

Orientation-dependent electronic structure in interfacial superconductors $\text{LaAlO}_3/\text{KTaO}_3$



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<http://creativecommons.org/licenses/by/4.0/>.

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

This paper tackles the important yet puzzling problem of orientation-dependent superconductivity in LaAlO₃/KTaO₃ interfaces. Superconductivity emerges up to 2K for certain orientations like (111), but is absent for (001) - an effect rarely seen in other superconductors. Resolving this phenomenon is key to unraveling the superconducting mechanism in complex oxides and enabling their engineering for next-generation devices. However, the microscopic origin of this orientation selectivity remains poorly understood, presenting a critical unsolved challenge.

To address this, the authors employ soft X-ray angle-resolved photoemission spectroscopy (SX-ARPES) to directly probe, for the first time, how the interfacial electronic structure in these oxide superconductors varies with orientation. This methodology enables access to the buried interface layers relevant for superconductivity.

Key findings include weak dimensional confinement and similar orbital characters across different orientations, arguing against selective orbital pairing theories. Instead, electron-phonon coupling between interfacial electrons and KTaO₃ substrate phonons exhibits orientation-dependent strength correlating with superconductivity.

In addition to detailed experimental results, the authors provide clear physical explanations and validations, making for a systematically complete analysis with ample supporting data. The paper represents a substantial advance in resolving an important problem through innovative application of SX-ARPES.

In summary, I recommend publication without reservation due to the thorough, well-reasoned analysis and sufficient supportive materials. This impactful work elucidates the orientational dependence mystery by directly measuring interfacial electronic states inaccessible until now. The new insights will aid engineering of multifunctional oxide heterostructures.

Reviewer #2 (Remarks to the Author):

The present paper reports experimental data on the interfacial electronic structure of LaAlO₃/KTaO₃ heterostructures. These heterostructures show superconductivity below critical temperatures that depend on the interface orientation: 1.22 K, 0.35 K and no superconductivity (or lower than 0.1 K) for (111)-, (110)- and (001)-oriented interfaces, respectively. The authors used soft-x-ray angle-resolved photoemission spectroscopy (SX-ARPES). All SX-ARPES measurements were done at 19 K, in the normal state.

The three main claims of the paper are:

i) The existence of three-dimensional-like, rather than the expected 2D-like, band structures for the three studied interfaces. This is deduced from the strong dispersion of the electronic

bands along the direction perpendicular to the interfaces.

ii) A similar orbital occupation (density of electrons in each of the t_{2g} -derived bands) for the three differently-oriented interfaces. This is deduced from an analysis of the photoemission selection rules following measurements with different photon polarisations, and is consistent (and expected) from the 3D-like band structure observed in the three interfaces.

iii) Different electron-phonon couplings for the differently-oriented interfaces, with the coupling constants λ being respectively $\lambda(111) > \lambda(110) > \lambda(001)$. The electron-phonon coupling strength is deduced from an analysis of the linewidths of the energy distribution curves for the three types of interfaces. The linewidths of the EDCs integrated over the whole range between Fermi momenta (WEDC) are respectively $WEDC(111) > WEDC(110) > WEDC(001)$.

The SX-ARPES data is of good quality, given the inherent constraints of the technique in terms of resolution, and the analyses are clearly explained. I find claims (i) and (ii) are well supported by the data, although I have concerns on how much the present SX-ARPES data represent the intrinsic electronic structure of the interfaces. On the other hand, I don't think claim (iii), which is the main claim of the paper, can be unequivocally inferred from the data. Therefore, I don't recommend publishing this paper in Nature Communications.

I summarise below my comments and remarks:

1) It is surprising that the three studied interfaces, which according to the magneto-transport data (Figure 1 and Figure S1) display very different characteristics even in the normal state (e.g., very different sheet resistances, different Hall magneto-resistances, different mobilities, different 2D carrier densities), show so similar electronic structures -the only slight difference being their linewidths. The irradiation with soft-X-rays seems not to be responsible of any modification of the interfacial electronic structure, as shown in figure S3. However, from that same figure, or from several other figures in the paper or supplementary information, one sees a clear peak of oxygen-vacancy states at around -2 eV binding energy. Are those oxygen vacancies created during the LAO deposition, hence intrinsic to the interfacial electronic structures, or are they rather created during the in-situ annealing to clean the surfaces before SX-ARPES experiments? In the second case, can the authors rule-out that an excessive creation of oxygen vacancies near the interface is rendering 3D-like the original interfacial, probably 2D-like, electron gas?

2) The sheet resistances RS and mobilities μ in the normal state are such that $RS(111) > RS(110) > RS(001)$ and $\mu(111) < \mu(110) < \mu(001)$. If the intrinsic electronic structures are 3D-like, as the paper claims, it seems surprising that transport is so much influenced by the interfacial orientations. In other words, why the interfacial orientation would affect an essentially bulk-like electronic structure? If this is due to different electron couplings to surface phonons, as the paper claims, then it is again surprising that Fuchs-Kliwer surface phonons, even at small q , have an effect on bulk-like states that is larger than the effect of bulk phonons, which should be the same and predominant for all 3D-like bands. Could it be that the different EDC linewidths are simply reflecting the different degrees of disorder at the three different interfaces? For instance, non-reconstructed KTO(111) surfaces are more

polar than KTO(110) surfaces, which in turn are more polar than KTO(001) surfaces, so interfacial reconstructions could lead to larger roughness and/or scattering potentials for, respectively, (111), (110) and (001) interfaces.

Reviewer #3 (Remarks to the Author):

I am also uploading the my reviewer comment as a PDF file.

Referee Report Ms#NCOMMS-23-45788, X. Chen et al.,

This manuscript reports the investigation of the crystallographic-orientation-dependent electronic structures at LaAlO₃/KTaO₃ interfaces by means of soft-x-ray angle-resolved photoemission spectroscopy (SX-ARPES). The superconductivity at the interface, which has been discovered recently, exhibits intriguing behaviors depending on crystallographic orientation. Understanding unique phenomena at the interface is a hot topic in condensed matter physics. The work gives valuable information to this hot field of oxide interface.

The main findings of this manuscript are 1) the mobile electrons that contribute to the interfacial superconductivity show strong k_z dispersion, indicating the existence of quasi-three-dimensional electron gas states in LaAlO₃/KTaO₃ interfaces, and 2) the orientation-dependent electron-phonon coupling correlates with the difference of superconducting temperatures.

The former claim is well substantiated by the state-of-the-art SXARPES results, where the clear k_z -dependence is demonstrated. However, the later claims are do not seem to be supported by their experimental data. The major problem is that the analysis relies on only a tail-like spectra weight without the peak-dip-hump structures, shown in Fig. 4(h-j). Specially,

1) In Frank-Condon model analysis, it is common to perform the analysis for ARPES spectra (energy distribution curves; EDCs) at the Fermi momentum (k_F), where quasiparticle peak is well defined, and the contributions from other EDCs at different k points are eliminated [Ref.8 and S. Moser et al., Phys. Rev. Lett. 110, 196403 (2013)]. Instead, the authors used momentum-integrated EDC (integrated ARPES intensity shown in Figs. 4b-d). In that case, the analysis of line shape is not straightforward since the renormalized main band and its replica (blue parabolas shown in Figs. 4e-g) have a considerable dispersion in the range of $\sim 0.3\text{\AA}^{-1}$ – $0.3\text{\AA}^{-1}$ and strong matrix-element effect. The authors should conduct a line-shape analysis with EDC at k_F .

2) The tail-like spectra weight is strongly influenced by the background subtraction method, as demonstrated in Supplementary Information. Thus, the analysis relying only on the spectral tail is questionable.

3) In addition, for the analysis of the momentum-integrated EDC, the width of the peak at E_F reflects the density of states (DOS) of the Ta 5d conduction band. Therefore, the carrier

concentration must be constant for three orientations to make fair comparisons. However, the number of carriers is different among orientations. The authors should present the results of the momentum-integrated EDC (Fig. 2b) also for (001) and (111) and provide evidence that there is no change in Ta-5d derived DOS at EF irrespective of carrier concentrations.

Minor comments:

A) To show a closed Fermi surface, the authors should show the MDC outside the Fermi surface (add the MDC outside the Fermi surface in Fig. 2e, Fig. S4C, and Figs. S5D&H, and Figs. S6D,E,I,J,N,O&P). Such a presentation will be helpful for the reader to ascertain its validity.

B) The background intensity at -0.8 eV is always less than half in the three-dimensional plots of ARPES spectra shown in Fig. 4(b-d). However, in their momentum-integrated EDC data shown in Fig. 4(h-i), the background intensity is well over half. Please explain this discrepancy.

The problems are that the peak-dip-hump structure, which is the evidence of electron-phonon coupling, has not been observed, and the conclusion relying on complicated data analysis is firmly model-dependent. If the present experimental results showed impeccable evidence for the interpretation and conclusion that the authors propose, the study would certainly be of great interest to the community of condensed-matter physics.

The responses to the reviewers' comments are listed as follows. The major modifications in the main text are highlighted in red color.

===== Authors' responses to the reviewers' comments =====

Response to the comment of Reviewer #1:

Reviewer #1 (Remarks to the Author):

This paper tackles the important yet puzzling problem of orientation-dependent superconductivity in LaAlO₃/KTaO₃ interfaces. Superconductivity emerges up to 2K for certain orientations like (111), but is absent for (001) - an effect rarely seen in other superconductors. Resolving this phenomenon is key to unraveling the superconducting mechanism in complex oxides and enabling their engineering for next-generation devices. However, the microscopic origin of this orientation selectivity remains poorly understood, presenting a critical unsolved challenge.

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In addition to detailed experimental results, the authors provide clear physical explanations and validations, making for a systematically complete analysis with ample supporting data. The paper represents a substantial advance in resolving an important problem through innovative application of SX-ARPES.

In summary, I recommend publication without reservation due to the thorough, well-reasoned analysis and sufficient supportive materials. This impactful work elucidates the orientational dependence mystery by directly measuring interfacial electronic states inaccessible until now. The new insights will aid engineering of multifunctional oxide heterostructures.

Reply (1): We thank Reviewer #1 for his/her careful review of our manuscript and for accurately summarizing our key findings. We sincerely appreciate his/her recognition of the significance of our work and strong recommendations.

Response to the comment of Reviewer #2:

Reviewer #2 (Remarks to the Author):

The present paper reports experimental data on the interfacial electronic structure of

LaAlO₃/KTaO₃ heterostructures. These heterostructures show superconductivity below critical temperatures that depend on the interface orientation: 1.22 K, 0.35 K and no superconductivity (or lower than 0.1 K) for (111)-, (110)- and (001)-oriented interfaces, respectively. The authors used soft-x-ray angle-resolved photoemission spectroscopy (SX-ARPES). All SX-ARPES measurements were done at 19 K, in the normal state.

The three main claims of the paper are:

- i) The existence of three-dimensional-like, rather than the expected 2D-like, band structures for the three studied interfaces. This is deduced from the strong dispersion of the electronic bands along the direction perpendicular to the interfaces.
- ii) A similar orbital occupation (density of electrons in each of the t_{2g}-derived bands) for the three differently-oriented interfaces. This is deduced from an analysis of the photoemission selection rules following measurements with different photon polarisations, and is consistent (and expected) from the 3D-like band structure observed in the three interfaces.
- iii) Different electron-phonon couplings for the differently-oriented interfaces, with the coupling constants λ being respectively $\lambda(111) > \lambda(110) > \lambda(001)$. The electron-phonon coupling strength is deduced from an analysis of the linewidths of the energy distribution curves for the three types of interfaces. The linewidths of the EDCs integrated over the whole range between Fermi momenta (WEDC) are respectively $WEDC(111) > WEDC(110) > WEDC(001)$.

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Reply (2): The Reviewer makes excellent suggestions for improving the focus of the manuscript. We follow these suggestions:

- We put more emphasis on claims (i) and (ii), which are well supported by the data, in our revised manuscript.
- We have revised the "orientation-dependent electron-phonon coupling" in the title of our manuscript to "orientation-dependent electronic structure", to emphasize the direct experimental findings.
- We have explicitly stated that the EPC scenario is a potential candidate to elucidate the orientation-dependent superconductivity.

In the following Reply (3) and Reply (4), we discussed the concern of whether the present data represents the intrinsic electronic structure and that claim (iii) is a candidate scenario to explain the data.

I summarise below my comments and remarks:

1) It is surprising that the three studied interfaces, which according to the magneto-transport data (Figure 1 and Figure S1) display very different characteristics even in the normal state (e.g., very different sheet resistances, different Hall magneto-resistances, different mobilities, different 2D carrier densities), show so similar electronic structures -the only slight difference being their linewidths. The irradiation with soft-X-rays seems not to be responsible of any modification of the interfacial electronic structure, as shown in figure S3. However, from that same figure, or from several other figures in the paper or supplementary information, one sees a clear peak of oxygen-vacancy states at around -2 eV binding energy. Are those oxygen vacancies created during the LAO deposition, hence intrinsic to the interfacial electronic structures, or are they rather created during the in-situ annealing to clean the surfaces before SX-ARPES experiments? In the second case, can the authors rule-out that an excessive creation of oxygen vacancies near the interface is rendering 3D-like the original interfacial, probably 2D-like, electron gas?

Reply (3): We thank the reviewer for the insightful comments.

As highlighted by the Reviewer, the presence of a valence band at -2 eV suggests the existence of oxygen vacancy states in our samples. It is reported that LAO deposition would induce oxygen vacancies, and these states are intrinsic to the interfacial electronic structures. Our LAO/KTO were grown at low oxygen pressures (1×10^{-5} mbar) and high temperatures (650 ~ 680 °C) by PLD, which is consistent with previous reports, and such conditions are shown to induce the formation of oxygen vacancies [Y. Sun *et al. Phys. Rev. Lett.* **127**, 086804 (2021)].

It's noteworthy that the in-situ annealing to clean the surface before SX-ARPES measurements were conducted at ~ 250 °C. According to prior research [F. Y. Bruno *et al., Adv. Electron. Mater.* **5**, 1800860 (2019)], the creation of oxygen vacancies in KTO typically occurs above 450 °C. Therefore, the formation of oxygen vacancies during the 250 °C annealing process should be negligible. There should be no additional oxygen vacancies prior to the ARPES measurements through the in-situ annealing process.

We agree with the reviewer that the transport data of differently orientated samples display very different characteristics in the normal state. In many cases, the changes in transport behavior, like the resistance and mobility, do not directly correspond to differences in band dispersion but reflect the differences in the lifetimes of quasiparticles. For example, in the underdoped-, optimal-doped- and overdoped-cuprate, though their resistance changes a lot, the Fermi surface and dispersion change only slightly [I. K. Drozdov *et al. Nat. Commun.* **9**, 5210 (2018); D. Pelc *et al. Phys. Rev. B* **102**, 075114 (2020)]. In the case of LAO/KTO, the electron-phonon coupling can shorten the lifetime of the quasiparticle, thereby increasing the normal state resistivity. The spectral tail and the quasiparticle lifetime likely contribute to the differences in normal state resistance and mobility.

Relevant changes:

- In the revised manuscript, we have added a discussion (lines 87-90) on the -2 eV shoulder and

the origin of the oxygen vacancies origin:

"The momentum-integrated energy distribution curve (EDC) of LAO/KTO(111) shows a peak near E_F , accompanied by a shoulder around -2 eV attributed to oxygen vacancy states [34] (Fig. 2b). These vacancies are reported to arise during LAO deposition and are intrinsic to the interfacial state [29]."

- We have added a reference to address that low-temperature annealing is not expected to generate additional oxygen vacancies (lines 276-278):

"Samples were heated to 250 °C for half an hour before ARPES measurements to remove the surface adsorbates owing to air exposure, and such low-temperature annealing is not expected to induce oxygen vacancies in KTO [37]."

- We have added a discussion on the different lifetime and the different transport behavior (lines 223-227):

"The electron-phonon coupling can shorten the lifetime of the quasiparticle, thereby increasing the normal state resistivity and reducing the mobility, which likely explains the differences in normal state resistance and mobility in differently-orientated LAO/KTO with $R_{\text{sheet}}(111) > R_{\text{sheet}}(110) > R_{\text{sheet}}(001)$ (Fig. 1b) and $\mu(111) < \mu(110) < \mu(001)$ (Fig. 1d)".

2) The sheet resistances RS and mobilities μ in the normal state are such that $RS(111) > RS(110) > RS(001)$ and $\mu(111) \ll \mu(110) \ll \mu(001)$. If the intrinsic electronic structures are 3D-like, as the paper claims, it seems surprising that transport is so much influenced by the interfacial orientations. In other words, why the interfacial orientation would affect an essentially bulk-like electronic structure? If this is due to different electron couplings to surface phonons, as the paper claims, then it is again surprising that Fuchs-Kliwer surface phonons, even at small q , have an effect on bulk-like states that is larger than the effect of bulk phonons, which should be the same and predominant for all 3D-like bands. Could it be that the different EDC linewidths are simply reflecting the different degrees of disorder at the three different interfaces? For instance, non-reconstructed KTO(111) surfaces are more polar than KTO(110) surfaces, which in turn are more polar than KTO(001) surfaces, so interfacial reconstructions could lead to larger roughness and/or scattering potentials for, respectively, (111), (110) and (001) interfaces.

Reply (4): We thank the Reviewer for the valuable comments.

First, we appreciate the reviewer's comment on the possible contribution from the different disorder levels at the three interfaces, which needs to be discussed. When a photoelectron is scattered by a local static disorder, either elastic or inelastic, it will lose its momentum resolution in the ARPES spectra. According to the theory and experimental verification by SX-ARPES, disorder can effectively reduce the spectral weight of dispersive feature (I_{coh}), and convert it into an incoherent non-dispersive background (I_{inc}) [Shevchik, N. J. *Phys. Rev. B* **16**(8), 3428 (1977). and Della Valle, E. *et al.* arXiv preprint arXiv:2308.03707 (2023)] while the effect of increasing the energy span is relatively minor. As shown by previous SX-ARPES on InAs (Fig. R1), when the increasing disorder density suppresses the I_{coh} by 70%, the EDC peak shows no detectable broadening in the energy scale. Therefore, the difference in spectral weight tails of LAO/KTO is not likely caused by different degrees of disorder. Alternatively, disorders can broaden the photoemission spectra of core levels by varying the local chemical potential [Newton, A. W. *et al. Journal of Electron Spectroscopy and*

Related Phenomena **107**(2), 185 (2000)]. However, to form dispersive features near Fermi energy, the domain size of a coherently doped region should reach tens of nanometers. As STEM measurements show homogeneous and amorphous LAO layer at LAO/KTO samples [Chen, Z. *et al. Science* **372**(6543), 721 (2021).], inhomogeneous dopings in mesoscopic scale is not likely to occur. Therefore, although we could not exclude disorder difference in our samples, it is not likely the origin of the spectral weight tail. Moreover, it is shown that the disorder is harmful to superconductivity in LAO/KTO, rather than enhancing it [Chen, Z. *et al. Science* **372**(6543), 721 (2021).]. If the larger EDC linewidth of 111 orientation is caused by higher degrees of disorder, it is not consistent with its higher superconducting T_c .

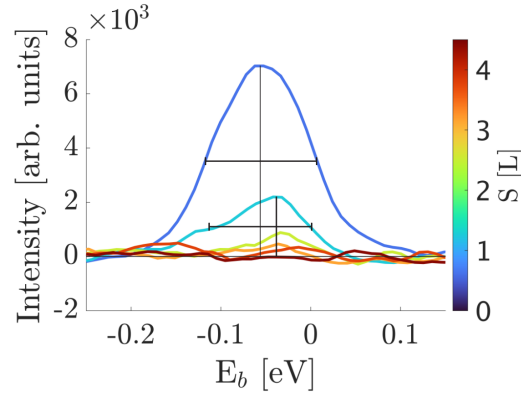


Fig. R1 EDCs of the dispersive electron Fermi surface in InAs as a function of increasing static disorders induced by sputtering dose S . This panel is reproduced from [Della Valle, E. *et al. arXiv preprint arXiv:2308.03707* (2023)]. The horizontal bars illustrate the FWHM of EDC peaks.

Second, we thank the Reviewer's insightful comments on the difference in polar strengths among the three interfaces which needs to be discussed. We agree with the Reviewer that "KTO(111) surfaces are more polar than KTO(110) surfaces, which in turn are more polar than KTO(001) surfaces". The different surface polar and the lattice relaxation can modify the Fuchs-Kliwer (FK) phonons and local EPC, which might play roles in the orientation-dependent electronic structure and superconductivity. We have added this to the revised manuscript.

Regarding the roles of FK phonons and bulk phonons, it's worth noting that FK phonons are known for their long-range nature, especially at small q . The quasi-3D interfacial states have a thickness around 5 ~ 7 nm, a thickness that can induce strong k_z dispersion [S. Moser *et al. J. Electron. Spectrosc. Relat. Phenom.* **225**, 16–22 (2018)]. This thickness of the interfacial states spatially overlaps with that of FK surface phonons, which allows a strong interaction between electrons and FK phonons. Concerning the bulk phonons, as the electron-doped KTO bulk crystal is not superconducting according to previous reports (refs. 59 and 60 in our manuscript) and it's not expected to show orientation dependence, the electrons coupling only with the bulk phonons is not sufficient to account for the orientation-dependent superconductivity. Therefore, we propose the FK surface phonon modes as the most probable candidate. We have made these conjectures clearer in the revised manuscript.

Relevant changes:

- In the discussion part (lines 197-199), we have added a sentence:
"In SX-ARPES, the dominant effect of disorder scatterings is converting the coherent dispersive spectra weight into the incoherent non-dispersive background, while the energy broadening of spectral function is relatively minor [47]. " to clarify the effect of disorder in SX-ARPES.
- In the discussion part (lines 244-246), we have added a sentence:
"...for instance, the different polar strength among the three interfaces and the induced lattice relaxation may modify the Fuchs-Kliwer modes and their coupling with the electrons" to discuss the possible role of different surface polar at differently oriented surfaces/interfaces.
- In the discussion part (lines 232-234), we modified the discussion on doped bulk KTO:
"Given that superconductivity is absent in chemically doped bulk KTO [58, 59], EPC with the bulk phonons, if present, should be less crucial for the superconductivity, while further investigations are required to understand this" to clarify the role of bulk phonons.

Response to the comment of Reviewer #3:

Reviewer #3 (Remarks to the Author):

I am also uploading the my reviewer comment as a PDF file.

Referee Report Ms#NCOMMS-23-45788, X. Chen et al.,

This manuscript reports the investigation of the crystallographic-orientation-dependent electronic structures at LaAlO₃/KTaO₃ interfaces by means of soft-x-ray angle-resolved photoemission spectroscopy (SX-ARPES). The superconductivity at the interface, which has been discovered recently, exhibits intriguing behaviors depending on crystallographic orientation. Understanding unique phenomena at the interface is a hot topic in condensed matter physics. The work gives valuable information to this hot field of oxide interface.

Reply (5): We thank Reviewer #3 for highlighting our work's importance.

The main findings of this manuscript are 1) the mobile electrons that contribute to the interfacial superconductivity show strong k_z dispersion, indicating the existence of quasi-three-dimensional electron gas states in LaAlO₃/KTaO₃ interfaces, and 2) the orientation-dependent electron-phonon coupling correlates with the difference of superconducting temperatures.

The former claim is well substantiated by the state-of-the-art SXARPES results, where the clear k_z -dependence is demonstrated.

Reply (6): We thank the reviewer for pointing out our main findings, and one of our main findings is "well substantiated by the state-of-the-art SXARPES results". Following the Reviewer's excellent suggestions for improving the focus of the manuscript. We have revised our manuscript as follows:

- We put more emphasis on claim 1), which is well supported by the data, in our revised

manuscript.

- We have revised the “orientation-dependent electron-phonon coupling” in the title of our manuscript to "orientation-dependent electronic structure", to emphasize the direct experimental findings.
- We have explicitly stated that the EPC scenario is a potential candidate to elucidate the orientation-dependent superconductivity.

However, the later claims are do not seem to be supported by their experimental data. The major problem is that the analysis relies on only a tail-like spectra weight without the peak-dip-hump structures, shown in Fig. 4(h-j). Specially,

1) In Frank-Condon model analysis, it is common to perform the analysis for ARPES spectra (energy distribution curves; EDCs) at the Fermi momentum (k_F), where quasiparticle peak is well defined, and the contributions from other EDCs at different k points are eliminated [Ref.8 and S. Moser et al., Phys. Rev. Lett. 110, 196403 (2013)]. Instead, the authors used momentum-integrated EDC (integrated ARPES intensity shown in Figs. 4b-d). In that case, the analysis of line shape is not straightforward since the renormalized main band and its replica (blue parabolas shown in Figs. 4e-g) have a considerable dispersion in the range of $-0.3\text{\AA}^{-1}$ – $0.3\text{\AA}^{-1}$ and strong matrix-element effect. The authors should conduct a line-shape analysis with EDC at k_F .

Reply (7): We thank the reviewer’s insightful suggestions.

Due to the utilization of high photon energy, the in-plane momentum resolution is limited, with $\Delta k \sim 0.02\text{\AA}^{-1}$, which is poor as compared to the small values of k_F in the order of $0.1\text{\AA}^{-1}$. Consequently, the differences between EDCs at k_F s and those integrated over the electron pocket region are not significant (Fig. R2). As a result, in our last manuscript submission, the integrated EDCs were analyzed to improve the signal-to-noise ratio.

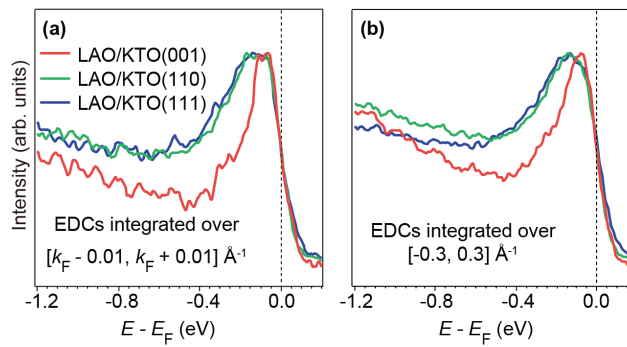


Fig. R2 (a) EDCs of LAO/KTO(001), (110) and (111) at the corresponding k_F . (b) EDCs of LAO/KTO(001), (110) and (111) integrated over $[-0.3, 0.3]\text{\AA}^{-1}$.

Following the reviewer's suggestion, here we analyze the EDCs at k_F . As shown in Fig. R3(d), the EDC linewidth at k_F s also follows $111 > 110 \gg 001$. We fit the EDCs with a series of Gaussian peaks whose intensities follow the Poisson distribution. As shown in Fig. R3(e-g), the fitted λ also follows $\lambda(111) > \lambda(110) > \lambda(001)$. We thank the Reviewer for pointing this out and we have added

these results into the revised manuscript.

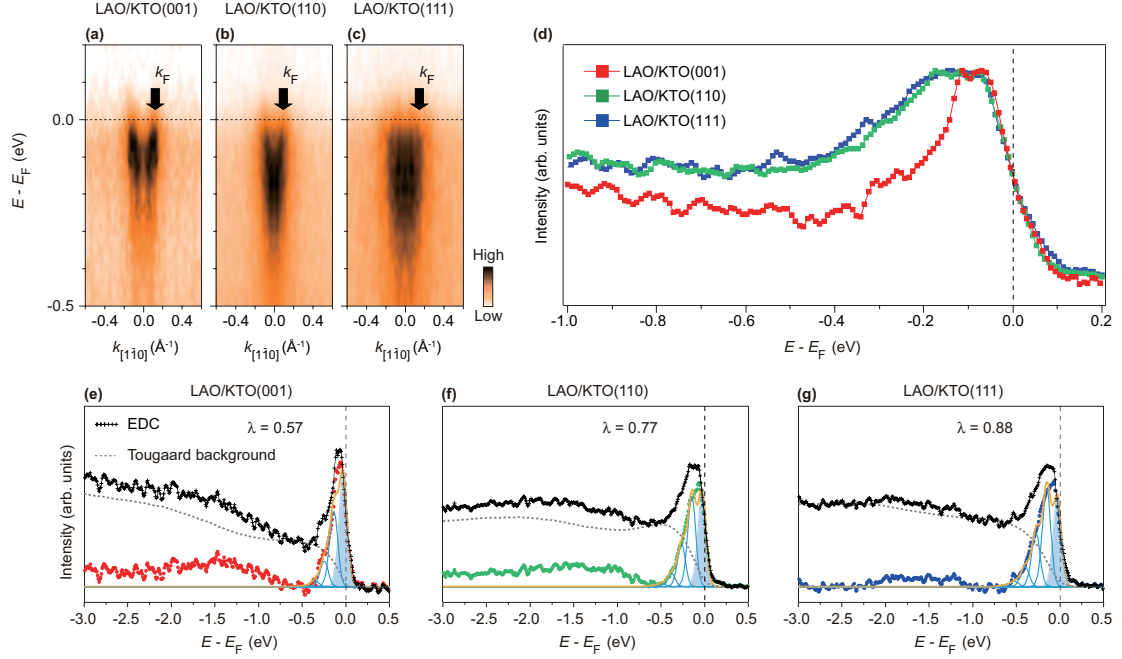


Fig. R3 (a-c) ARPES spectra measured along $k_{[1-10]}$ direction of LAO/KTO(001), (110) and (111), respectively. (d) EDCs of LAO/KTO(001), (110), and (111) at the corresponding k_F indicated in (a). (e-g) Franck-Condon fitting results of EDCs in (d). The Tougaard backgrounds are used in these fittings. The main band and replicas are represented by a series of Gaussian peaks whose FWHM are set as the energy resolution in ARPES measurements.

Relevant changes:

- We have added a line-shape analysis with EDC at k_F to the revised manuscript as Figs. 4k-4m.

2) The tail-like spectra weight is strongly influenced by the background subtraction method, as demonstrated in Supplementary Information. Thus, the analysis relying only on the spectral tail is questionable.

Reply (8): We thank the Reviewer for this insightful comment. While we fully agree with the referee that a more convincing analysis should be conducted on spectra with peak-dip-hump structures, it is unfortunate that SX-ARPES with approximately 1000 eV photons lacks the resolution required to observe such features (Supplementary Fig. S14). Note that the KTO surface under electric gating shows superconductivity similar to LAO/KTO, and KTO surface states, which are surface-doped KTO similar to the interfacial states, can be studied by surface-sensitive VUV-ARPES with much higher resolution. Therefore, here we performed additional experiments on KTO surface mobile electrons as a complement to our analysis on LAO/KTO. As shown in Fig. R4, KTO(110) shows the peak-dip-hump (PDH) lineshape with an energy separation of around 100 meV, close to the energy of the LO4-derived Fuchs-Kliever surface phonon mode. In a previous ARPES study on the KTO(111) surface 2DEG [C. Bareille *et al. Scientific Reports* **4**, 3586 (2014)], a prominent PDH structure is also observed. These facts emphasize the general presence of EPC with ~ 100 meV phonons at KTO (110) and (111) surfaces, which can be superconducting under electric gating.

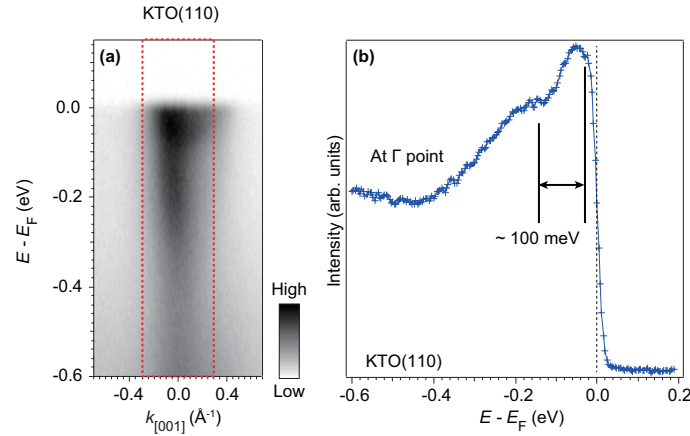


Fig. R4 (a) ARPES spectra of KTO(110) surface 2DEG. The spectra are measured using 63 eV photons at 11 K and the total energy resolution is ~ 30 meV. **(b)** EDC integrated over $[-0.3, 0.3] \text{ \AA}^{-1}$ around the Γ point of KTO(110) surface 2DEG, the integrated region is indicated as a dashed red box in (a). A peak-dip-hump structure is resolved with an energy separation of ~ 100 meV.

Relevant changes:

- Added the discussion on PDH to the Discussion part of the revised manuscript, and addressed the assumption before analyzing the EPC of LAO/KTO in a more explicit way (lines 205-213):
 "...Due to the insufficient resolution of SX-ARPES with ~ 1000 eV photons, the peak-dip-hump structure with the separated main band and replicas by phonon energy could not be resolved (Supplementary Note 9). Note that superconductivity has recently been achieved at the KTO surfaces under electric gating, displaying a similar orientation dependence as the interface [50, 51]. Therefore, as a supplement, we studied the spectral weight tail on KTO(110) surface using VUV-ARPES with much better energy resolution, where the peak-dip-hump structure can indeed be resolved, showing an energy separation between the peak and hump around 100 meV (Supplementary Fig. 15). Assuming that the spectral weight tail observed in LAO/KTO is the combination of quasiparticle peaks and their shake-off replicas by 100 meV phonons,..."
- Added Fig. R4 as Supplementary Fig. 15 to show the presence of PDH at the KTO surface.

3) In addition, for the analysis of the momentum-integrated EDC, the width of the peak at E_F reflects the density of states (DOS) of the Ta 5d conduction band. Therefore, the carrier concentration must be constant for three orientations to make fair comparisons. However, the number of carriers is different among orientations. The authors should present the results of the momentum-integrated EDC (Fig. 2b) also for (001) and (111) and provide evidence that there is no change in Ta-5d derived DOS at E_F irrespective of carrier concentrations.

Reply (9): Following Reviewer's suggestion, here we display the large energy scale valence band of LAO/KTO(111), (110) and (001) and their zoomed-in view near the E_F in Fig. R5. W_0 varies from 169 meV, 128 meV, to 174 meV for (111), (110), and (001) orientations, respectively, which does not match the orientation dependence of the EDC linewidth. Therefore, although the difference in carrier concentration leads to slight variations in band filling and DOS, it cannot explain the tail

behavior of EDCs. In our Franck-Condon fittings, we have taken into account the different bandwidth (W_0 in Fig. R5(b)), caused by their doping discrepancies.

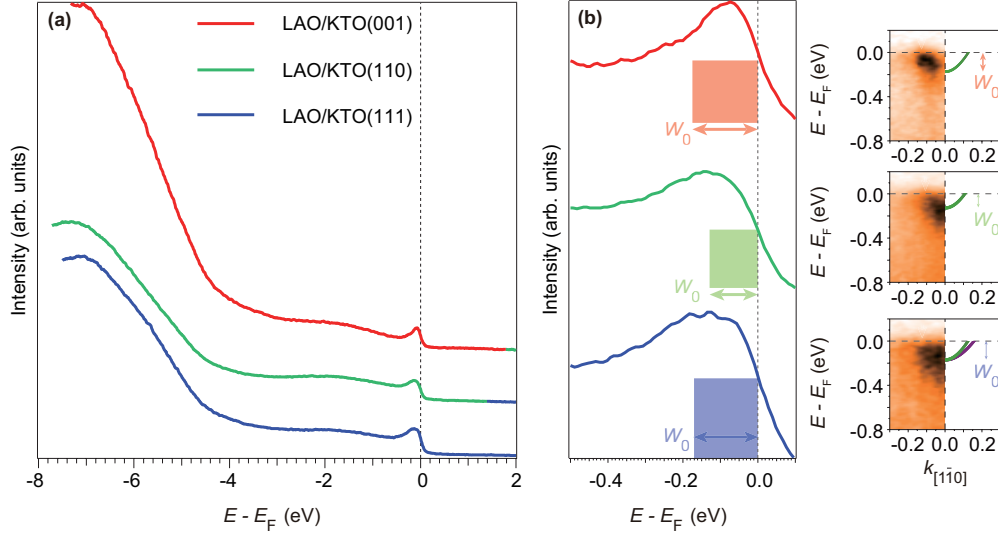


Fig. R5 (a) The momentum-integrated EDCs of LAO/KTO(111), (110) and (001). The integrated region is $[-0.3, 0.3] \text{ \AA}^{-1}$. **(b)** Zoomed-in view near the E_F of the momentum-integrated EDCs. W_0 is the bare bandwidth obtained by aligning experimental k_F with the DFT calculations, as shown on the right side of (b).

Relevant changes:

- We have added the results of the momentum-integrated EDC for (001) and (110) as Supplementary Fig. 13.
- In the revised manuscript, we have added the discussion on band filling (lines 216-218):
"Note that the slight variations in band filling and density of states attributing to different carrier concentrations have already been considered in the fitting (see details in Supplementary Note 8 and Supplementary Note 13)."

Minor comments:

A) To show a closed Fermi surface, the authors should show the MDC outside the Fermi surface (add the MDC outside the Fermi surface in Fig. 2e, Fig. S4C, and Figs. S5D&H, and Figs. S6D,E,I,J,N,O&P). Such a presentation will be helpful for the reader to ascertain its validity.

Reply (10): The Reviewer makes an excellent suggestion for improving the data presentation. Following the reviewer's suggestion, we have shown the MDC outside the Fermi surface in the revised manuscript and display the corresponding panels here (Fig. R6). It allows for a better display of the electrons pocket along k_z direction by adding extra MDCs.

Relevant changes:

We have updated the corresponding figures in the revised manuscript and added the MDC outside the Fermi surface at the k_z -dispersive Fermi pocket of (111) (Fig. 2e, Supplementary Fig. 4C), (110) (Supplementary Fig. 5D), and (001) orientations (Supplementary Fig. 5H).

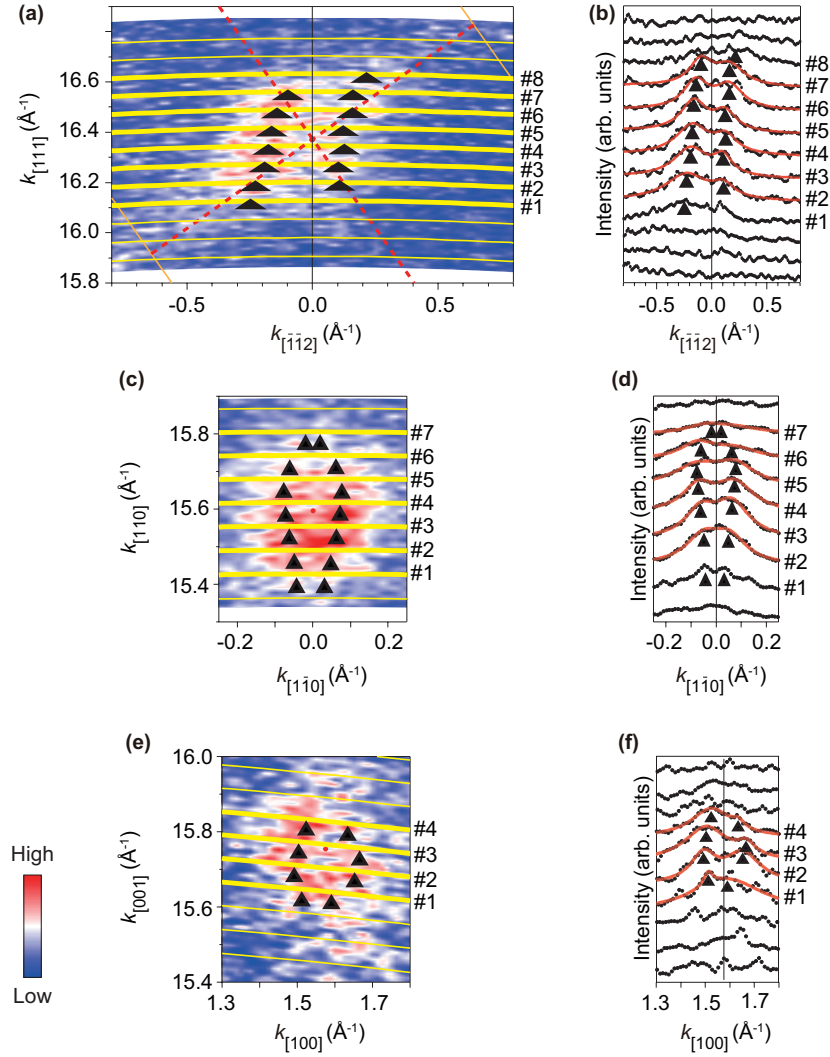


Fig. R6 The closed Fermi surface along k_z of LAO/KTO(111), (110) and (001). The MDCs at different k_z and corresponding Lorentzian fittings are displayed on the right side.

B) The background intensity at -0.8 eV is always less than half in the three-dimensional plots of ARPES spectra shown in Fig. 4(b-d). However, in their momentum-integrated EDC data shown in Fig. 4(h-i), the background intensity is well over half. Please explain this discrepancy.

Reply (11): Yes, we agree that the relative spectral weight at -0.8 eV to the maximum intensity is higher after momentum integration, which is as expected in ARPES data. As shown in Figs. R7(a) and (c), the highest intensity in the three-dimensional plots of ARPES around -0.2 eV (denoted as $I_{\max}$) is more than twice the intensity at -0.8 eV (denoted as $I(-0.8 \text{ eV})$). When these fifty individual EDCs are added together as shown in Fig. R7(b), because the feature around -0.2 eV is highly dispersive, the maximum intensity in the integrated EDC is much less than $50 \times I_{\max}$, while the intensity at -0.8 eV, with mostly non-dispersive background, is about $50 \times I(-0.8 \text{ eV})$. Consequently, the relative spectral weight at -0.8 eV to the maximum intensity is higher after integration [Fig. R7(b)].

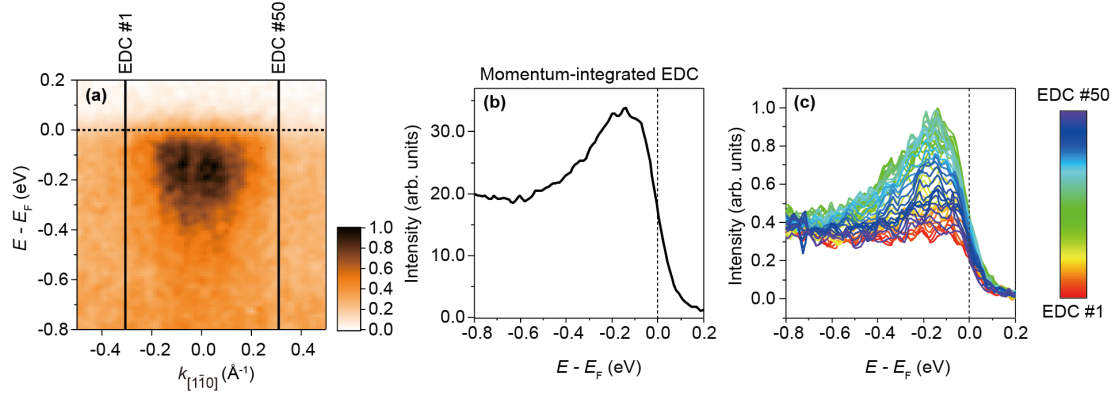


Fig. R7 (a) ARPES spectra of LAO/KTO(111) along $k_{[1-10]}$ direction. (b) The momentum-integrated EDC of LAO/KTO(111) integrated over $[-0.3, 0.3] \text{ \AA}^{-1}$, which is equivalent to the sum of fifty individual EDCs with a momentum step of $0.0126 \text{ \AA}^{-1}$. (c) The fifty individual EDCs in the momentum range $[-0.3, 0.3] \text{ \AA}^{-1}$ as shown in (a), and their integration is the EDC present in (b).

The problems are that the peak-dip-hump structure, which is the evidence of electron-phonon coupling, has not been observed, and the conclusion relying on complicated data analysis is firmly model-dependent. If the present experimental results showed impeccable evidence for the interpretation and conclusion that the authors propose, the study would certainly be of great interest to the community of condensed-matter physics.

Reply (12): We thank the Reviewer's comments. The mysterious orientation-dependent superconductivity in LAO/KTO is a complex problem, involving so many factors that may lead to it. In our work, we have provided solid evidence on the quasi-three-dimensional electronic structure and similar orbital occupancy of these novel interfacial superconductors, as acknowledged by all the Reviewers. Based on this, we have excluded some existing scenarios on the orientation-dependent superconductivity. Unfortunately, the current soft X-ray techniques cannot provide high enough resolution to resolve the peak-dip-hump to provide impeccable evidence on the interfacial EPC. We performed additional experiments on KTO surfaces, where the observed peak-dip-hump feature provides complementary support for the EPC scenario. Considering the technical limits and the Reviewer's comments, we have added discussions and softened the conclusions on the EPC scenario of the orientation-dependent superconductivity. The revised manuscript is much more rigid and thorough after addressing the Reviewers' comments. We thank all the Reviewers for their nice suggestions and comments.

REVIEWERS' COMMENTS

Reviewer #1 (Remarks to the Author):

I would like to express my appreciation to the authors for their comprehensive response to the reviewers' previous comments and for the substantial revisions made to the manuscript. The authors have carefully considered and addressed the suggestions and feedback provided by the reviewers, resulting in a significantly enhanced and more robust manuscript that effectively communicates their findings.

The authors have successfully addressed the main points raised in the initial review, including the presentation of additional data and analyses to support their findings. The complementary experiments on KTO surfaces, which exhibit clear peak-dip-hump (PDH) structures, provide valuable evidence for the presence of electron-phonon coupling (EPC) in related systems. The authors have made a reasonable effort to address the importance of directly observing PDH features in LAO/KTO, given the current technical limitations.

The inclusion of the EDC line-shape analysis at kF and the consideration of carrier concentration differences through momentum-integrated EDCs and Franck-Condon fittings strengthen the authors' conclusions. The additional data and explanations regarding the closed Fermi surfaces and the discrepancy in relative background intensity between 3D plots and integrated EDCs enhance the overall presentation of the ARPES results.

I commend the authors for their balanced interpretation of the data, particularly in softening the conclusions on the EPC scenario and emphasizing it as a potential candidate explanation for the orientation-dependent superconductivity in LAO/KTO. This demonstrates a rigorous and comprehensive approach to the complex problem at hand.

In conclusion, I believe that the revised manuscript presents a significant contribution to the understanding of the electronic structure and potential mechanisms behind the orientation-dependent superconductivity in LAO/KTO interfaces. The authors have successfully addressed the concerns raised in the initial review, and the manuscript is now suitable for publication in Nature Communications. I have no further questions or comments and fully support the publication of this work, as it will undoubtedly stimulate further research into this intriguing system.

Reviewer #3 (Remarks to the Author):

Referee Report NCOMMS-23-45788A, X. Chen et al.,

I have gone through the revised manuscript along with the detailed responses sent by the authors of this manuscript. I have also looked at the responses to the other reviewers' comments. Evidently, the authors have put in substantial effort to improve the manuscript.

The technical issues raised in the first round of refereeing have been answered by the

revisions to the manuscript and the additional supplementary material. Not all of the answers are completely satisfying, but I do not think it is reasonable to expect more from the authors at this point in the development of the subject. Thus, it is probably now best to leave it to the scientific community to judge this aspect of the work.

I think the main experimental observations merit publication in Nat. Commun. and therefore, support the publication of this work.