’s in-depth attention to and suggestions for revising the introduction. According this comment, we have combined and reorganized the first two paragraphs of the introduction.

“*Aerosol jet (AJ) printing, as an emerging non-contact additive manufacturing technique, is capable of* *fabricating various conformal electronics including passive components[1–5], sensors[6–10], and high-performance interconnects[11–15] over different substrates. Compared to other direct ink-writing printing methods[16], such as syringe/extrusion and ink-jet printing, AJ printing offers several key advantages. For example, the aerosolized ink stream enables a nozzle-to-build plate standoff distance of 2-5 mm and print ink stream widths having diameters as small as 10-20 μm[17]. The large working distance provides a distinct advantage for printing on surfaces with significant roughness or topographical features, especially along sharp corners over a complex surface. Most studies[6,12] declare that AJ printing has a resolution down to 10 μm. However, it remains a challenge when it comes to fabricating a pattern with multi-ultrafine lines and extremely small line gaps. Moreover, the heavy overspray and bulged line geometry largely hindered its application in conformal electronics[18], especially under high-frequency operation and confined printing areas[19]. As a consequence, the proximity of the planned deposition parts may result in inadvertent contact due to overspray[20], potentially resulting in short circuits and substantial impairment to their electrical capabilities[19]. In addition, overspray is critical to the electrical properties of a printed device, as an excess of overspray may lead to short circuits and increased resistance[21]. Concurrently, the presence of overspray reduces the density of a given trace, leading to the actual effective deposition being insufficient compared to the intended amount[22]. Furthermore, deficient focus during the deposition procedure poses obstacles to achieving the desirable densification, thereby restricting conductivity and giving rise to insufficient print strength and compromised electrical performance[23]. Many efforts have been made to improve the printing resolution and uniformity, as well as to explore effective ways to reduce overspray issues, mainly involving three aspects, including optimization of operating parameters[21,24–26], nozzle geometry[27–29], and ink properties[30]. The focusing ratio, defined as the ratio of sheath gas flow rate to carrier gas flow rate, was considered as a key parameter to determine the geometry of printed traces[31], which has an optimal value corresponding to the highest resolution obtained, while both the final resolution and its stability is constrained by the unstable turbulent and printing equipment. Das et al. established a detailed and accurate three-dimensional computational fluid dynamics model for AJ printing to reveal the interplay between the monodisperse droplet size and gas flow[20] and pointed out that higher print resolution requires a higher sheath gas velocity, however, overspray increases after decreasing to its minimum value with the increasing sheath gas velocity. The fundamental fluid mechanics principles that govern the deposition process in AJ printing were also explored[32], which proposed that those aerosols deposited along the center of the line are distorted much more significantly than the edge aerosols. The ink composition variation and the interaction between drying induced by the sheath gas and impaction also have a dramatic influence on the line quality[33,34], and it is also necessary to consider whether they meet the physicochemical conditions of AJ printing when formulating inks to improve printing quality and conductivity. The above multi-directional research has effectively explored the key factors of AJ printing and their influence on the output, providing sufficient theoretical guidance for optimization. Despite the advancements in AJ printing technology, the micro-electronic devices produced exhibit suboptimal electrical performance due to the inferior geometric characteristics of the printed lines[35]. Specifically, the low controllability of the printed line width may induce overlap of narrowly spaced circuits or unnecessary intertrack voids, which will hinder the wide application of AJ printing in the advanced electronic manufacturing industry[36]. Therefore, ensuring high precision and low overspray for high-density is essential for AJ printing, which urgently needs an effective, real-time, stable, and low-constrained optimization approach.*" (please see page 1 in the revised manuscript)

In section 2.1 (and relevant methods section), I have the following concerns/questions:

Comment #4

The claim made in line 94-96 about the reason for ink flow diameter blames 'environmental disturbances'. Given that this is determined in the nozzle itself, what disturbances do the authors mean? In addition, 'limited pneumatic focusing' should include an explanation about why there is a limit to pneumatic focusing.

Response

We thank the reviewer for the careful consideration and in-depth questioning regarding the "environmental disturbances" and "limited pneumatic focusing".

(1) For "environmental disturbances", it can be attributed to several factors: (a) Real-time data from the gas flow controller shows fluctuation of the input volume of carrier gas and sheath gas; (b) Impurities accumulate inside the nozzle flow channel due to prolonged and repeated operation, which can make the transported aerosol ink deviated from the expected effect; (c) Some nozzles used in printing have insufficient forming quality. The roundness of their outlets and standardization degree of internal channels may deviate from design specifications, affecting the distribution pattern of ink flow in printing process. These and even more factors of change will collectively affect the ink flow of the jet, ultimately leading to a certain degree of fluctuation in the printed line width. Then we have conducted four continuous experiments to get large sample data, and provided explanations on the statement of LW fluctuation."

"*The initial diameter of the ink flow, denoted as D_0, was predominantly influenced by factors such as limited pneumatic focusing*[20,31] *and environmental disturbances, which are specifically stated later.*" (please see page 5 in the revised manuscript)

"*It is noted that the slight disparity observed in trace morphology before and after the AF process can be ascribed to the gas flow fluctuation and drift in the ink composition over time which is common issues during AJ printing, mainly because of (a) fluctuation of carrier gas and sheath gas volumes; (b) impurities accumulate inside the nozzle, due to prolonged and repetitive use, leading to dislocation of the aerosol ink during transport; (c) manufacturing defect of the outlet roundness and internal channel standardization degree of nozzles affecting the ink flow distribution.*" (please see page 11 in the revised manuscript)

(2) For "limited pneumatic focusing", the current research results indicate that the optimization of pneumatic focusing has an optimal focus ratio (sheath gas flow rate/carrier gas flow rate). The printing effect of other all focus ratios is worse than that of the optimal focus ratio. However, in the face of current application requirements such as higher resolution, less overspray, higher deposition quality, and higher conductivity, there is still considerable room for optimization in the printing effect of the optimal focus ratio.

"*Das et al. established a detailed and accurate three-dimensional computational fluid dynamics model for AJ printing to reveal the interplay between the monodisperse droplet size and gas flow*[20] *and pointed out that higher print resolution requires a higher sheath gas velocity, however, overspray increases after decreasing to its minimum value with the increasing sheath gas velocity.*" (please see page 2 in the revised manuscript)

"*The initial diameter of the ink flow, denoted as D_0, was predominantly influenced by factors such as limited pneumatic focusing*[20,31] *and environmental disturbances.*" (please see page 5 in the revised manuscript)

"*20. Chen, G., Gu, Y., Tsang, H., Hines, D. R. & Das, S. The Effect of Droplet Sizes on Overspray in Aerosol-Jet Printing. Adv. Eng. Mater.* ***20****, 1701084 (2018).*

31. Dalal, N. et al. Effect of Gas Flow Rates on Quality of Aerosol Jet Printed Traces With Nanoparticle Conducting Ink. J. Electron. Packag. ***142****, 011012 (2020).*" (please see page 25 in the manuscript)

Comment #5

The model appears to be built on previous work studying the acoustic forces on microparticles in a liquid-filled microchannel (the author's reference 48). There is no discussion on the potential applicability of this model in aerosol jet printing, which utilizes nanoparticles suspended in microdroplets (hence aerosol). This is a large assumption which the authors must thoroughly justify in order for the modeling in this work to be plausible.

Response

We thank the reviewer for considering the relevance of this simulation work and the referenced literature. In this regard, we supplemented the following additional explanation.

Ref. 48 is the boundary condition example guide we found from the help documentation of COMSOL software in the process of simulating design boundary conditions, mainly used for reference in acoustic transmission mechanism design. The simulation work of the Ref. 48 was carried out in a water-based creeping flow, where acoustic radiation force was applied to particles originally driven by Stokes drag force, causing their motion to change. The acoustic radiation force gradually became the dominant force instead of drag force. The AJ printing process is very similar to the scenario described in Ref. 48. This similarity can be mainly observed in several aspects: (a) the characteristics of aerosol ink particles are almost equivalent to those described in Ref. 48; (b) gas-based laminar flow can also serve as a driving flow for particle motion with low Stokes number and significant Stokes drag force; (c) as for acoustic radiation force, its transmissibility and driving effect on particles are essentially identical to those in the referred cases. However, this study needs to additionally consider the acoustic transmission mechanism at the nozzle wall and then design the distribution of acoustic field and direction of acoustic radiation force. In general, the simulation is a different design based on the same principle compared to the one Ref. 48.

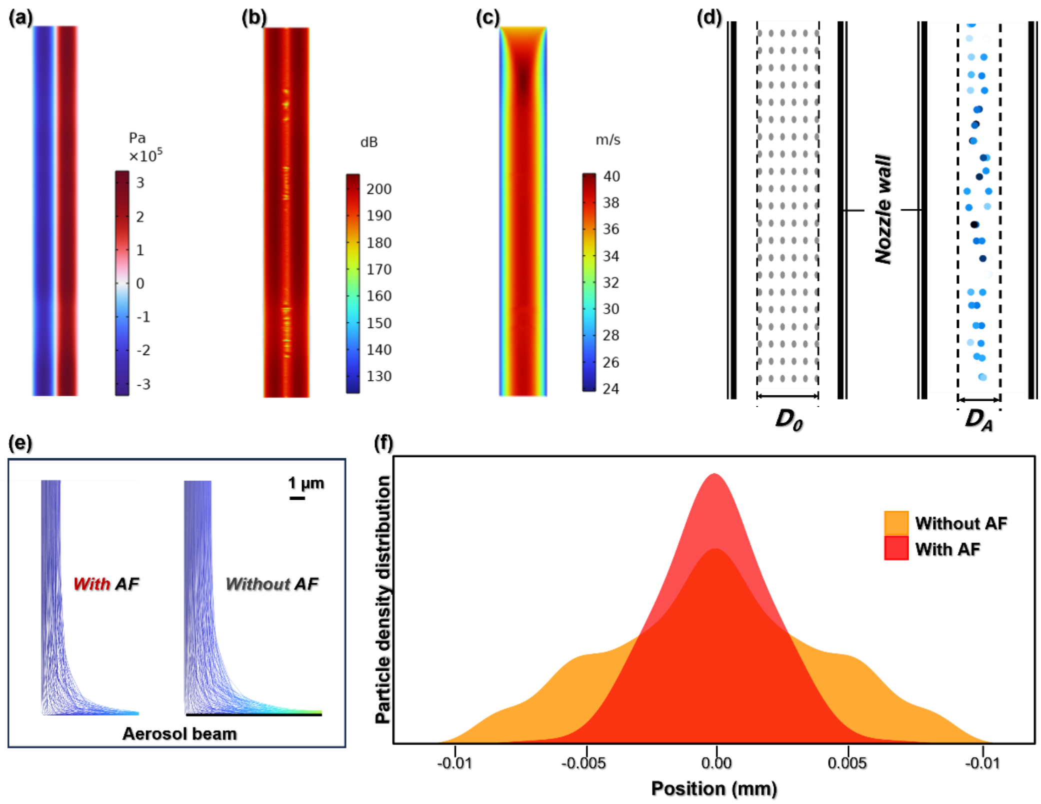
Comment #6

The work described in lines 174-178 needs much more description.

Response

We thank the reviewer for the attention to the simulation work, and we have processed the particle position information to obtain particle density distribution, and conducted the following analysis.

"*By tracking the trajectory of ink particles in the acoustic-flow coupled physical field model, the spatial arrangement of particle clusters deposited on the substrate is obtained (Fig. S2e). The particle density distribution, as shown in Fig. S2f, was derived from statistical analysis of the position information of all particles. The longitudinal trend of its distribution is closer to the topography profile of actual printed lines. The distance between the two ends of the distribution can be approximately determined as LW. Remarkably, our calculations reveal that the utilization of simulated AF has resulted in a reduction of LW to approximately 70% of its original magnitude.*" (please see page 8 in the revised manuscript)



“

***Fig. S2** Simulated process of AJ printing with/without AF of the optimal frequency (6.035 MHz). **a** Acoustic pressure distribution. **b** Acoustic intensity distribution. **c** Flow field distribution. **d** Particle motion distribution in aerosol ink flow before and after AF. **e** Simulated particle trajectories with/without AF. **f** Particle density distribution of the AJ printed trace with/without AF.*” (please see page 3 in the revised supplementary information)

In section 2.2 (and relevant methods section), I have the following concerns/questions:

Comment #7

Given that aerodynamic properties dominate aerosol jet, and a new nozzle is used here, it is imperative that that design is included in the main text. The authors do not even mention the nozzle diameter until much later in the work. In addition, the work shown here should be compared to typical commercially-available nozzles. Without this, it is impossible to evaluate the effectiveness of this new nozzle alone, which from what is here seems to be impressive in its own right.

Response

We appreciate the reviewer's in-depth inquiries about our work. The newly designed nozzle for AF strategy is indeed a so important component of this project that imperative to be included in the main text. We apologize for not having in-deeply explained this part of the work in a comprehensive manner. “*Based on the conical design of the original nozzle for pneumatic focusing, combined with the standard tubular geometry of the piezoelectric tube for AF and considering its assembly stability, a standard tubular channel has been integrated into the conical nozzle's structure for the AF mechanism in the AJ system, in order to ensure consistent radial transfer of acoustic radiation force. The modified nozzle* *structure is depicted in Fig. S3a. Its inlet maintains the original structure and parameters, connected to the printer. There are also five parameters to constrain this structure, defined as upper focusing length (UFL), lower focusing length (LFL), acoustic channel sizes (length and diameter), and outlet diameter, which jointly determine the printing effect of the corresponding nozzle. To ensure the AF quality, the acoustic channel length is calculated to be 30 mm which is determined by the focusing time and velocity of the APS. Here, the velocity is approximately 30 m/s from the reported result*[24] *and the focusing time is established to be 1 ms from the simulated result. The inner and outer diameters of the acoustic channel are determined by theoretical calculations using the available parameters of the piezoelectric tube. The outlet diameter is determined by the minimum aperture achievable through current processes. The other two parameters, namely UFL and LFL, jointly determine the pneumatic focusing printing effect without AF in this structure. A numerical combination method, as shown in Fig. S3a, was used to optimize these two parameters without AF. As shown in Fig. S3d, the investigation resulted in a LW of 10.13 μm and an overspray of 0.84 μm which met the high resolution requirement of the AJ printing manufacturers. Therefore, the nozzle of this type can be selected for AF-assisted AJ printing, as shown in Fig. S3b, c, and whose structural parameters are shown in Table. S2.*" (please see page 8 in the revised manuscript)

"***3.1 Device design and nozzle manufacturing***

……

The nozzles were manufactured by laser cladding deposition 3D printing (Sonic Mini 8K, Phrozen), as shown in Fig. S3b, c. Then, the nozzles underwent a meticulous cleaning regimen involving both acetone and isopropanol within an ultrasonic bath. Subsequently, compressed air was employed to meticulously expunge any contaminants and excess resin from the intricate internal configurations of the nozzles. This rigorous purification process is critical to maintaining the integrity of the internal structure, thereby precluding any potential functional discrepancies attributable to structural imperfections in the nozzles." (please see page 21 in revised manuscript)

"*For the printed nozzles, we used red wax (Photosensitive resin 121, Funcrecol), selected for its superior rigidity and minimal deformation characteristics (3%), which contributed to exceptional mold precision and negligible discrepancies.*" (please see page 21 in the revised manuscript)

"***3 New nozzle design***

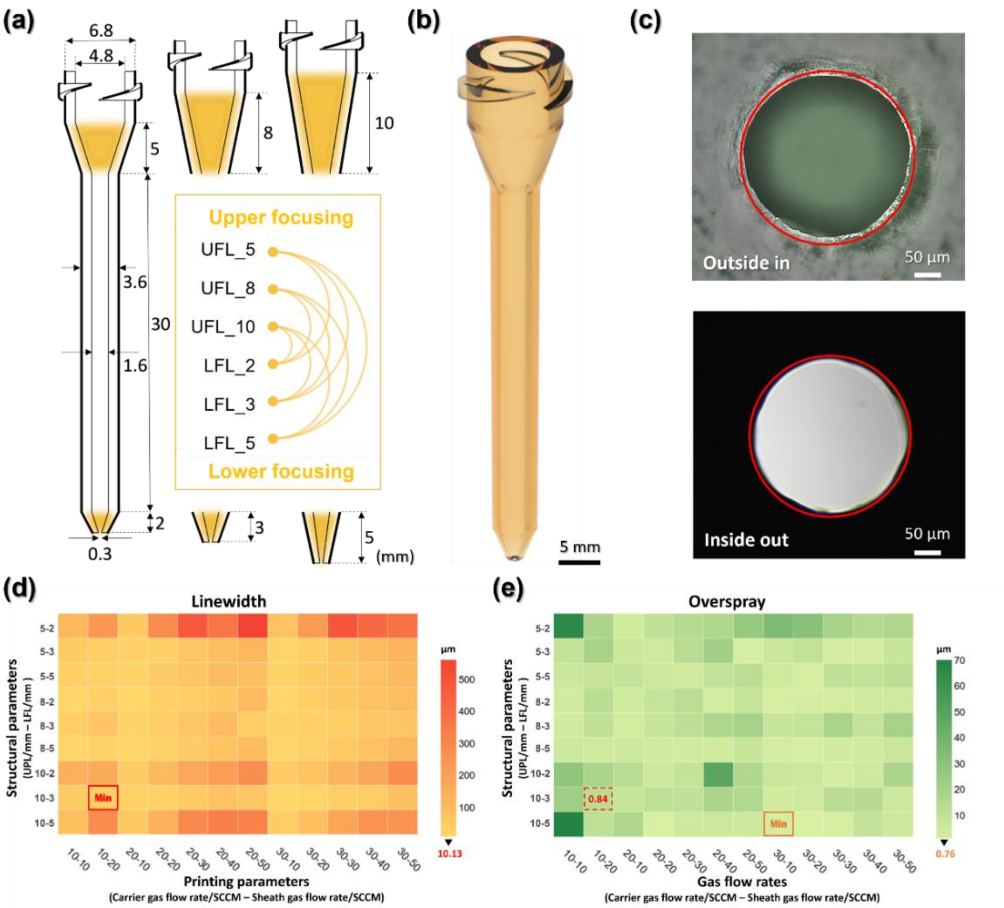


Fig. S3 a *Geometric structure and design parameters of the nozzle.* ***b*** *Optical image of the nozzle.* ***c*** *Outlet of the stereolithography printed nozzle.* ***d, e*** *Quantitative comparisons of the printed LW (d) and overspray (e) under different printing parameters for nozzles with different structural parameters.*

Table. S2 *Structural parameters of the new nozzle*

Upper focusing length (mm)	*Lower focusing length (mm)*	**Acoustic channel** *inner diameter (mm)*	**Acoustic channel** *outer diameter (mm)*	*Outlet diameter (mm)*	*Inlet diameter (mm)*
10	*3*	*1.3*	*3.5*	*0.3*	*4.8*

” (please see page 4 in the revised supplementary information)

Comment #8

This entire section has severely inadequate details about the work, including the details in the later methods section. Details about the piezoelectric actuator, for example, are vital to evaluating and reproducing the outcomes achieved here and must be included.

Response

Thanks very much to the reviewer for asking detailed questions and requesting supplements about the details of the AF assistance device, especially the piezoelectric actuator. This is indeed meaningful for the completeness of the AF strategy and the article. We have supplemented relevant details of the piezoelectric actuator in section 3.1. In addition, we have also added other device details.

"*The piezoelectric actuator adopts a piezoelectric tube (YIJIA, China), with the dimensions of 30 mm × 5.6 mm × 3.6 mm (length × outer diameter × inner diameter), operating frequency range of 5–8 MHz, operating voltage ≤ 1000 V.*" (please see page 21 in the revised manuscript)

Comment #9

There is a mention in line 194-195 that differences between simulations and reality could be the result of material property variation. Nowhere does this work mention what material properties (density, specific heat, viscosities, thermal conductivity, etc.) were used in the simulations.

Response

We are grateful for the reviewer's careful consideration the simulation differences between simulations and reality and meticulous question regarding the material properties used in the simulations. There are indeed certain differences in material properties between the standard materials properties used in simulations and the corresponding ones in the experiment, for the ink, the gas composition, the nozzle, and the piezoelectric tube, and specially the exact material of the piezoelectric tube is not provided completely. We will supplement the relevant material properties used in the simulation as follows.

"*It is known that there are indeed disparities exist between the designated material properties for ink, gas components, nozzles, and piezoelectric tubes in simulation, and their corresponding actual material characteristics in experiments, especially the precise material properties of the piezoelectric tube are not completely understood, thereby resulting in a minor incongruity between the optimal frequencies derived from simulations and those deduced from practical experiments.*" (please see page 10 in the revised manuscript)

"*The material properties used in the simulation are listed in Table. S1.*" (please see page 21 in the revised manuscript)

"

Table. S1 *Material properties used in the simulation*

Domain	***Material properties***		
	Parameter	***Definition***	***Value***
Ink	μ_{ink}	*Viscosity of ink (cP)*	*15*
	ρ_{ink}	*Density of ink (kg/m^3)*	*789.9*
	C_{ink}	*Heat capacity of ink (J/kg/K)*	*1280*
	k_{ink}	*Thermal conductivity of ink (W/m/K)*	*1.9762*
	c_{ink}	*Speed of sound of ink (m/s)*	*1207*
	γ_{ink}	*Ratio of specific heats of ink*	*1.1928*
Gas	μ_{gas}	*Viscosity of gas (cP)*	*0.0183*
	ρ_{gas}	*Density of gas (kg/m^3)*	*1.25*
	C_{gas}	*Heat capacity of gas (J/kg/K)*	*1038*
	k_{gas}	*Thermal conductivity of gas (W/m/K)*	*0.02583*
	c_{gas}	*Speed of sound of gas (m/s)*	*343*
	γ_{gas}	*Ratio of specific heats of gas*	*1.407*
Nozzle	c_{nozzle}	*Speed of sound of nozzle (m/s)*	*2760*
	μ_{nozzle}	*Density of nozzle (kg/m^3)*	*1120*
Piezoelectric tube	μ_{piezo}	*Density of piezoelectric tube (kg/m^3)*	*7750*
	E_c	*Elasticity matrix of piezoelectric tube*	*COMSOL database*

E_{se}	*Coupling matrix of piezoelectric tube*	*COMSOL database*
E_{ε}	*Relative permittivity of piezoelectric tube*	*COMSOL database*

” (please see page 2 in the revised supplementary information)

Comment #10

In lines 195-200, the authors claim to have found an optimum frequency but do not show any data to demonstrate that claim. In addition, the range of frequencies considered for experimentation include frequencies higher than those included in the modeling. Why were higher frequencies not considered in the modeling section?

Response

We are grateful for the reviewer’s deep concern and inquiry about the optimum acoustic frequency of AF-assisted AJ printing. We apologize for not providing the printing effect of AF-assisted AJ printing inputted by optimum and other frequency to demonstrate the claim in the initial draft, however, we did conduct variable parameter experiments on acoustic frequencies within a certain range to obtain the optimal acoustic frequency and its corresponding optimal AJ printing results. In response to this comment, we provide optical image results of all printed traces within the frequency range of 5-7 MHz with a step size of 0.2 MHz and perform quantitative comparison of their LWs. In combination with the simulated acoustic field under different frequency input, we explain below the impact of acoustic frequency on AF-assisted AJ printing, enabling a comprehensive and sufficient explanation of the optimal acoustic frequency suitable for AF-assisted AJ printing.

In addition, for modeling, we started from the perspective of theoretical waveform distribution to obtain the corresponding frequency range, and considering the comprehensiveness of optimization verification in experiments, we expanded the experimental frequency input range. Although this presents the final optimal results, it does not indeed achieve scientific comprehensive validation and comparison in modeling. Therefore, we will supplement a frequency range in the simulation experiment. The size of the supplemented frequency range is consistent with the existing frequency range and is located above the current optimal frequency value to enhance the comprehensiveness and credibility of the optimum frequency value obtained in this article.

“*The morphology variations of the AJ printed traces were recorded by an optical camera, as shown in Fig. S5, which enabled us to identify the optimal focusing frequency. By measuring and comparing their LWs, it can be seen that the LW under frequency changes does not vary monotonically, and the instantaneous values of LW at some single frequencies clearly cannot be stably maintained. This phenomenon can be associated with the simulated outcomes of acoustic field distribution established under different frequency inputs (Fig. S1b) and explained. When the acoustic pressure nodes are not at the neutral axis position, ink particles are not sufficiently focused to the central axis, so the ink flow is not constrained to be dense enough, resulting in a larger LW. When the distribution of pressure node is erratic, ink particles are driven to move in chaotic directions, causing noticeable unstable LW variations of the printed traces. Thereby, the optimal focusing frequency of Ag nanoparticle ink employed in this study has been determined to be 5.8 MHz.*” (please see page 9 in the revised manuscript)

“

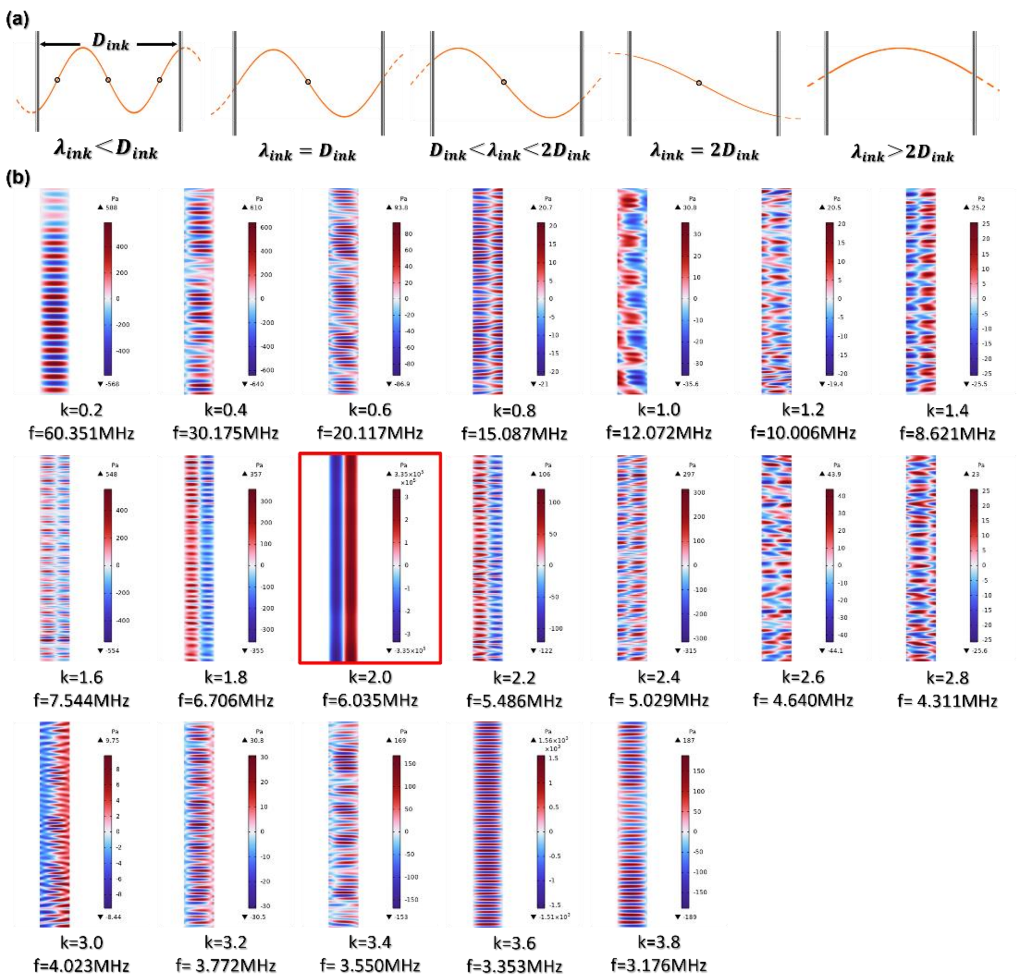


Fig. S4 a *Schematic diagram of the relationship between acoustic pressure node distribution and wavelength in ink flow.* ***b*** *Parameterized scanning ultrasound frequency search for the optimal acoustic field distribution for focusing aerosol-jet (AJ) printing particles*” (please see page 1 in the revised supplementary information)

“***5 Optimal AF frequency search experiment***

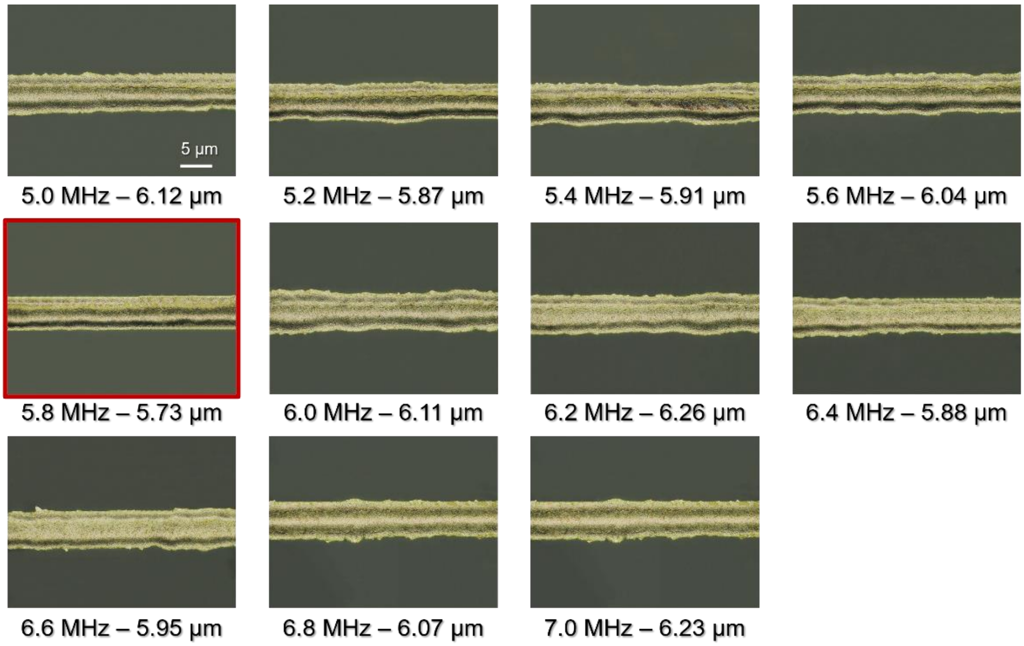


***Fig. S5** AF-assisted AJ printed trace images with a step size of 0.2 MHz in the acoustic frequency range of 5-7 MHz.*" (please see page 5 in the revised supplementary information)

Comment #11

The mention of the necessity of a power amplifier in lines 200-202 does not include details about what model or specifications were used here.

Response

Thanks very much to the reviewer for asking detailed questions and requesting supplements about the details of the AF assistance device, especially the power amplifier. This is indeed meaningful for the completeness of the AF strategy and the article. We have supplemented relevant details of the power amplifier, including model and specifications, in section 3.1. In addition, we have also added other device details.

"*The power amplifier adopts the model LZY-22+ (Mini-Circuits, USA), with an operating frequency range of 0.1-200 MHz and an output voltage of 24 V.*" (please see page 21 in the revised manuscript)

Comment #12

In line 204, values for line width and overspray (presumably width although the manuscript is not clear) include no details about the statistical significance of this data. How were these values measured. Given the nearly Gaussian curve the authors claim, what was the criteria used to determine the edge of the line and the edge of the overspray? This is not a trivial question and will have a significant effect on the results of this study. Without these details, this number provides little meaning.

Response

We appreciate your inquiry about the criteria for defining the edges of the line and the overspray. Acknowledging its importance, we regret any confusion caused and will clarify this in the revised manuscript. We have detailed the definitions and quantification of line width (LW) and overspray to enhance data extraction and analysis, thus improving our study's rigor.

"*Both the LW and the overspray in this work were defined through the utilization of the average LW. Along the printing direction, the maximum deposition width* L_{max_i} *was extracted on each pixel column in orthogonal directions. Next, the discontinuous pixel units along the printing direction were removed and a continuous deposition width* L_i *along the printing direction was obtained. Finally, the LW of the AJ printed traces was denoted as* $LW = \frac{1}{n}\sum L_i$. *The maximum printing range was defined as* $PW = \frac{1}{n}\sum L_{max_i}$. *The width of overspray was denoted as* $OW = \frac{1}{2}(PW - LW)$." (please see page 22 in the revised manuscript)

Comment #13

'minimal overspray' in line 207 has no meaning. This type of result must have some quantification of the detection limits of the measurement and analysis technique. Moreover, this reviewer disputes this claim based on Fig. 2a-b. The overspray size seems nearly identical in both cases.

Response

We are grateful for your perceptive observations on the overspray in our research and concerning on the quantification of the detection limits of the measurement and analysis.

(1) We apologize for not providing clear definition and measurement/analysis methods before. The minimal overspray of the experimental results has been quantitatively and analytically measured and reported as follows, which was grounded in the definitions and analytical methods delineated in *Comment #12*.

"*We also proposed a new print nozzle geometry, resulting in ultrafine traces (line width <6 μm, overspray <0.1 μm).*" (please see page 1 in the revised manuscript)

"*Experimentally, an ultrafine-printed Ag ink stream with a line width (LW) of 5.7 μm and overspray of 0.08 μm was achieved under AF, the corresponding focusing and defocusing process were observed which indicates the focusing behavior is fully reversible and omnidirectional.*" (please see page 4 in the revised manuscript)

"*As illustrated in Fig. 2b, an ink stream with a LW of 5.71 μm and a minimal overspray of 0.08 μm was observed without altering any printing parameters.*" (please see page 10 in the revised manuscript)

"

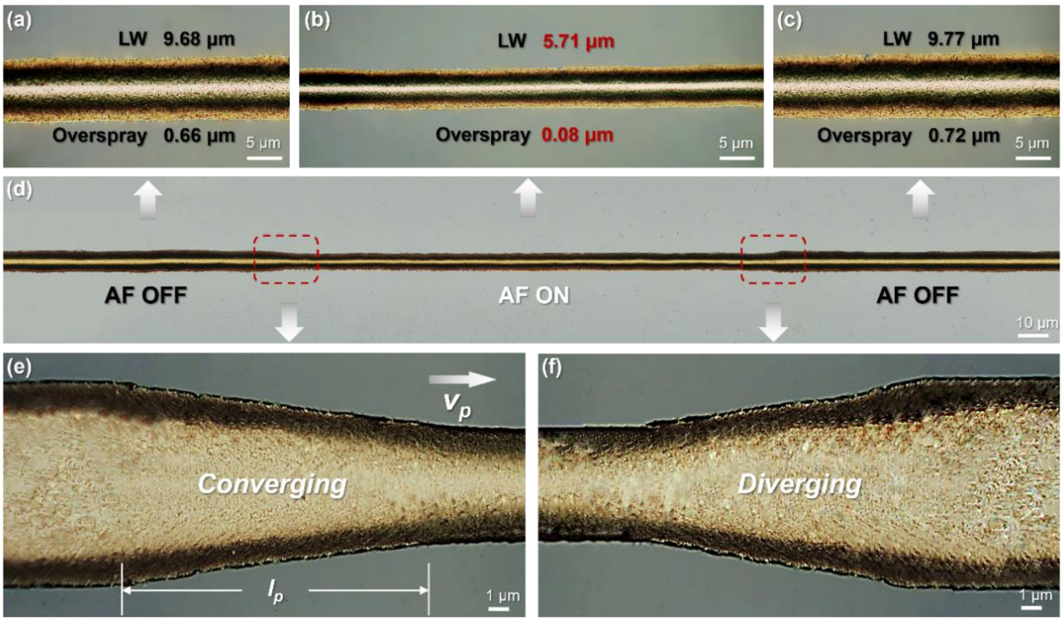


Fig. 2 The impact of AF on the geometric morphology of AJ printed line. ……" (please see page 12 in the revised manuscript)

"*As a result, it achieves an ultra-high resolution in AJ printing, up to 5.7 μm, while minimal overspray occurred to less than 0.1 μm, which completely breaks through the results of previous optimization studies, as shown in Fig. 5c.*" (please see page 18 in the revised manuscript)

(2) The overspray of the AJ printed lines in Fig. 2a, b is indeed difficult to visually discern and quantify as displayed in the text. We apologize again for any inconvenience and concerns caused to the reviewer. Therefore, we have provided additional quantitative test results for the overspray of the lines in Fig. a-c, as well as enlarged images of the edge morphology, as shown in Fig. S6d-f. (please see page 6 in the revised supplementary information)

Comment #14

Differences before and after the AF region of a line are attributed to process drift in line 210. This reviewer would like to see an analysis on how often line dimensions change positively or negatively. Without such analysis of a large sample size, the effect of AF (or lack thereof) cannot be stated with any degree of confidence.

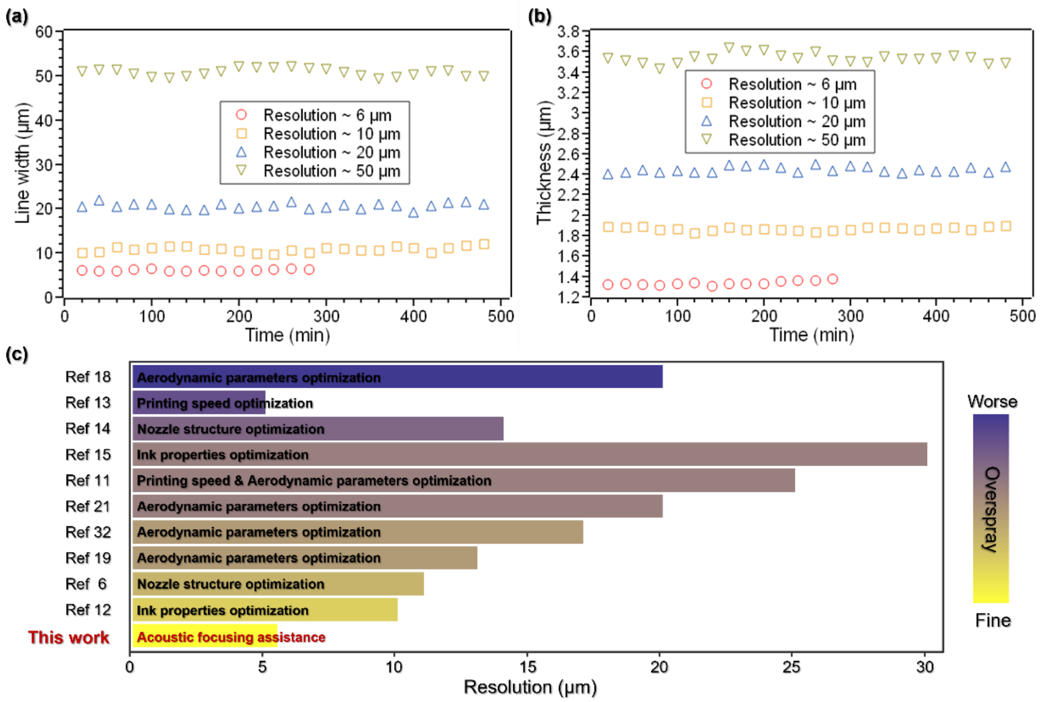
Response

We thank the reviewer for the careful consideration and in-depth questioning regarding the variation in LW of the printed traces. In the case of slight fluctuations in AJ printed LW, we have conducted four continuous experiments to get large sample data, and provided explanations on the statement of LW fluctuation.

"*It is noted that the slight disparity observed in trace morphology before and after the AF process can be ascribed to the gas flow fluctuation and drift in the ink composition over time which is common issues during AJ printing, mainly because of (a) fluctuation of carrier gas and sheath gas volumes; (b) impurities accumulate inside the nozzle, due to prolonged and repetitive use, leading to dislocation of the aerosol ink during transport; (c) manufacturing defect of the outlet roundness and internal channel standardization degree of nozzles affecting the ink flow distribution.*" (please see page 11 in the revised manuscript)

"*To assess AJ printing stability with AF, four printing stability tests were conducted under ambient conditions, which involved printing lines with the resolution of 6, 10, 20, and 50 μm respectively. Among these, the stability of 6 μm resolution lasted for approximately 4.5 hours. This duration was notably shorter compared to the other three resolutions with 10 μm, 20 μm, and 50 μm, which all maintained stability for almost 8 hours.*" (please see page 18 in the revised manuscript)

"*Although a small variation of LW was observed during the long-term printing, the average LW (6.0±0.3 μm, 10.8±1.2 μm, 20.6±1.4 μm, 50.7±1.4 μm) of the AF-assisted AJ printed traces indicates that the excellent printing stability compared to traditional AJ printing method.*" (please see page 18 in the revised manuscript)



"

Fig. 5 Stability quantification and performance comparison of AF-assisted AJ printing. *For 8-hour continuous AJ printing with resolution ~6 μm, ~10 μm, ~20 μm, and ~50 μm,* ***a*** *LWs and* ***b*** *thickness were recorded at intervals of 20 mins.* ***c*** *Comparison of different optimization methods on printing resolution and overspray of AJ printing with other reported results.*" (please see page 19 in the revised manuscript)

Comment #15

Line 214 – what is meant by 'channel'?

Response

We thank the reviewer for the meticulous review and inquiries. The term "channel" in this context specifically denotes the acoustic channel within the nozzle. We apologize for any ambiguity in our initial explanation and the subsequent lack of clarity in that section of the manuscript. The external acoustic field induces a focusing effect on the aerosol ink flow within the nozzle's channel.

"*These transition areas exhibit noteworthy patterns of focusing and defocusing in the aerosol ink flow which are hard to observe in the acoustic channel of nozzle due to the opacity and diminutive structure of the nozzle channels as well as the fast jet speed of the ink flow.*" (please see page 11 in the revised manuscript)

Comment #16

Line 218 – can the jet velocity be compared to inlet gas flows or some reference to ensure its reasonableness? Again, all of these values should come from multiple measurements and be presented with some measure of data spread.

Response

We thank the reviewer for the meticulous review and in-depth questioning of this jet velocity. If direct measurement of this value can be obtained, it would provide more conclusive evidence for this analysis work. However, considering that the measurement object is located inside a small and enclosed nozzle, and we do not yet have access to equipment such as ultrahigh-speed cameras for measuring relevant jet velocity. But through literature comparison, the average velocity inside the nozzle is similar to conventional empirical data. In addition, AF works radially and has no effect on the axial velocity.

"*The average velocity within the nozzle is in the range of the conventional empirical data*[24,61]." (please see page 11 in the revised manuscript)

"*23. Ramesh, S. et al. Numerical and experimental investigation of aerosol jet printing. Addit. Manuf.* ***59****, 103090 (2022).*

58. Rurup, J. D. & Secor, E. B. In-situ qualification and physics-based process design for aerosol jet printing via spatially correlated light scattering measurements. Addit. Manuf. ***82****, 104037 (2024).*"

(please see page 25 in the revised manuscript)

<u>Comment #17</u>

What substrate material was used here? And what platen temperature? Both of these will have a large effect on line width.

<u>Response</u>

We thank the reviewer for the meticulous review and inquiries. The choice of the substrate material and the regulation of the platen temperature do have a large effect on the printing effect. To elucidate the optimization effect of AF on AJ printing in this work, we have adopted the experimental method of controlling variables to ensure that the printing conditions both before and after the application of AF remain consistent, including the substrate material and the platen temperature. In our study, glass slides were employed as the substrate, with the platen temperature consistently maintained at 80 °C, adhering to the usage requirement of the ink.

"

Table. S3 *Printing parameters of the experiments*

Line width (μm)	***Nozzle diameter (μm)***	***Carrier gas flow rate (sccm)***	***Sheath gas flow rate (sccm)***	***Printing speed (mm/s)***	***Standoff distance (mm)***	***Platen temperature (°C)***	***Substrate material***
10	*300*	*8*	*21*	*9*	*2.5*	*80*	*Glass*
30	*300*	*15*	*33*	*9*	*2.5*	*80*	*Glass*
50	*300*	*22*	*47*	*9*	*2.5*	*80*	*Glass*
80	*300*	*36*	*78*	*9*	*2.5*	*80*	*Glass*

" (please see page 6 in the supplementary information)

In section 2.3 (and relevant methods section), I have the following concerns/questions:

<u>Comment #18</u>

Why for 'repeatability' tests does there only seem to be one cross-section measurement taken for each data set? Without multiple sections across multiple prints, the repeatability cannot be assessed. In addition, there is a high precision given on measurements here – this again begs the question of how the 'edge' of such a curve was determined.

Response

We thank the reviewer for the meticulous review and consideration of the "repeatability" tests and results. We conducted three sets of repeated experiments with four print resolutions respectively and performed three slice contour measurements for each part (before, during, and after AF) of the printed traces to get 108 data of cross-sections. We averaged the three cross-sectional areas obtained from each part, then averaged the three average areas of each type of printed part, and finally determined the values of cross-sectional area at different stages under different print resolutions. As an illustration only to express the change of the cross-section, we drew the cross-section contours using only single set of measurement data for each different print resolution. For the specific definition regarding the edge of the printed traces, we have provided explanations in *Comment #12*.

"*Three sets of repeated experiments with four print resolutions respectively were conducted in this study, and three slice contour measurements for each part (before, during, and after AF) of the printed traces were conducted, as depicted in Fig. S7, to get 108 data of cross-sections. …… The three cross-sectional areas obtained from each part were averaged, then the three average areas of each type of printed part were averaged again. Finally, the values of the cross-sectional area at different stages under different print resolutions were determined.*" (please see page 23 in the revised manuscript)

"***7 Measuring the cross-sectional profile of AJ-printed traces***

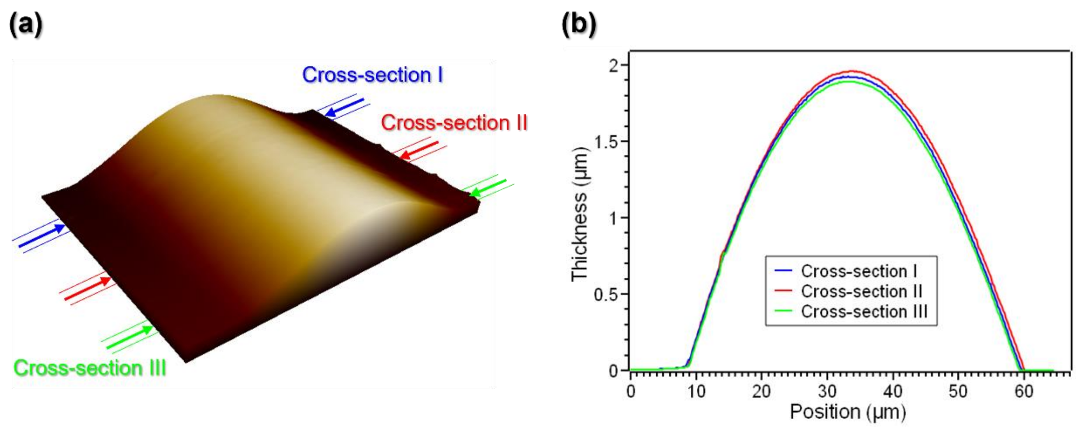


***Fig. S7 a** 3D model of the AJ-printed trace scanned by AFM. **b** Three randomly selected parts of the trace were chosen as the cross-sections (I, II, III).*" (please see page 6 in the revised supplementary information)

Comment #19

The authors define line thickness using the largest measurable – this is an acceptable choice, but I must again question how this can be a statistically significant, meaningful value based on a single measurement.

Response

We are grateful for your profound inquiry concerning the criteria used to determine the thickness of the printed trace, which is indeed a statistically significant, meaningful value in this study and article. Now we clarify the quantification and measurement method of it.

"*Within the range of both edges of the printed trace, from one edge to the opposite edge in the orthogonal direction to the printing direction, the height H_{a_i} of each pixel row along the printing direction are collected and averaged. Finally, take the maximum value $H_{a_{max}}$ of all obtained average heights as the thickness of this trace.*" (please see page 22 in the revised manuscript)

Comment #20

The definition of 'change rate' is not given.

Response

We are grateful for the reviewer's detailed review of the article, especially for their rigorous questioning regarding the definition of the physical quantity "change rate". This definition indeed needs to be clarified, otherwise it would make the article lack rigor and potentially lead to misunderstandings. We have supplemented this definition and improved other similar issues.

"*The change rate for all physical quantities in this work is defined as the ratio of the changed value to the original value, mainly related to LW, thickness, cross-sectional area, resistance and conductivity.*" (please see page 23 in the revised manuscript)

Comment #21

It is again unclear to this reviewer how the data in Fig. 3e can be considered proof of repeatability without some quantitative measure of line dimensions, particularly when the authors admit there are variations. There is no evidence on convincing reference that these variations can be 'primarily attributable to the fluctuation in gas flow'. I would also dispute that these results can be combined with others to prove the techniques applicability to multiple directions.

Response

We are grateful for the reviewer's concern about the fluctuation of LW in the repetitive test and the applicability of AF-assisted AJ printing to multi-directional. First, we would like to explain the situation of minor LW fluctuation in AJ printing itself, and the same one in AF-assisted AJ printing, as described in both theoretical analysis and experimental results in *Comment#14*. Secondly, we have quantified the LW at each stage of this result, including image representation and supplementary data as follows.

"*It is noted that a slight variation in the LW of printed traces under the same AF circumstances, quantified in Fig. 3g, is primarily attributable to the fluctuation in gas flow within the AJ printing system. ……*

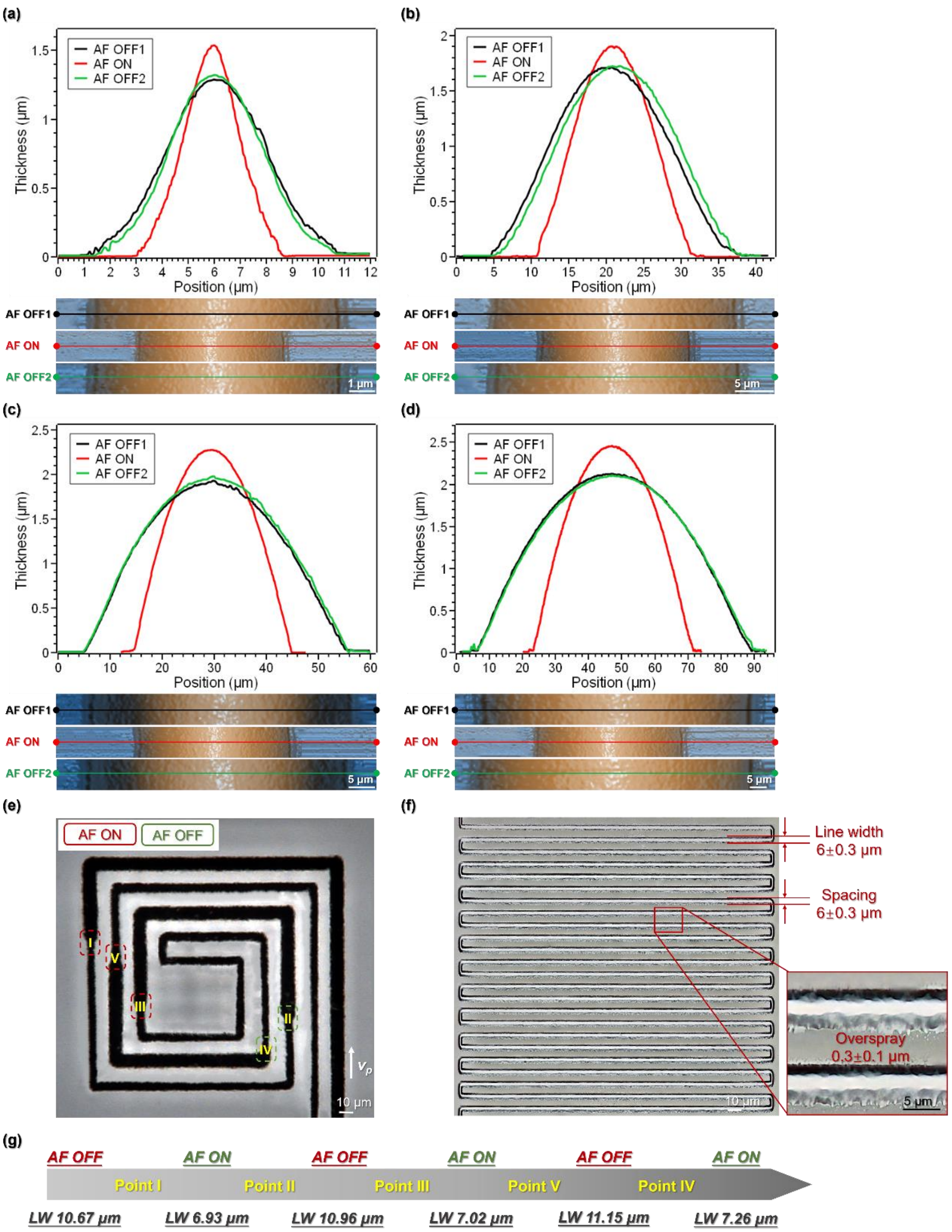


Fig. 3 …… ***g*** *LW of a continuously AJ printed trace with multiple AF ON/OFF varies in different directions.*" (please see page 13 in the revised manuscript)

Comment #22

Lines 256-263 give the only true measure of repeatability in this section, but vital details are again omitted here. How many data points are used in these values? What statistical measure is used in the +/-? Analysis is again qualitative in describing overspray as 'tiny' and of questionable veracity. Fig. 3f has visible overspray, particularly in the inset. This again brings the question of how is overspray being measured? As an average deviation from the line? Given that electrical shorting is the concern, it seems more useful to measure maximum distance at which overspray is found or some measure of density vs. distance from the line's edge. Much more detail is required to describe the work.

Response

We are grateful for the reviewer's deep concern regarding the geometric measurement method and repeatability verification of printed array, as well as the line density and short circuit issues in high-density electronic printing applications. We have provided supplementary explanations on the definition and measurement method of LW and overspray width in *Comment #12&13*. In this work, we quantitatively processed both the LW and overspray of the 24 printed circuits of the displayed array separately, and finally obtained the average values of the presented LW and overspray width. Additionally, based on the suggestions from the reviewer regarding short circuit issues in high integration printed circuit applications, we have provided corresponding analysis and evaluation.

"*The array was composed of 24 lines and corresponding morphological characteristic values (LW: 5.9±0.3 μm, overspray: 0.3±0.1 μm, line gap: 6.0±0.3 μm) were obtained separately based on the definition and measurement methods of LW and overspray in this study. The range and average value of them were obtained from the 24 sets of data. In terms of theoretical analysis, the sum of the maximum overspray width and half of the maximum LW was much smaller than the axial spacing between adjacent printed lines, i.e.* $2 \cdot OW_{max} < spacing$*. In addition, we measured all adjacent lines, and the results showed that there was indeed no electrical shorting in any adjacent printed lines.*" (please see page 13 in the revised manuscript)

Comment #23

Fig. 2b – why the apparent lateral shift before and after the AF?

Response

We are very grateful for the reviewer's meticulous attention to the morphology of the AF-assisted AJ printed traces. This phenomenon is not a real lateral shift of the deposition, but rather caused by the image acquisition. The printed trace under the microscope was not horizontally aligned with respect to the measurement field of view.

Comment #24

Fig. 2c & d – the is an apparent shoulder in the AF ON lines – is this not evidence of measurable overspray that could be quantified?

Response

We thank the reviewer for the careful review of the printing results and the meticulous consideration of the definition of overspray for this work. From a visual perspective, it is indeed possible to define the seemingly shoulder-like deposition as overspray. We have comprehensively defined overspray from geometric and electrical perspectives, defining it as adjacent discontinuous deposition along the printing direction/current direction, and indicating quantification methods under this definition.

"*Both the LW and the overspray in this work were defined through the utilization of the average LW. Along the printing direction, the maximum deposition width* L_{max_i} *was extracted on each pixel column in orthogonal directions. Next, the discontinuous pixel units along the printing direction were removed and a continuous deposition width* L_i *along the printing direction was obtained. Finally, the LW of the*

AJ printed traces was denoted as $LW = \frac{1}{n}\sum L_i$*. The maximum printing range was defined as* $PW = \frac{1}{n}\sum L_{max_i}$*. The width of overspray was denoted as* $OW = \frac{1}{2}(PW - LW)$*.*" (please see page 22 in the revised manuscript)

In section 2.4 (and relevant methods section), I have the following concerns/questions:

Comment #25

The data in Fig. 3a-d and Supplementary Table S2 do not seem to agree on quantities. Given the definition of line thickness, line width, and apparent Gaussian cross-section, the values given for cross-sectional area seem to be quite large. It seems warranted to check these values again for any calculation error or provide further details on how this area was calculated.

Response

We are grateful for the reviewer's detailed review of the geometric data, especially the thorough inquiry into the measurement details of the cross-sectional area. The definition and measurement method of line width and thickness are provided in *Comment #12*. Here, we supplement the definition and measurement method of cross-section area as follows.

"*For cross-sectional area calculating, the entire contour discrete data points are imported into XEI software and fitted by polynomial fitting. Then a trapezoidal discrete integration was applied to calculate the cross-sectional contour area.*" (please see page 23 in the revised manuscript)

Comment #26

In line 283, it is suddenly mentioned that the traces were 'annealed'. None of the conductivity data has any meaning whatsoever without details about the heating process. Was this merely drying the traces? Given the ink used, the manufacturer gives a range of resistivities equal to about 2.5 orders of magnitude from 120C to 200C. These values also cover the entire range of conductivities shown in this work. Given this information, the method of heating the samples and is vitally important to evaluating the degree of conductivity as well as how likely any differences can actually be attributed to the effects claimed here.

Response

We are grateful for reviewer's careful attention to the research method of conductivity. The method of heating printed traces indeed has an extremely important impact on their conductivity. To fully illustrate the effect of AF on conductivity, we maintained consistent experimental procedures outside of AF, including the sintering process. Additionally, no optimization or adjustment was made to the initial conductivity during the research process and the original conductivity may not seem very excellent.

"*To fully illustrate the effect of AF on conductivity, we maintained a consistent sintering process, and all printed samples were sintered using the same sintering program.*" (please see page 15 in the revised manuscript)

"*The sintering process involved heating to 200 °C for 30 minutes, then holding for one hour, and finally cooling down to room temperature in the heating oven (OGH60-S, Thermo Scientific).*" (please see page 23 in the revised manuscript)

Comment #27

The definition of 'change rate' is not given.

Response

We are grateful for the reviewer's detailed review of the article, especially for their rigorous questioning regarding the definition of the physical quantity "change rate". This definition indeed needs to be clarified, otherwise it would make the article lack rigor and potentially lead to misunderstandings. We have supplemented this definition and improved other similar issues.

"*The change rate for all physical quantities in this work is defined as the ratio of the changed value to the original value, mainly related to LW, thickness, cross-sectional area, resistance and conductivity.*" (please see page 23 in the revised manuscript)

Comment #28

In Fig. 4a & b, what do the error bars represent?

Response

We thank the reviewer's detailed review and questions on the electrical performance results (Fig. 4a&b). Since the resistance values are obtained through measurements, we conducted 5 sets of resistance tests of AJ printed traces with/without AF. The average resistance value, error bar, and change rate shown in Fig. 4a were derived from all the results obtained. The conductivity results for different depositions were also calculated based on the corresponding resistance values and their geometric values to obtain average values and error bars.

Comment #29

Line 293 – was particle size checked before or after the annealing process? Small differences could be obscured by the onset of sintering.

Response

We thank the reviewer for giving deep thoughts on the possible particle size variation. The sintering effect considered by the reviewer is indeed rigorous, however, for deposited ink before annealing, it is unable to check the particle size using SEM or other methods. Here, we will supplement and limit the wording and conditions mentioned in the text.

"*It should be noted that the annealed particle size remains unchanged after undergoing the AF process, which indicates that the AF has no damage to the nanoparticles for AJ printing that requires sintering.*" (please see page 15 in the revised manuscript)

Comment #30

How were the samples cross-sectioned? The images appear to be different in geometry. With poor adhesion between particles, the method of sectioning can have a large effect on the observed morphology.

Response

Thank you for the thorough review and consideration of sectioning sample. Considering that the substrates used for AJ printing in this study were glass slides, in order to avoid external contact and its impact on the sample, we used a glass knife to cut a line, which was perpendicular to the printed trace, on the other side of the glass slide. Then, we bent it towards this side of no deposition until it broke off, thus obtaining cross-sections of the printed traces. This method of sectioning does introduce some errors into the results, but it ensures that the obtained cross-section is not affected by external contact and does not affect the internal structure. In addition, a better sampling method that meets this basic requirement has not been considered yet.

Comment #31

Without more details the porosity measurement is not meaningful. Algorithms to measure porosity on a cross-section which is not flat and polished can have high error. Images showing the detected pores on top of the relevant image would be one way of demonstrating this.

Response

We thank the reviewer for the thorough review of the porosity measurement method. Here, we supplement the whole porosity measuring process, as depicted in Fig. R1, including printing traces, making cross-sections, and SEM measurements. We designed adjacent parallel printing tracks and added AF midway to obtain three completely parallel and continuously printed traces. Then we used a glass knife to slide in a direction perpendicular to the traces on the other side of the glass slide and bended the glass to break it, thus obtaining three cross-sections of the printed traces. It is worth noting that the flatness of these three cross-sections obtained in this way is relatively consistent. Finally, we used SEM to measure the three cross-sections on the same glass slide. It ensures that the observation angles of the three sections are also consistent. In summary, even if there may be errors in measurement angles in this study, but the measurement angles from three sections are relatively error-free.

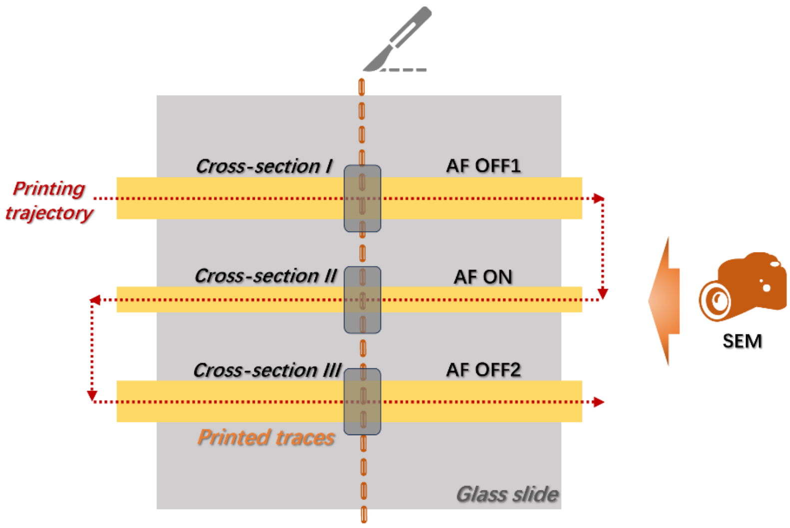


Fig. R1 Schematic illustration of the whole porosity measuring process.

Comment #32

Density is also limited by the stacking of circular particles. A comparison of the relative contribution is warranted. That is to say – how close or far to the theoretical maximum packing density is either case discussed here?

Response

Thank you for the deep consideration of packing density of ink particles. This discussion currently started from an obvious qualitative perspective or approximate quantitative analysis to discuss the change of the packing density of the printed ink particles, in order to explain the excellent effect of a significant improvement in conductivity. For the quantitative manipulation of particle stacking density by AF and optimization research on theoretical stacking, we consider conducting further research on it as future work.

Comment #33

The statements in lines 304-313 seem quite out of place. This information is relevant during the modeling portion of the work more than in this location (as mentioned above in previous comments).

Response

The particle focusing motion for resolution optimization has been explained in Fig. 1b-f, while the particle aggregation here aims to explain the increase in conductivity through the increase in the particle packing density.

Comment #34

In Fig. 4i, you show the coalescing of microdroplets as a result of nanoparticle focusing. To this reviewer's best judgement, none of the work here proves this mechanism. A smaller material stream with consistent volumetric flow will have a higher velocity. It is, therefore, possible that droplets in the aerosol stay the same size and merely their arrangement changes. Unless there is compelling evidence of droplet size increasing (already expected to be 1-5 um in diameter, close to the minimum line width shown here), this schematic should be reconsidered or given with sufficient disclaimer that it is merely a hypothesis of the flow state.

Response

We thank the reviewer's deep attention and consideration to this schematic illustration. The particle aggregation changes envisioned in this schematic are based on studies of similar particle distribution changes (Ref. 63), aiming to explain the decrease in cross-sectional area and the increase in conductivity through an increase in particle packing density. We acknowledge that it is only a hypothesis, but we maintain support for this hypothesis and are committed to further confirming or exploring the microstructural nature in future research.

In section 2.5 (and relevant methods section), I have the following concerns/questions:

Comment #35

Why was printing stability not measured for the 6 um line width? Without this information, the value of this work is significantly diminished. Its lack also makes this reviewer concerned about the actual viability of the claims in this work.

Response

We agree with the reviewer's comment that the stability for the smallest line width would be a very important point for readers who may wish to use this approach. Therefore, this has now been discussed in the revised manuscript.

"*To assess AJ printing stability with AF, four printing stability tests were conducted under ambient conditions, which involved printing lines with the resolution of 6, 10, 20, and 50 μm respectively. Among these, the stability of 6 μm resolution lasted for approximately 4.5 hours. This duration was notably shorter compared to the other three resolutions with 10 μm, 20 μm, and 50 μm, which all maintained* *stability for almost 8 hours. The discrepancy can be attributed to the instability observed in the original optimal resolution of 10 μm without AF, but still has greatly improved stability under ultrahigh printing resolution. An optical image was captured every 20 minutes as shown in Supplementary Fig. S9, the LW, and thickness of the trace at various time points can be viewed in Fig. 5a, b. Although a small variation of LW was observed during the long-term printing, the average LW (6.0±0.3 μm, 10.8±1.2 μm, 20.6±1.4 μm, 50.7±1.4 μm) of the AF-assisted AJ printed traces indicates that the excellent printing stability compared to traditional AJ printing method. Compared to the stability shown by the LW approach, there was a significant fluctuation in trace thickness which suggests that the deposition rate varies during printing, a common occurrence in practical applications. Specifically, there is a variation in deposition volume with consistent width but noticeable difference in height. However, this highlights the optimizing impact of AF in AJ printing to maintain the quality and stability of printed LW despite fluctuations in deposition rate. ……*

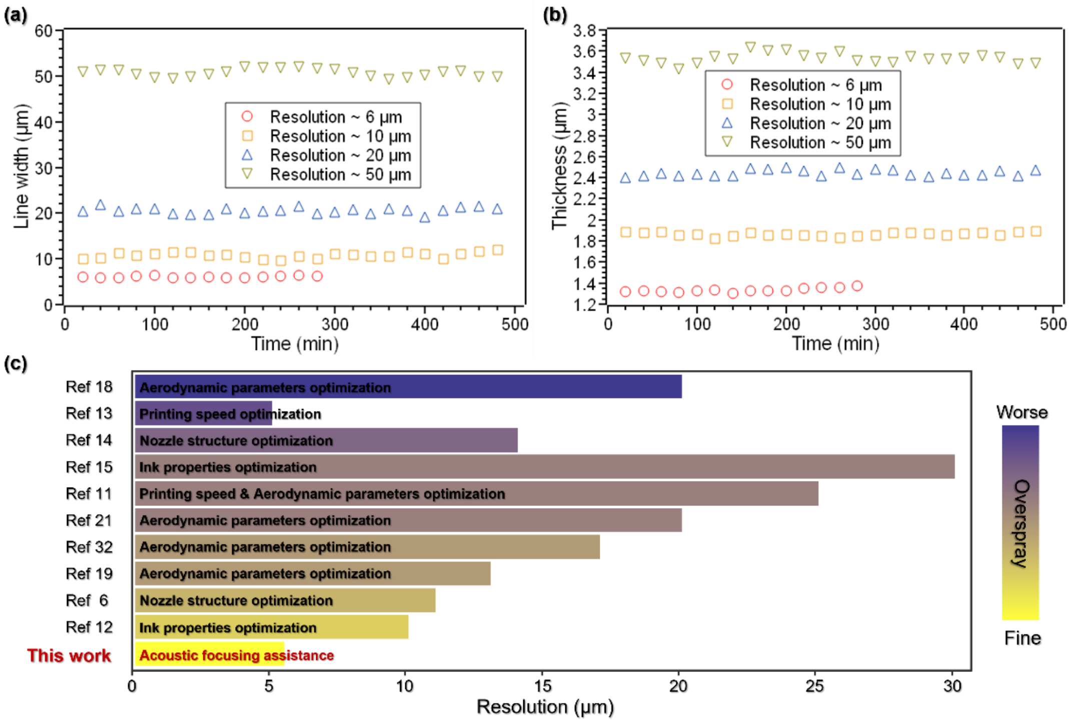
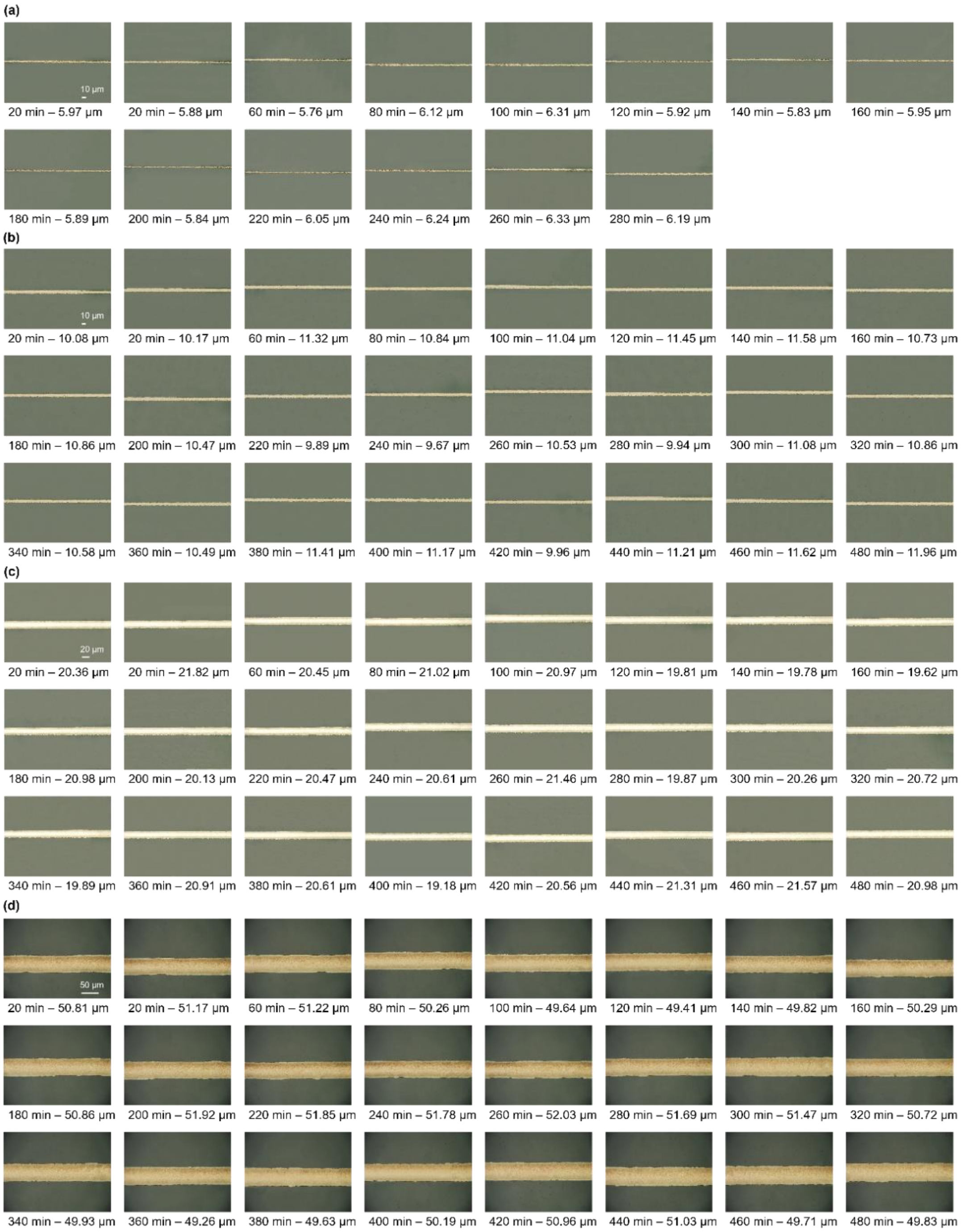


Fig. 5 Stability quantification and performance comparison of AF-assisted AJ printing. a *For 8-hour continuous AJ printing with resolution ~6 μm, ~10 μm, ~20 μm, and ~50 μm,* ***a*** *LWs and* ***b*** *thickness were recorded at intervals of 20 mins.* ***c*** *Comparison of different optimization methods on printing resolution and overspray of AJ printing with other reported results."* (please see page 18 in the revised manuscript)



“*Fig. S9 Continuous AJ printing assisted with AF, during which the morphology and geometric values of the printed line were measured in steps of 20 minutes.* ***a*** *LW ~6 µm for 4.5 hours.* ***b*** *LW ~10 µm for 8 hours.* ***c*** *LW ~20 µm for 8 hours.* ***d*** *LW ~50 µm for 8 hours.*” (please see page 9 in the revised supplementary information)

Comment #36

Line widths seem always above the planned values in Fig. 5a. Why?

Response

We thank the reviewer for the carefully reviewing and asking detailed questions about the article. Here, we conducted continued printing experiments and analyzed the stability of AF assisted AJ printing at different printing resolutions. Given that stability is based on a certain initial resolution level, we did not strictly control the initial resolution value to planed level, but instead maintained it at a level slightly higher originally intended. Therefore, the reported line widths remained consistent and their values were above the planned value. However, under such randomly obtained printing resolution, the AF assistance enables it to demonstrate good stability as well. This also fully demonstrates that the stability of AF-assisted AJ printing is quite considerable. To address this concern of the reviewer, we have re-conducted stability tests at different initial resolution, in which each initial resolution was adjusted to a nearly planed level.

We agree with the reviewer's comment that the stability for the smallest line width would be a very important point for readers who may wish to use this approach. Therefore, this has now been discussed in the revised manuscript.

"*To assess AJ printing stability with AF, four printing stability tests were conducted under ambient conditions, which involved printing lines with the resolution of 6, 10, 20, and 50 μm respectively. Among these, the stability of 6 μm resolution lasted for approximately 4.5 hours. This duration was notably shorter compared to the other three resolutions with 10 μm, 20 μm, and 50 μm, which all maintained stability for almost 8 hours. The discrepancy can be attributed to the instability observed in the original optimal resolution of 10 μm without AF, but still has greatly improved stability under ultrahigh printing resolution. An optical image was captured every 20 minutes as shown in Supplementary Fig. S9, the LW, and thickness of the trace at various time points can be viewed in Fig. 5a, b. Although a small variation of LW was observed during the long-term printing, the average LW (6.0±0.3 μm, 10.8±1.2 μm, 20.6±1.4 μm, 50.7±1.4 μm) of the AF-assisted AJ printed traces indicates that the excellent printing stability compared to traditional AJ printing method. Compared to the stability shown by the LW approach, there was a significant fluctuation in trace thickness which suggests that the deposition rate varies during printing, a common occurrence in practical applications. Specifically, there is a variation in deposition volume with consistent width but noticeable difference in height. However, this highlights the optimizing impact of AF in AJ printing to maintain the quality and stability of printed LW despite fluctuations in deposition rate.* ……

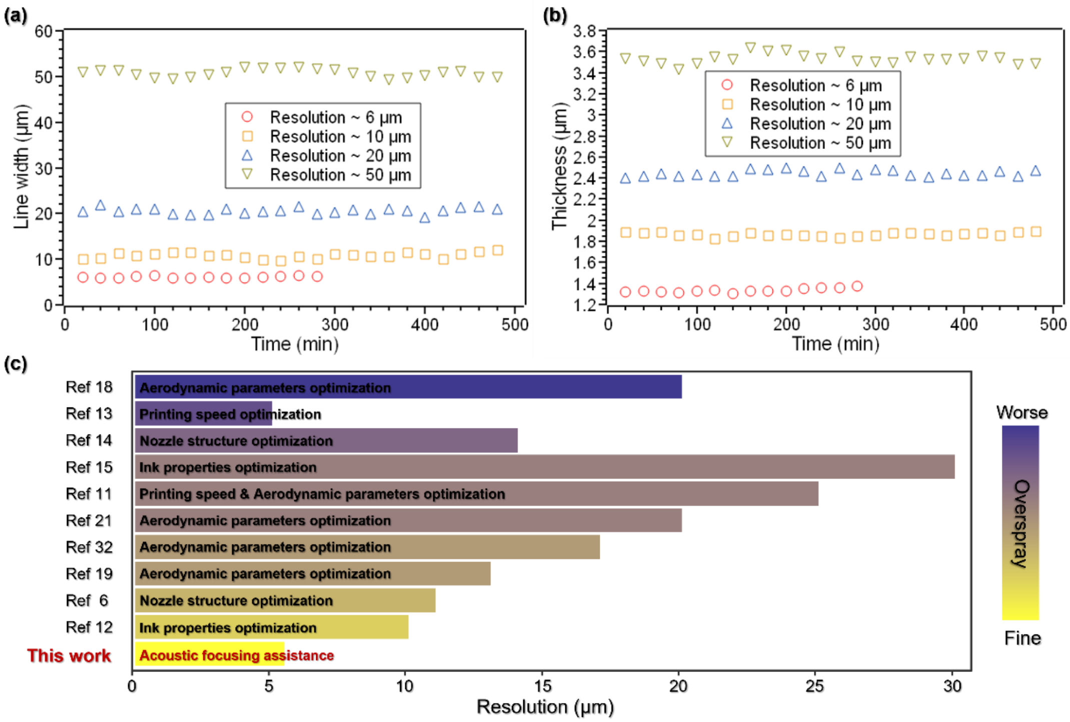


Fig. 5 Stability quantification and performance comparison of AF-assisted AJ printing. a *For 8-hour continuous AJ printing with resolution ~6 μm, ~10 μm, ~20 μm, and ~50 μm,* ***a*** *LWs and* ***b*** *thickness were recorded at intervals of 20 mins.* ***c*** *Comparison of different optimization methods on printing resolution and overspray of AJ printing with other reported results.*" (please see page 18 in the revised manuscript)

Comment #37

All of the discussion in line 330-338 is speculation without data from smaller nozzles. In addition, clogging issue in aerosol jet are a matter of statistics rather than a yes/no scenario. Actual tests with multiple nozzle sizes would give statistical meaning to this analysis. Otherwise, it should be removed from this section entirely.

Response

We are grateful for the reviewer's meticulous and in-depth inquiry into the clogging issue in AJ printing. The discussion here is indeed based on speculative experience. However, our current equipment conditions cannot yet produce nozzles with smaller outlets and conduct more comprehensive statistical experiments. We agree to remove this part of the content for now.

Comment #38

Fig. 5b aggregates data but adds little without additional discussion on the methods used in the cited works. Also, how was overspray compared between these works? It is highly material and process parameter dependent.

Response

We appreciate the reviewer's detailed questions on comparative studies and method comparison standards. To address this, we add to our discussion on the methods from cited works. We only focus on the direct overspray ratio of these excellent advanced research results, defined as the ratio of overspray width to effective line width. We uniformly adopt their printed result images using the LW and overspray definition method in this study for image recognition, obtaining the overspray ratio, and finally obtaining the contrasting results presented in Fig. 5c.

"*…… which completely breaks through the results of previous optimization studies, as shown in Fig. 5c. This comparison is based on the intuitive printing results mainly including the resolution and overspray ratio, which is defined as the ratio of overspray width to effective line width, attached with the excellent advanced research methods worth learning and thinking about respectively.*

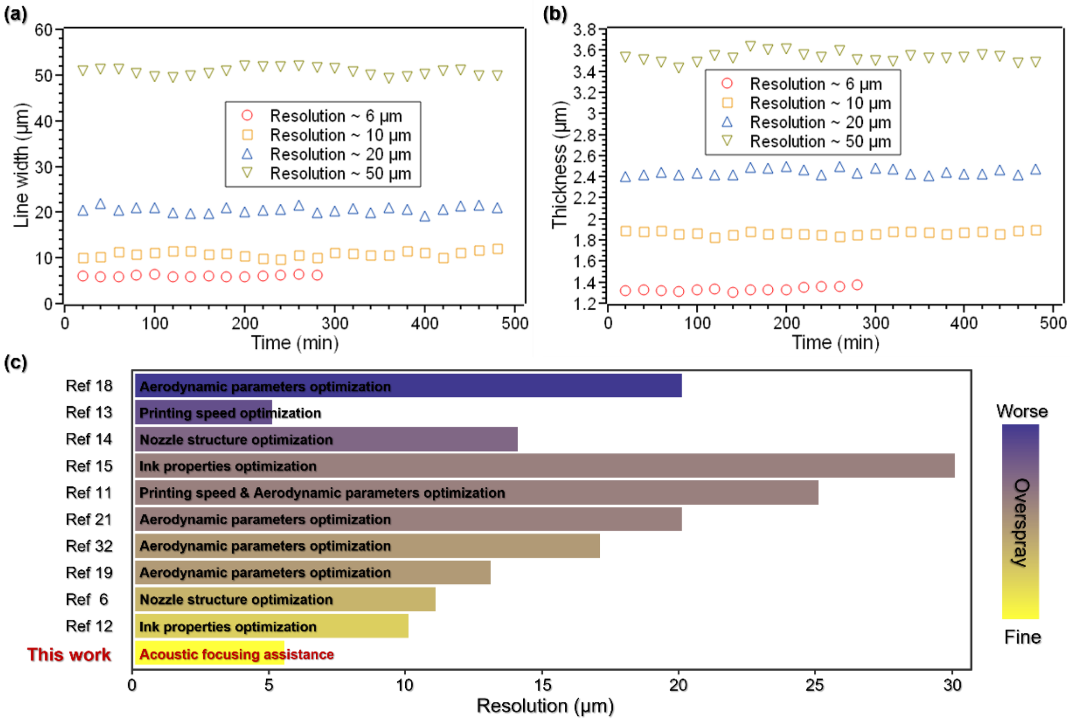


Fig. 5 Stability quantification and performance comparison of AF-assisted AJ printing. a *For 8-hour continuous AJ printing with resolution ~6 μm, ~10 μm, ~20 μm, and ~50 μm,* ***a*** *LWs and* ***b*** *thickness were recorded at intervals of 20 mins.* ***b*** *Comparison of different optimization methods on printing resolution and overspray of AJ printing with other reported results.*" (please see page 18 in the revised manuscript)

Comment #39

As a general comment, the authors attribute multiple changes to process drift without any evidence and between claims of efforts made to mitigate process drift. Please reconsider the veracity of these claims. In addition, authors claim this work is 'material agnostic' in line 349, while only demonstrating one material. Although AJ printing has little dependency on the target material, the work shown here depends on the fluid properties of solvent and additives. As such, this phrase should be removed or proven.

Response

We thank the reviewer for the thorough review of process drift and material agnosticism. The explanations for each of the questions regarding these two aspects are provided as follows.

(1) The process drift is explained with practical factors and outcomes, and the optimization effect of AF on this situation is shared and analyzed.

"*It is noted that the slight disparity observed in trace morphology before and after the AF process can be ascribed to the gas flow fluctuation and drift in the ink composition over time which is common issues during AJ printing, mainly because of (a) fluctuation of carrier gas and sheath gas volumes; (b) impurities accumulate inside the nozzle, due to prolonged and repetitive use, leading to dislocation of the aerosol ink during transport; (c) manufacturing defect of the outlet roundness and internal channel standardization degree of nozzles affecting the ink flow distribution.*" (please see page 11 in the revised manuscript)

(2) For the "material agnostic" of this work, first of all, the acoustic field and its focusing function are not designed and acted based on the material properties of a certain ink. Secondly, the purpose of this method is to optimize the original printing effect (printing resolution, overspray, conductivity) of a certain ink to a certain extent. Of course, for inks with different material properties, the optimization rate is another matter that we will further explore in future research.

"*It is worth mentioning that this remains almost consistent for each type of ink. Based on that the acoustic frequency is directly determined by the wavelength, and the design of the wavelength is based on the nozzle geometry, which will be explained as follows, the optimal focusing frequency is uniquely determined by the nozzle geometry. For the purpose of focusing the particles, it is required that the pressure node of the acoustic wave be set at this axis position. There exists an optimal wavelength that satisfies this distribution, which enables the entire acoustic field to form an optimal acoustic pressure distribution and achieve an optimal focusing effect for printing ink particles. This design is not about the ink. For different inks printed using a nozzle of the type in this study, their optimal focusing frequencies are consistent, however, due to differences in ink particle properties, there may be variations in achievable focusing degree at this optimum frequency. What's more, although the nozzle geometry design in this study has achieved comparable printing results compared with ones using ordinary nozzle, there is still room for optimization in its geometry and corresponding printing effects. As a result, the corresponding optimum frequency will undergo corresponding changes accordingly when using a possible more excellent nozzle geometry.*" (please see page 10 in the revised manuscript)

Comment #40

Overall, it is unclear to this reviewer whether the claimed improved resolution of 6 um can actually be achieved readily using this method. The data shown here seems to be the result of one or two successful prints without any measure of statistical significance. Without much more detail, likely several pages of methodology, and many repetitions in experimental data, it is impossible to have any confidence in the repeatability or impact of this work. As such, I must recommend rejection of this manuscript at this time, despite the novelty of the idea presented here.

Response

We appreciate your thorough assessment of our manuscript and the insights you've provided regarding the clarity and robustness of our methodology. We understand your concerns regarding the claimed improved resolution of 6 μm and the need for additional evidence to support our findings. To address this concern, we have supplied additional experimental details, including a more extensive methodology section outlining the printing process and parameters used, as well as a thorough analysis of the repeatability of our method through multiple repetitions of the experiments. Continuous 4.5-hour printing with the ultrahigh resolution of approximately 6 μm should serve as direct evidence of our research. Thanks again to the reviewer for the comprehensive review and suggestions. It is believed that with the addition of the above research content, the quality of our manuscript can be greatly improved.

"*As a result, it achieves an ultra-high resolution in AJ printing, up to 5.7 µm, while minimal overspray occurred to less than 0.1 µm, which completely breaks through the results of previous optimization studies, as shown in Fig. 5c.*

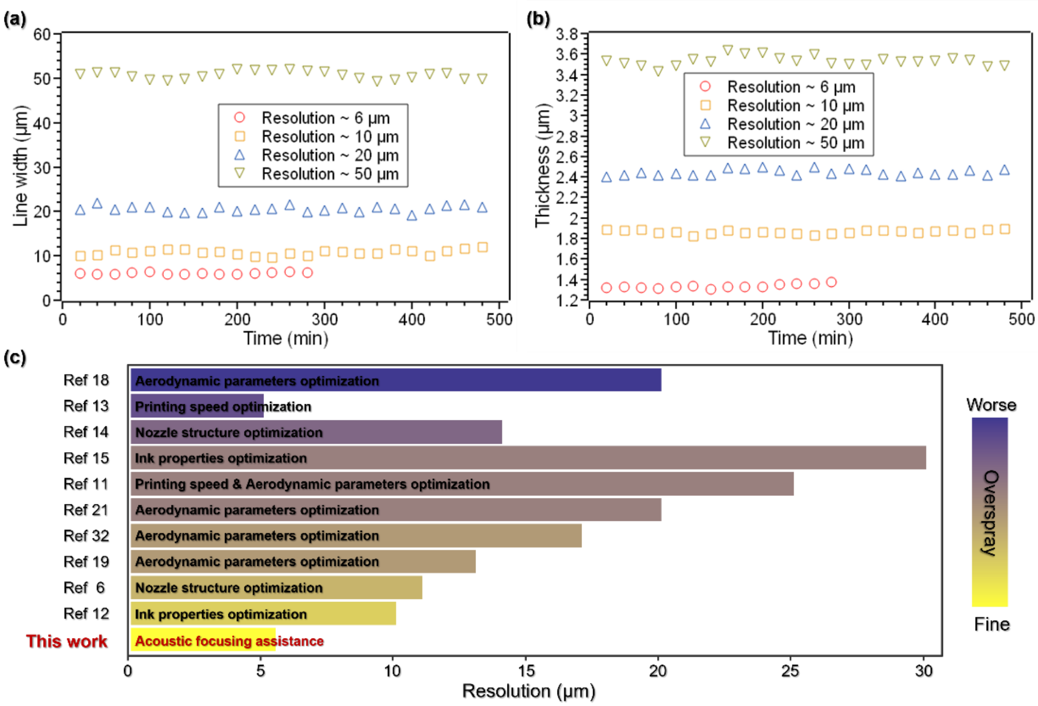


Fig. 5 Stability quantification and performance comparison of AF-assisted AJ printing. *For 8-hour continuous AJ printing with resolution ~6 µm, ~10 µm, ~20 µm, and ~50 µm,* ***a*** *LWs and* ***b*** *thickness were recorded at intervals of 20 mins.* ***c*** *Comparison of different optimization methods on printing resolution and overspray of AJ printing with other reported results.*" (please see page 18 in the revised manuscript)

REVIEWER COMMENTS

Reviewer #2 (Remarks to the Author):

Review for manuscript NCOMMS-23-48632:
The paper has been substantially improved. At this second round of review, the paper should be accepted pending some minor revisions outlined below.

1. In line 55, please revise the statement "which has an optimal value corresponding to the highest resolution obtained, while both the final resolution and its stability is constrained by the unstable turbulent and printing equipment". What is turbulent in this case? Consider changing the end of the statement to: … obtained. This optimal value was reported to depend on the level of turbulence in the flow field.

2. In line 98, consider changing the end of the statement to: in order to form an annular acoustic field that radially focuses the APS toward the center of the piezoelectric tube. The statement "from all angles" has the same meaning as "radially focuses".

3. For line 88, please replace the term "nozzle analog" to "nozzle analog(ref 55), as well as demonstrating acoustic focusing within a working powder-fed DED machine, to increase catchment efficiency(ref 56)." Consider using the same term "powder-fed directed energy deposition (DED) additive manufacturing", also introducing the DED acronym, when discussing Huang's work in line 83. Can also include this term for Smith's work: … processing conditions in powder-fed DED as an adaptive control technique ...

4. In line 288, it says 30 m/s from the reported result. In the previous review I suggested using "reported result", but a more clear statement could be "measured result from the experiments in this article". I'm not sure why reference 24 is used, is this where the number actually came from or does it refer to how this number was measured? Please clarify what numbers where measured and simulated for this article.

5. In line 533, what is the unit for the number 5 in the statement: "density distribution of order 5 at the inlet"? What is it meant by "order 5"?

6. For Fig. S2, what are the units for Particle density? Is this particle concentration? Consider using "Particle concentration (# particles/mm^3)" for the y-axis. This will require an estimate of the mass flowrate, or could be stated as normalized with the flowrate. Also, how was this particle density computed? Note that if doing a 2D simulation of particles along a line (is this what is shown in Fig. S2 (e)?), one should take into account that the concentration for a fixed number of particles will change in proportion to the area of the concentric circles (this radial concentration calculation is like a weighted histogram) around the center of the tube where they fall. Reference 55 discusses this.

7. In line 289, it is noted that 2- and 3-dimensional track measurements are included in Fig. S6 g, h, however, no mention of how this is related to reference 60 is included. The reference can either be removed, or it might be best to move it to the conclusion section, along with a comment that for future work it could be shown that the track profile (Fig. S6 h) may be predicted from the particle concentration distribution in S2 f using the convolution model from reference 60.

Reviewer #3 (Remarks to the Author):

Impressive and interesting result. All concerns have been addressed.

Reviewer #4 (Remarks to the Author):

The authors have addressed several of my comments well. However, following major comments need to be addressed before the paper can be accepted in the journal:

A] The authors have not fully address comment #7 where no commercially available nozzle was compared. There was no explanation provided either.

B] Comment #29, authors should use XRD or other methods to confirm that the particle sizes remain unchanged with and without AF (with XRD, the authors can use HWFM technique).

C] Comment #31, authors should conduct cross-sectioning methods so that flat and comparable cross-sections can be achieved after polishing. The response did not address the reviewer's concerns.

D] Comment #34, there is still no sufficient disclaimer indicating that Figure 4i is merely a hypothesis. No evidence has been provided to prove the nanoparticle focusing due to AF either.

Response to Reviewers of NCOMMS-23-48632A

We extend our heartfelt appreciation to the reviewers for their insightful comments and to the Editor for affording us the chance to enhance our manuscript (NCOMMS-23-48632A) entitled "Enhanced aerosol-jet printing using annular acoustic field for ultrahigh resolution and minimal overspray with improved conductivity" submitted to Nature Communications. After careful consideration of the feedback received, we agree that the manuscript has been substantially improved by integrating the recommended revisions. We have highlighted all revisions in red in the revised version. In the following sections, the reviewer's comments are in black, our responses to the comments are in blue and red indicates the text added/modified in the manuscript.

Reviewer #2 (Remarks to the Author):

Review for manuscript NCOMMS-23-48632:

The paper has been substantially improved. At this second round of review, the paper should be accepted pending some minor revisions outlined below.

Response

We thank the reviewer for the positive evaluation of our revised manuscript and we are pleased to hear that the paper has been substantially improved. We are committed to addressing the minor revisions you have outlined. We have carefully revised the manuscript accordingly, and believe that these changes will further enhance the quality and clarity of our work.

Comment #1

In line 55, please revise the statement "which has an optimal value corresponding to the highest resolution obtained, while both the final resolution and its stability is constrained by the unstable turbulent and printing equipment". What is turbulent in this case? Consider changing the end of the statement to: … obtained. This optimal value was reported to depend on the level of turbulence in the flow field.

Response

We appreciate the reviewer's careful review and consideration of the wording of the article. The term "turbulent" in the original manuscript was indeed unclear, and we accept the suggested revisions and have made the corresponding changes in the article.

"*The focusing ratio, defined as the ratio of sheath gas flow rate to carrier gas flow rate, was considered as a key parameter to determine the geometry of printed traces*[31]*, which has an optimal value corresponding to the highest resolution obtained. This optimal value was reported to depend on the level of turbulence in the flow field.*" (please see page 2 in the revised manuscript)

Comment #2

In line 98, consider changing the end of the statement to: in order to form an annular acoustic field that radially focuses the APS toward the center of the piezoelectric tube. The statement "from all angles" has the same meaning as "radially focuses".

Response

We appreciate the reviewer's careful review of the article. We accept this comment and have revised the corresponding statement in the article.

"*A new print nozzle geometry with two tapered focusing zones was proposed to fix the annular piezoelectric tube, in order to form an annular acoustic field that radially focuses the APS toward the center of the piezoelectric tube.*" (please see page 4 in the revised manuscript)

Comment #3

For line 88, please replace the term "nozzle analog" to "nozzle analog (ref 55), as well as demonstrating acoustic focusing within a working powder-fed DED machine, to increase catchment efficiency (ref 56)." Consider using the same term "powder-fed directed energy deposition (DED) additive manufacturing", also introducing the DED acronym, when discussing Huang's work in line 83. Can also include this term for Smith's work: … processing conditions in powder-fed DED as an adaptive control technique ...

Response

We appreciate the reviewer's detailed suggestions. Accordingly, we have adjusted the position of the citations and added introductions of DED in the current research status of the relevant experts.

"*Huang et al. presented an electrodynamic concentration approach for submillimeter-sized conductive metal particles focusing on moving gas-powder streams to generate a tightly focused powder stream in powder-fed directed energy deposition (DED) additive manufacturing*[53]*. Smith et al. demonstrated a methodology allowing magnetic fields to dynamically manipulate processing conditions in powder-fed DED as an adaptive control technique in cases where greater control of track dimensions or enhanced material feed control is required*[54]*. Martinez-Marchese et al. used an ultrasound phased array to produce a small volume of high-intensity ultrasound to acoustically focus and deflect particle streams exiting from a nozzle analog*[55]*, as well as demonstrating acoustic focusing within a working powder-fed DED machine, to increase catchment efficiency*[56]*.*" (please see page 3 in the revised manuscript)

Comment #4

In line 288, it says 30 m/s from the reported result. In the previous review I suggested using "reported result", but a more clear statement could be "measured result from the experiments in this article". I'm not sure why reference 24 is used, is this where the number actually came from or does it refer to how this number was measured? Please clarify what numbers where measured and simulated for this article.

Response

We are grateful for the reviewer's deep concern and inquiry about the particle velocity result. We apologize for the unclear data explanation here. As it is in a small and inaccessible acoustic focus area, the particle velocity in this area is calculated through simulation and is consistent with the relevant empirical values and the range described in the literature. Here, we have modified the data explanation and supplemented it with corresponding simulation result charts.

"*Here, the velocity is approximately 30 m/s, as determined from the aerosol particle jetting simulation (Fig. S2d), which falls within the normal ejection speed range*[60] *of 10-100 m/s, and the focusing time is established to be 1 ms from the simulated result.*" (please see page 9 in the revised manuscript)

"

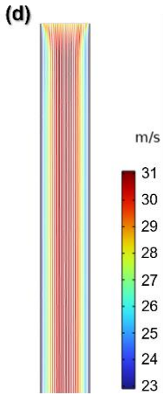


Fig. S1 *......* ***d*** *Particle trajectories and their velocities.* ***......*** " (please see page 3 in the revised supplementary information)

Comment #5

In line 533, what is the unit for the number 5 in the statement: "density distribution of order 5 at the inlet"? What is it meant by "order 5"?

Response

We appreciate the reviewer's careful review. We apologize for the unclear enunciating for particle release layout planning in the original manuscript, and the "order 5" here refers to the release distribution accuracy order of 5, and which has been revised in the revised manuscript.

"The quantity of particles emitted at the nozzle inlet is 100, positioned initially in accordance with a release distribution accuracy order of 5 at the inlet." (please see page 22 in the revised manuscript)

Comment #6

For Fig. S2, what are the units for Particle density? Is this particle concentration? Consider using "Particle concentration (# particles/mm^3)" for the y-axis. This will require an estimate of the mass flowrate, or could be stated as normalized with the flowrate. Also, how was this particle density computed? Note that if doing a 2D simulation of particles along a line (is this what is shown in Fig. S2 (e)?), one should take into account that the concentration for a fixed number of particles will change in proportion to the area of the concentric circles (this radial concentration calculation is like a weighted histogram) around the center of the tube where they fall. Reference 55 discusses this.

Response

We appreciate the reviewer's careful review of the article's expression and in-depth consideration of the research analysis. The "particle density" here actually refers to "particle concentration", and we have corrected the expression in the revised manuscript. In addition, here we only focused on the final printed deposition, so only focused on the particle concentration at the substrate. The specific research method was to collect the radial distance from the center of the tube of all particles that moved to the substrate in the simulation as shown in Fig. S2e, and the particle concentration shown in Fig. S2f was obtained by automatic processing of all radial distance data by commercial software Hiplot, which was stated as normalized. Regarding the research analysis method (Ref. 55) proposed by the reviewer, which take into account that the concentration for a fixed number of particles will change in proportion to the area of the concentric circles (this radial concentration calculation is like a weighted histogram) around the center of the tube where they fall, we have carefully learned it and will adopt it in future in-depth studies on the particle focusing process, and we thank this research suggestion from the reviewer again.

"*The particle concentration distribution, stated as normalized as shown in Fig. S2g, was derived from statistical analysis of the position information of all particles at the substrate in the simulation.*" (please see page 8 in the revised manuscript)

"

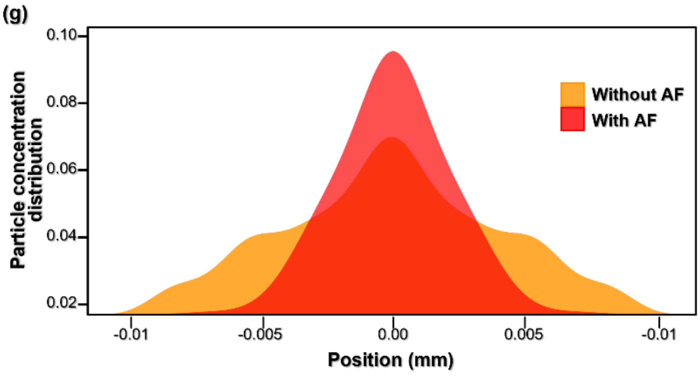


Fig. S2 ...... ***g*** *Particle concentration distribution of the AJ printed trace with/without AF.*" (please see page 3 in the revised supplementary information)

Comment #7

In line 289, it is noted that 2- and 3-dimensional track measurements are included in Fig. S6 g, h, however, no mention of how this is related to reference 60 is included. The reference can either be removed, or it might be best to move it to the conclusion section, along with a comment that for future work it could be shown that the track profile (Fig. S6 h) may be predicted from the particle concentration distribution in S2 f using the convolution model from reference 60.

Response

We thank the reviewer for the careful checking. We accept this opinion and have removed the reference cited here as suggested. We thank the reviewer again for the suggested methods for this study.

"*Moreover, the entire track geometries of the trace printed before, during, and after a short-term AF process, incorporating both two-dimensional and three-dimensional measurements, are supplemented in Fig. S6g, h.*" (please see page 11 in the revised manuscript)

Reviewer #3 (Remarks to the Author):

Impressive and interesting result. All concerns have been addressed.

Response

We appreciate your thorough review and valuable feedback throughout the revision process. Your insights have been instrumental in enhancing the quality of our manuscript. We are delighted to hear that our results are both impressive and interesting. We look forward to the possibility of our research being published in this esteemed journal.

Reviewer #4 (Remarks to the Author):

The authors have addressed several of my comments well. However, following major comments need to be addressed before the paper can be accepted in the journal:

Response

We appreciate the reviewer's positive feedback, careful review of our paper and the suggested methods. Through in-depth consideration and amelioration of the comments raised by the reviewer, the integrity and rigor of the research paper have been greatly enhanced. Once again, we thank the reviewer for his or her positive evaluation, thorough review, and helpful suggestions. We look forward to our paper being accepted by this prestigious journal!

Comment #1

The authors have not fully address comment #7 where no commercially available nozzle was compared. There was no explanation provided either.

Response

We thank the reviewer's in-depth attention and suggestions on our claim. We have supplemented the relevant information of the available nozzle as required, including the structure, outlet, and its limit printing effect, and compared the limit printing effect of the new nozzle with it, in order to fully and accurately explain the effectiveness of the new nozzle.

"*As shown in Fig. S3d, e, the investigation resulted in a LW of 10.13 μm and an overspray of 0.84 μm, which has completely broken through the limit printing accuracy and printing quality of the commercial nozzle with an outlet diameter of 260 μm (Fig. S3f-h), meeting the high-resolution requirement of AJ printing manufacturers.*" (please see page 9 in the revised manuscript)

"

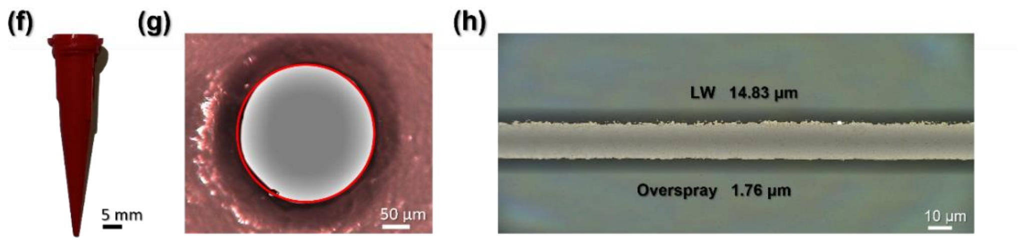


***Fig. S3 a** Geometric structure and design parameters of the new nozzle. **b** Optical image of the new nozzle. **c** Outlet of the stereolithography printed new nozzle. **d, e** Quantitative comparisons of the printed linewidth (d) and overspray width (e) under different printing parameters for the new nozzles with different structural parameters. **f** Optical image of the commercial nozzle. **g** Outlet of the available nozzle. **h** Geometric morphology of the available nozzle printed line.*" (please see page 4 in the revised supplementary information)

Comment #2

Comment #29, authors should use XRD or other methods to confirm that the particle sizes remain unchanged with and without AF (with XRD, the authors can use HWFM technique).

Response

We appreciate the reviewer's careful questioning and suggested methods. Regarding the question of whether the particle size before and after sintering was affected by AF, we have adopted the measurement methods suggested by the reviewer and used XRD to scan the depositions with and without AF before and after sintering, and calculated the corresponding silver particle sizes using HWFM technique. This provides concrete evidence and explanations to support the assertion that AF does not affect particle size.

"*It should be noted that the annealed particle size remains almost unchanged after undergoing the AF process, which indicates that the AF has no damage to the nanoparticles for AJ printing that requires sintering. As illustrated in Fig. S8a, b, the particle size was determined using Scherrer formula after scanning the AJ printed deposition with an X-ray diffractometer (XRD).*" (please see page 15 in the revised manuscript)

"*The particle size was calculated using Scherrer formula after scanning the AJ printed sample with an XRD.*" (please see page 23 in the revised manuscript)

"

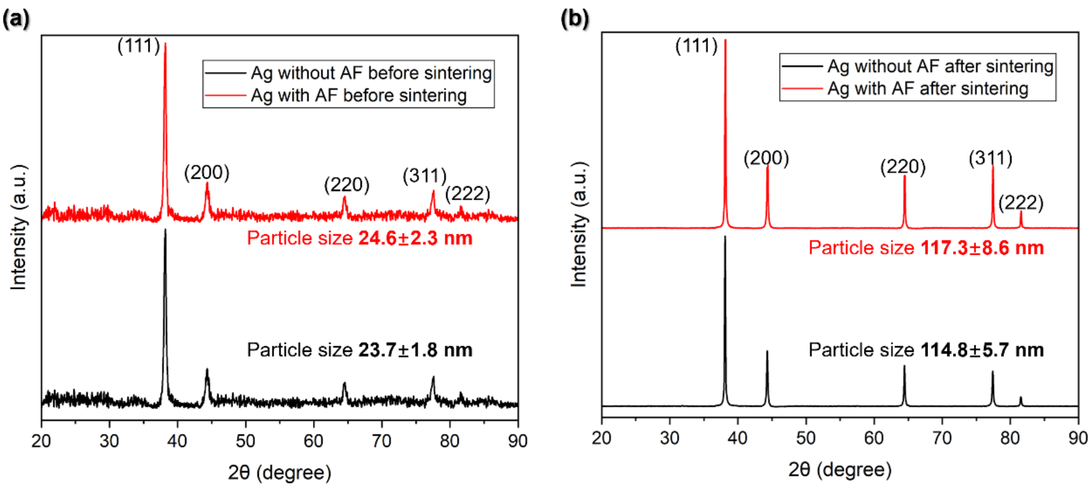


***Fig. S8** XRD spectra of Ag printed without and with AF **(a)** before and **(b)** after sintering respectively.*" (please see page 8 in the revised supplementary information)

Comment #3

Comment #31, authors should conduct cross-sectioning methods so that flat and comparable cross-sections can be achieved after polishing. The response did not address the reviewer's concerns.

Response

We appreciate the careful review by the reviewer on the experimental process and results of the cross-sections. For the porosity measurement, we have adopted the reviewer's cross-sectioning method to achieve new flat and comparable cross-sections of the original samples, and re-measured the cross-section and calculated the porosity. We would like to thank the reviewers again for his or her rigorous review and suggestion on cross-sectional fabrication and porosity measurement, which has made the research methods and results of the article more reliable.

"*By using pore analysis software, we analyzed the porosity of AJ printed cross-sections before and after*

A

F

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A

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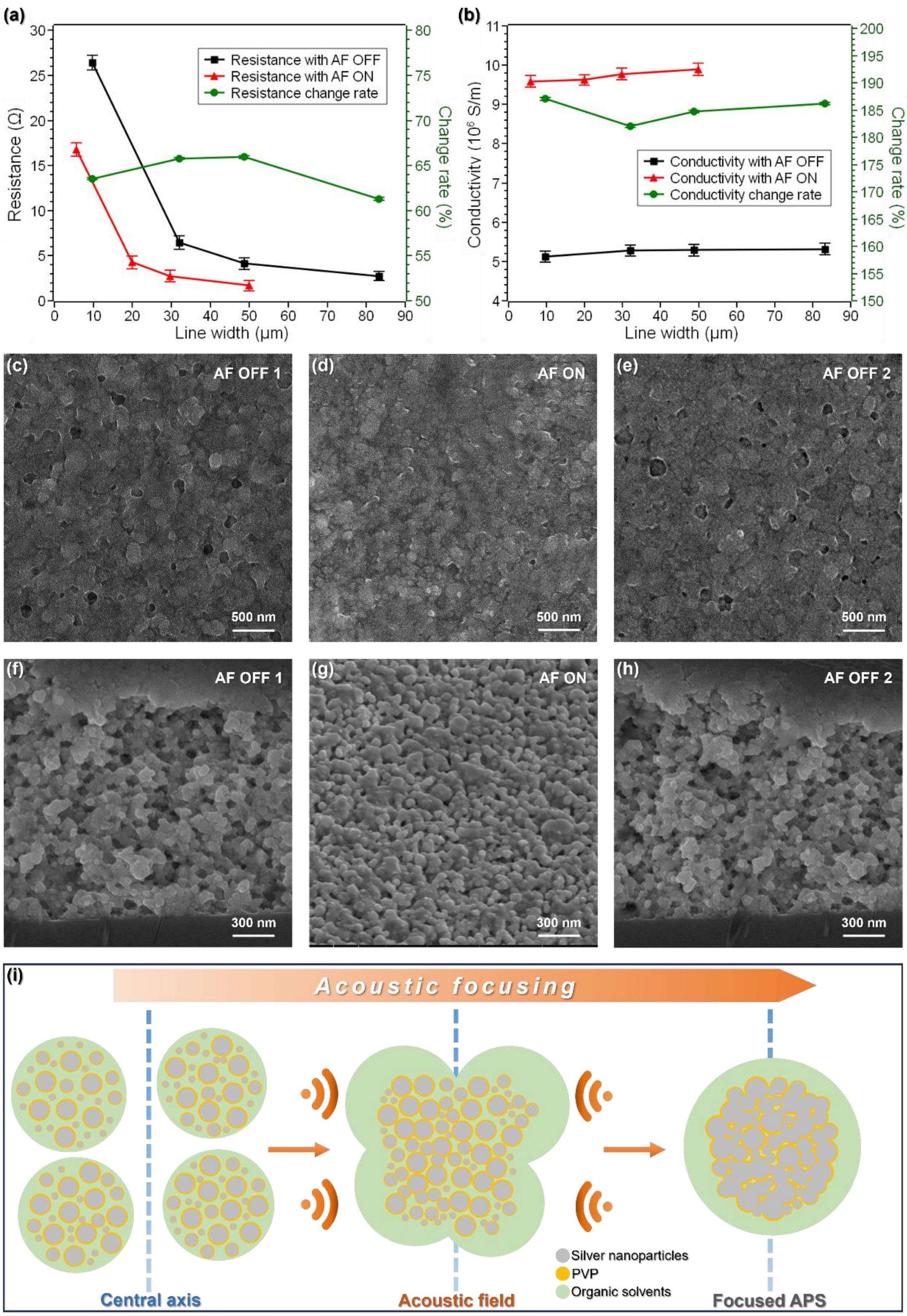

"

Fig. 4 Electrical properties and internal structure of AJ printed deposition under AF OFF1/ON/OFF2 state. …… i *Hypothesis schematic illustration showing the motion of APS during the AF process.*" (please see page 17 in the revised manuscript)

"***11 Porosity analysis***

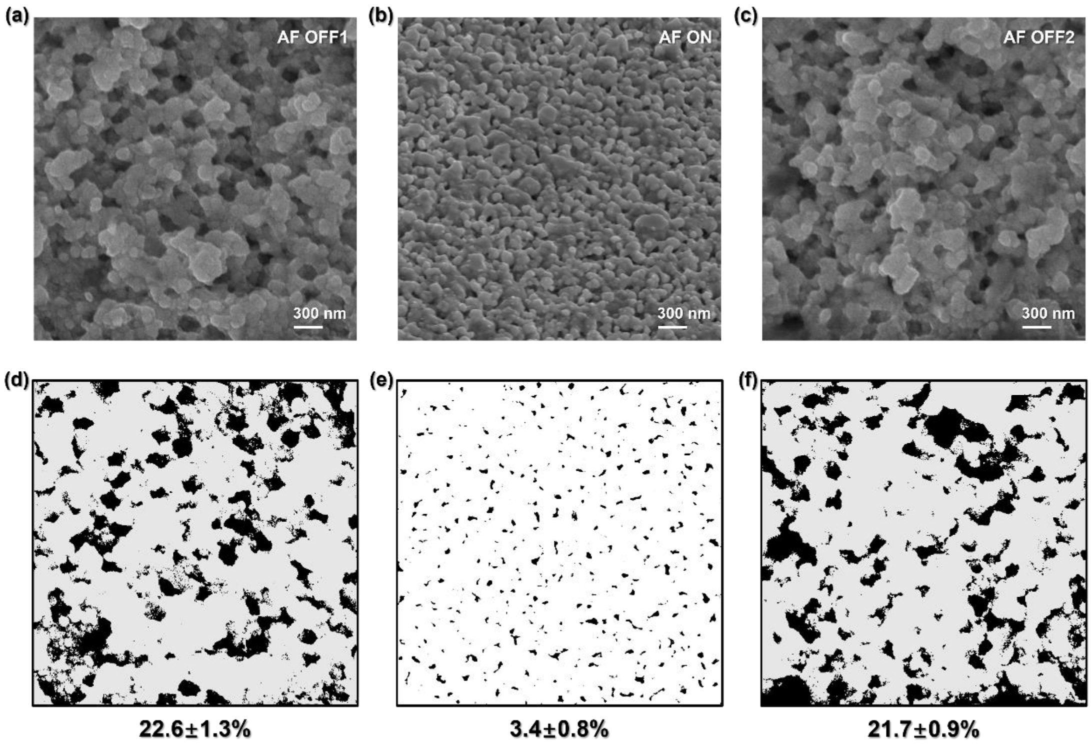

***Fig. S8** SEM images of the cross-sectional area of AJ printed trace **a** AF OFF1, **b** AF ON, **c** AF OFF2. **d, e, f** the calculated corresponding porosity.*" (please see page 8 in the revised supplementary information)

Comment #4

Comment #34, there is still no sufficient disclaimer indicating that Figure 4i is merely a hypothesis. No evidence has been provided to prove the nanoparticle focusing due to AF either.

Response

We appreciate the reviewer's careful examination and inquiries regarding the hypothesis (Fig. 4i). In response, we will explain and demonstrate it from the following two aspects. (1) As indicated by Comment #2, AF had no effect on particle size; (2) As explained by Comment #3, the particle distribution density in the deposition increased after the application of AF. Based on the above proof and analysis, we derived the hypothesis of the APS movement during the AF process shown in Fig. 4i. Here, we are willing to provide a sufficient disclaimer indicating that this is a hypothesis, and we support this hypothesis, committing to further confirm or explore the properties of this microstructure in future research. We thank the reviewer again for the rigorous review of this hypothesis.

"*As the hypothesis schematic drawing shown in Fig. 4i,* ……

***Fig. 4** …… **i** Hypothesis schematic illustration showing the motion of APS during the AF process.*" (please see page 17 in the revised manuscript)

REVIEWERS' COMMENTS

Reviewer #2 (Remarks to the Author):

All my previous revisions have been addressed.

Response to Reviewers of NCOMMS-23-48632B

We extend our heartfelt appreciation to the reviewers for their insightful comments and to the Editor for affording us the chance to enhance our manuscript (NCOMMS-23-48632A) entitled "Enhanced aerosol-jet printing using annular acoustic field for ultrahigh resolution and minimal overspray with improved conductivity" submitted to Nature Communications. After careful consideration of the feedback received, we agree that the manuscript has been substantially improved by integrating the recommended revisions. We have highlighted all revisions in red in the revised version. In the following sections, the reviewer's comments are in black, our responses to the comments are in blue and red indicates the text added/modified in the manuscript.

Reviewer #2 (Remarks to the Author):

All my previous revisions have been addressed.

Response

We appreciate your thorough review and valuable feedback throughout the revision process. We look forward to the possibility of our research being published in this esteemed journal.