nature portfolio

Peer Review File

Universal mechanism of shear thinning in supercooled liquids

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The manuscript by Mizuno and co-authors studies the shear-thinning of the supercooled Gaussian core model (GCM) at high density identifying a 2-step behaviour. The topic is timely and would definitely be of interest to the large community of Statistical Physics, Soft Matter and Material Science. In general, the work is well structured and nicely written, however the conclusions are not fully convincing.

The authors start by showing that, according to previous studies, there are 2 possible scenarios responsible for shear-thinning: 'advection' (from sheared MCT) and 'distortion' (theory by Furukawa). The authors then integrate the distortion effect within the sheared MCT formalism and show that these 2 scenarios emerge at different shear rates. The theoretical results obtained from the 'extended' sheared MCT are compared to results from MD simulations. The main claim is that the relaxation time as well as the diffusion coefficient of the GCM show 2 distinct power-scalings, each with its specific onset shear rate. While the mathematical formalism developed in this manuscript is interesting, there are some points that seem to suggest that this pictures does not actually provide a good description for the GCM. In particular, please see the points below:

1. The claim of 2 power-law scalings is not fully justified. By looking for instance at Fig 2. one can see that the first power-law trend extends at most for 1 decade, making it more a smooth transition from the Newtonian to then non-Newtonian regime rather then a clear separate regime. To support this criticism there are 2 additional main points: i) the corresponding exponent obtained from the theoretical calculations differs quite a lot from the one obtained from simulations, thus suggesting that the theoretical picture described in the paper might not be a good explanation for this system. ii) the authors briefly mention at the end of the manuscript that the viscosity does not show a 2-step shear-thinning and relocate results about this to a future publication. However, I believe that this is quite an important point to be addressed, in particular to fully support or discard the observation of 2 distinct power-law scalings.

2. The study often refers to the cage mechanism as main interpretation of the dynamics in supercooled liquids. However, it is known that the supercooled GCM at high density shows a dynamics which is quite different to other standard glassy fluids, as shown by some of the co-authors of this work (ref 24-25 in the manuscript) and in particular by Coslovich et al (ref 26 in the manuscript), where it was stressed that the dynamics is rather driven by cooperative motion and not by standard cage mechanism (in this direction see also a recent paper by Sposini et al. Soft Matter, 2023). Thus, I wonder, how much of what shown in this paper is based on the assumption that the cage mechanism is valid in this system while clearly this is not an appropriate description for the high density GCM?

3. Finally, how general are the results obtained in this paper given that, as said above, the dynamics of the supercooled high density GCM is quite different to the one of other supercooled liquids?

The authors should address all the points stated above in order for their paper to be re-considered for publication in Communications Physics.

Reviewer #2 (Remarks to the Author):

The authors of "Two-step shear thinning in deeply supercooled liquids" present a numerical analysis of the rheology of the so-called Gaussian core model (GCM) - a dense, glassy fluid of particles interacting with a Gaussian potential. The numerical data is interpreted in terms of a schematic mode-coupling theory (MCT), inspired by previous related work (Refs. 9, 10) and a more recent phenomenological model (Refs. 14, 15). The authors claim that, at variance with most other glassy liquids, the GCM displays a two-step shear thinning around the MCT transition temperature and that this behavior is qualitatively captured by their extended MCT analysis.

The idea of revisiting theoretical approaches for the rheology of dense glassy systems using the GCM as a benchmark is nice and novel. The expectation is that MCT for shear liquids should work well for the GCM - in analogy to what was found at equilibrium. The authors are in the position to give a clear answer to this question, thus orienting future theoretical work in this field. Unfortunately, the work contains some presentation flaws and an erroneous interpretation of the data. These issues make the manuscript unsuitable for publication in Nature Communications or, for what matters, in any other scientific journal. I hope that the authors can revise their manuscript to correct these issues, as I feel that the motivation for this work is compelling.

There are three main issues. The first two are easy to fix, the third one may require a thorough revision, because it affects the very message of the paper.

1) The authors state that the GCM

- "mimics soft materials like star polymers and dendrimers" (abstract)
- "mimics soft materials like star polymers and dendrimers [21, 22]" (p.2)
- "is a good model that mimics soft materials such as star polymers and dendrimers [21]" (p.9)

Besides the useless repetition, these sentences are wrong. The GCM is a model for the effective interactions between linear polymer chains (not star polymers or dendrimers) and the large density / low temperatures used in this study are, anyway, far beyond the range of validity of the model as an effective interaction. It is also well-known that the effective interaction between dendrimers is not Gaussian and this has a serious impact on the phase diagram, especially at high density. The reference to star polymers is also misplaced as the effective potential diverges at small distances, at variance with the GCM. Always concerning star polymers, the authors write that "A recent experiment [22] successfully demonstrated that block copolymer micelles display equilibrium dynamics which match those of the GCM.", but this is again wrong: Ref. 22 used a logarithmic / Yukawa potential and focused on low density states. In Ref. 22 one reads "Our findings for an ultrasoft coarse-grained model which quantitatively reproduces the structural and dynamical behavior of frozen block copolymer micelles as a realization of starlike soft colloids, are in agreement with those of simulations of the Gaussian core model". Thus, those authors just noted a similarity of their findings (weak Stokes-Einstein violation) with similar observations for the GCM. This does not mean that block copolymer micelles "display equilibrium dynamics which match those of the GCM" in particular in the present density / temperature regime. Please revise these sentences related to the link with soft materials.

2) The authors write that their simulations are carried out in a "deeply supercooled" liquid (title, p.2 and p.7). For present-days computer simulations, "deeply" supercooled states are those close and below the experimental Tg, which became accessible thanks to the swap Monte Carlo method. The simulations carried out in this work cover instead a few decades in structural relaxation times, are limited above the MCT temperature and anyway fall short of experimentally relevant time scales. Please avoid this exaggerated wording.

3) The MCT analysis suggests the presence of a two-step shear thinning with two distinct exponents depending on shear rate. The authors build their interpretation on the idea that these two regimes are actually found in the GCM and not in the conventional Kob-Andersen (KA) binary mixture. However, an

unbiased comparison of the data in Fig. 2 (GCM) and Fig. 8 (KA) shows instead that the two models behave pretty much the same way. I attach a figure in which I digitized the data and superposed them (I did a little 5% scaling of the x axis, which does not change the exponents). The good collapse shows there is no qualitative difference between the curves obtained for the two models.

Honestly, I do not understand how the authors can claim the data in Fig. 1 and 2 are well described by $\dot{\gamma}^{-\nu_1}$ in this putative first shear thinning regime: this power law would not even cover a decade in relaxation times and the data clearly show a continuous curvature in this range... just look at the data: there is no consistent power law regime (linear in log-log) in this range of shear rates. One can fit a power law anywhere in this crossover range, but it will only work locally. This means there is no well-defined two-step shear thinning in the GCM. One observes instead a smooth crossover with what looks like a reasonable power law with exponent nu_2<1 only at large shear rates. Contrary to what the authors write, the analysis of Fig.2 b gives no further evidence of a well-defined power law. It would work similarly (i.e. locally) also for the KA model, if one used an exponent nu_1 < nu_2.

The interpretation of the physical origin of the putative two regimes, as governed by "distortion" and "advection" respectively, in terms of the schematic MCT (p.6: "these results validate our expectation that the distortion induces the first thinning while the advection drives the second.") then becomes inconsistent because (i) there is no clear evidence of a well-defined intermediate power-law regime and (ii) the exponent predicted by MCT for the advection regime remains wrong - surprisingly, also for the GCM (!)

Finally, the authors mention at the very end (next to last line of conclusions) that the viscosity does not show any two-step shear thinning in the GCM (!!). This final observation throws the whole interpretation into confusion and it is difficult to accept if one has followed the line of reasoning of the authors (there should be little violation of Stokes-Einstein relation within MCT). This observation would instead make sense if one accepted that there is no two-step shear thinning, not even in the structural relaxation time.

To sum up: I find the study interesting and well-motivated, but the interpretation of the data appears biased. The insistence on power laws is motivated the specific theoretical settings used by the authors, but there is little correspondence with the numerical data. The fact that MCT for sheared liquids does not work well for the GCM is by itself an interesting and somewhat surprising result, which needs to be addressed carefully and without bias. I hope that the authors will use this occasion to revise their manuscript and propose a clear and robust interpretation of the data.

Minor points:

p.2: "The monatomic GCM exhibits slow glassy dynamics close to the dynamical transition point Tc without being intervened by unwanted crystallization."

-> The wording is clumsy: a liquid is not "intervened by crystallization", one may say perhaps "affected by crystallization"?

p.4: "In both cases of such the liquids and the GCM"

-> "of such liquids"

Reviewer #3 (Remarks to the Author):

Report on manuscript: JCP23-AR-02944
Title: Two-step shear thinning in deeply supercooled liquids
Authors: H. Mizuno, A. Ikeda, T. Kawasaki, and K. Miyazaki
Date: 12-Dic-2023

The manuscript by Mizuno et al reports numerical and theoretical results concerning the shear-thining phenomena in supercooled glassy liquids. Using the Gaussian core model (GCM) in the deep supercooled regime (rho = 2.0, T ≳ 3*10^{-6}), the authors performed non-equilibrium molecular dynamic simulations to analyze the effects of a steady shear flow on the system relaxation time and on the (long-time) self-diffusion coefficient. The simulation results indicate that the relaxation time presents two different scaling dependences on the shear rate which are associated with two different shear-thinning regimes. By properly normalizing the numerical results for all temperatures and shear rates, the authors establish a functional form for the relaxation time as a function of the shear rate with the corresponding estimated power-law exponents (Eq. 3 in the manuscript).

To rationalize their simulation results, the authors make use of two approximations. In the first place, they consider the distortion scenario according to which the shear flow modifies the effective density along both the compression and decompression axis, which in turn facilitates the speeding-up of the dynamics of the particles. Using such an argument, the authors were able to determine the onset shear rate for the first shear-thinning regime. Secondly, the authors present a theoretical model based on a schematic version of the MCT equation adapted to take into account the shear flow effects. The authors generalized such an approach by including in the memory kernel of the MCT equation not only an advection term but also a term associated with the previously described distortion scenario.

To rationalize their simulation results, the authors make use of two approximations. In the first place, they consider the distortion scenario according to which, the shear flow modifies the effective density along both the compression and decompression axis, which in turn facilitates the speeding-up of the dynamics of the particles. Using such an argument, the authors were able to determine the onset shear rate for the first shear-thinning regime. Secondly, the authors present a theoretical model based on a schematic version of the MCT equation adapted to take into account the shear flow effects. The authors generalized such an approach by including in the MCT memory kernel not only the advection term but also a distortion term, which follows their previous argumentation. In this way, they can reproduce the two shear thinning regimes and proportionate an explanation of the related two scalings exponents.

In my opinion, the manuscript is well written and correct within the applied methodology and interpretations, it is scientifically sound and the work presented gives some interesting insights for understanding the complex rheology observed in dense suspensions of soft colloids. In this way, I feel the results shown in this paper are worth to be published in "Communications Physics". Therefore, I recommend publication after the minor points detailed below have been addressed.

>> On page 4, 1st column, 1st paragraph, the second sentence reads "Since the dynamics accelerate as the density increases in the GCM, the direction along the compression axis, where bρ is positive and ρeff increases, contributes to the shear thinning". However, in note [20] one can read, "The present work focuses on fragile glass formers, where a decrease in effective density leads to shear thinning according to the distortion scenario." I think these are a little bit contradictory statements and should be made clearer. Moreover, the note [20] is indeed an important statement that should be

included in the main text.

>> The Gaussian Core Model is not bonded and therefore, it is expected that at such high-density many particles will overlap at the same time. In more realistic systems many-body effects would arise, would the crossover between the different shear-thinning behaviors be hindered by accounting for these many-body effects? Please mention that in the manuscript.

>> Page 6, 1st column, last paragraph: I do not understand the term "dephrasing" in the sentence '... the advection and its resultant “dephrasing” of the nonlinear coupling of density fields...'. Is it a typo?

Response to reviewers

Please find attached a revised version of our manuscript titled "Universal mechanism of shear thinning in supercooled liquids" (COMMSPHYS-23-1216). (We have changed the title of our manuscript.)

We would like to express our sincere gratitude for the three crucial referees' reports. We have taken into consideration the referees' comments and suggestions and have thoroughly rewritten our manuscript. After carefully considering the referees' comments, especially those of the second referee, and re-examining our simulation data, we have come to realize that our original claims regarding the Gaussian core model (GCM) and its two-step shear thinning were *not* accurate; instead, it is actually just a crossover. The GCM and the Kob-Andersen (KA) model show very similar scaling behavior, as the second referee correctly pointed out. In addition, we gathered data on the soft sphere (SS) model and the van Beest-Kramer-van Santen (BKS) model from published papers. Surprisingly, all of these glass models exhibit very similar scaling behavior as shown in Fig R1 below, and our theory can unequivocally explain this behavior of all of them. It is really embarrassing that we formulated the theory correctly, presented all the numerical data, and still misinterpreted the data to draw a wrong conclusion. We are immensely grateful to the referees for their valuable insights and feedback, which have helped us significantly improve the quality of our work.

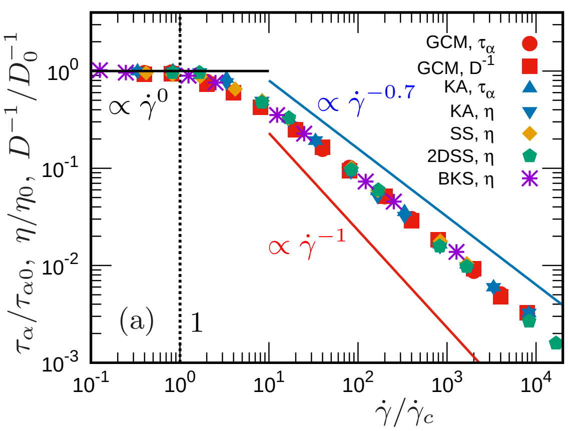

FIG. R1. Shear thinning in all the systems studied in the revised manuscript. This is identical to Fig. 1(a) in the revised manuscript.

In the revised manuscript, while keeping all theory and simulation data intact, we have entirely modified our conclusions. The resulting picture and message are now stronger. All the simulation data on shear thinning are explained by a deceptively simple equation derived from combining the mode coupling theory with the distortion mechanism. We believe that the new manuscript is much stronger, more important, and surprising as it can explain all the shear-thinning data reported in the past.

Below we have addressed all the comments provided by the three referees in the manuscript. We believe that the manuscript is now appropriate for reconsideration for publication in Communications Physics. We would like to express our gratitude once again for the referees' comprehensive and insightful reviews.

Yours sincerely,

Hideyuki Mizuno and Kunimasa Miyazaki on behalf of the authors

Reviewer 1

(1) The manuscript by Mizuno and co-authors studies the shear-thinning of the supercooled Gaussian core model (GCM) at high density identifying a 2-step behaviour. The topic is timely and would definitely be of interest to the large community of Statistical Physics, Soft Matter and Material Science. In general, the work is well structured and nicely written, however the conclusions are not fully convincing.

The authors start by showing that, according to previous studies, there are 2 possible scenarios responsible for shear-thinning: 'advection' (from sheared MCT) and 'distortion' (theory by Furukawa). The authors then integrate the distortion effect within the sheared MCT formalism and show that these 2 scenarios emerge at different shear rates. The theoretical results obtained from the 'extended' sheared MCT are compared to results from MD simulations. The main claim is that the relaxation time as well as the diffusion coefficient of the GCM show 2 distinct power-scalings, each with its specific onset shear rate. While the mathematical formalism developed in this manuscript is interesting, there are some points that seem to suggest that this pictures does not actually provide a good description for the GCM. In particular, please see the points below:

[**Reply**] We would like to express our gratitude to the referee for taking the time to evaluate our manuscript. After carefully re-examining our simulation data, we realized that the referee's observations were correct. Our previous claim about the two-step shear thinning was inaccurate, and it is actually a crossover. In the revised manuscript, we gathered data on the KA, SS, and BKS models. Surprisingly, we found that the GCM displays a very similar scaling behavior as these glass formers, as shown in Fig. R1 (please also see Fig. 1 of the revised manuscript), and our theory can explain this behavior of all of them. In our original manuscript, we correctly formulated the theory and presented all the numerical data, but unfortunately, we misinterpreted the data and came to a wrong conclusion. This is really embarrassing for us. However, we have entirely modified the manuscript's conclusions and believe that the revised version is much stronger, more important, and surprising as it can explain all the shear-thinning data on not only the GCM but also the other canonical models (the KA, SS, and BKS models). Once again, we appreciate the referee's careful comment.

(2) 1. The claim of 2 power-law scalings is not fully justified. By looking for instance at Fig 2. one can see that the first power-law trend extends at most for 1 decade, making it more a smooth transition from the Newtonian to then non-Newtonian regime rather then a clear separate regime. To support this criticism there are 2 additional main points: i) the corresponding exponent obtained from the theoretical calculations differs quite a lot from the one obtained from simulations, thus suggesting that the theoretical picture described in the paper might not be a good explanation for this system. ii) the authors briefly mention at the end of the manuscript that the viscosity does not show a 2-step shear-thinning and relocate results about this to a future publication. However, I believe that this is quite an important point to be addressed, in particular to fully support or discard the observation of 2 distinct power-law scalings.

[**Reply**] We would like to express our gratitude to the referee for providing constructive criticism. As mentioned in our response to the comment **(1)**, we completely agree with the referee that the two-step shear thinning was inaccurate and it should be considered as a crossover. As a result, our theoretical prediction of the exponent $\nu = 0.73$ now matches the numerical observation in the GCM.

In addition, as per the suggestion of the referee, we have added data and discussion on viscosity η in Figures 7 and 8 of the revised manuscript. We found that while the viscosity exhibits the single-step shear thinning behavior as does the relaxation time, its scaling behavior is different from that of the relaxation time. It is widely accepted that the viscosity is directly proportional to the relaxation time, $\eta \propto \tau_\alpha$. However, we have observed that this relationship does not hold true in the GCM at high shear rates. This suggests that the shear modulus, measured as $G = \eta/\tau_\alpha$, is dependent on the shear rate ($\dot{\gamma}$). In the GCM, as $\dot{\gamma}$ increases, G also increases. To better understand this crucial observation, further analysis is necessary in the future.

(3) 2. The study often refers to the cage mechanism as main interpretation of the dynamics in supercooled liquids. However, it is known that the supercooled GCM at high density shows a dynamics which is quite different to other standard glassy fluids, as shown by some of the co-authors of this work (ref 24-25 in the manuscript) and in particular by Coslovich et al (ref 26 in the manuscript), where it was stressed that the dynamics is rather driven by cooperative motion and not by standard cage mechanism (in this direction see also a recent paper by Sposini et al. Soft Matter, 2023). Thus, I wonder, how much of what shown in this paper is based on the assumption that the cage mechanism is valid in this system while clearly this is not an appropriate description for the high density GCM?

[**Reply**] We wish to express our sincere appreciation to the referee for providing us with such a valuable comment. We agree with the referee's comment that the (high density) GCM exhibits equilibrium dynamics, which are not characterized by the standard cage mechanism. In our current study, we have analyzed the relaxation time data in terms of the distortion scenario, which is formulated through equations (2) to (7) in the revised manuscript. These formulations do *not* rely on the cage mechanism. Therefore, in the revised manuscript, we have explained our results without any reference to the cage mechanism.

On the other hand, this point brought up by the referee is certainly significant. Although the GCM and the other models (the KA, SS, and BKS models) show similar shear-rate dependence of τ_α or η, their microscopic dynamics may differ in the shear-thinning regime. To discuss this point, we have added an additional paragraph in the Conclusion section of the revised manuscript, citing the suggested reference.

(4) 3. Finally, how general are the results obtained in this paper given that, as said above, the dynamics of the supercooled high density GCM is quite different to the one of other supercooled liquids?

[**Reply**] We express our gratitude to the referee for their important comment. In our revised manuscript, we collected data on the GCM, KA, SS, and BKS models, and found that these different types of models exhibit very similar scaling behaviors, which are described by $\tau_\alpha,\ \eta \propto \dot{\gamma}^{-\nu}$ with $\nu \sim 0.7$ and $\gamma_c \propto \tau_{\alpha 0}^{-\delta}$ with $\delta \sim 1.4$ (please see Fig. 1 in the revised manuscript). Based on these findings, we believe that our results can be applied to many different systems. However, we acknowledge that the microscopic dynamics of the systems may differ, as the referee pointed out in the above comment **(3)**. We have discussed this point in the Conclusion section of the revised manuscript.

(5) The authors should address all the points stated above in order for their paper to be re-considered for publication in Communications Physics.

[**Reply**] Once again, we would like to express our gratitude to the reviewer for their valuable feedback. We hope that the revised manuscript can be reconsidered for publication in Communications Physics.

Reviewer 2

(1) The authors of "Two-step shear thinning in deeply supercooled liquids" present a numerical analysis of the rheology of the so-called Gaussian core model (GCM) - a dense, glassy fluid of particles interacting with a Gaussian potential. The numerical data is interpreted in terms of a schematic mode-coupling theory (MCT), inspired by previous related work (Refs. 9, 10) and a more recent phenomenological model (Refs. 14, 15). The authors claim that, at variance with most other glassy liquids, the GCM displays a two-step shear thinning around the MCT transition temperature and that this behavior is qualitatively captured by their extended MCT analysis.

The idea of revisiting theoretical approaches for the rheology of dense glassy systems using the GCM as a benchmark is nice and novel. The expectation is that MCT for shear liquids should work well for the GCM - in analogy to what was found at equilibrium. The authors are in the position to give a clear answer to this question, thus orienting future theoretical work in this field. Unfortunately, the work contains some presentation flaws and an erroneous interpretation of the data. These issues make the manuscript unsuitable for publication in Nature Communications or, for what matters, in any other scientific journal. I hope that the authors can revise their manuscript to correct these issues, as I feel that the motivation for this work is compelling.

There are three main issues. The first two are easy to fix, the third one may require a thorough revision, because it affects the very message of the paper.

[**Reply**] We would like to express our sincere gratitude to the referee who took the time to evaluate our manuscript. We are also thankful for the constructive criticism provided by the referee which has helped us to significantly improve our work. After an extensive review of our numerical data, we have come to agree with the referee's comments. We have thoroughly rewritten our manuscript to address the concerns raised and ensure that our work is of high quality. Below we have provided our responses to the comments raised by the referee, and we hope that they will find our revised manuscript to be satisfactory.

(2) 1) The authors state that the GCM
- "mimics soft materials like star polymers and dendrimers" (abstract)
- "mimics soft materials like star polymers and dendrimers [21, 22]" (p.2)
- "is a good model that mimics soft materials such as star polymers and dendrimers [21]" (p.9)

Besides the useless repetition, these sentences are wrong. The GCM is a model for the effective interactions between linear polymer chains (not star polymers or dendrimers) and the large density / low temperatures used in this study are, anyway, far beyond the range of validity of the model as an effective interaction. It is also well-known that the effective interaction between dendrimers is not Gaussian and this has a serious impact on the phase diagram, especially at high density. The reference to star polymers is also misplaced as the effective potential diverges at small distances, at variance with the GCM. Always concerning star polymers, the authors write that "A recent experiment [22] successfully demonstrated that block copolymer micelles display equilibrium dynamics which match those of the GCM.", but this is again wrong: Ref. 22 used a logarithmic / Yukawa potential and focused on low density states. In Ref. 22 one reads "Our findings for an ultrasoft coarse-grained model which quantitatively reproduces the structural and dynamical behavior of frozen block copolymer micelles as a realization of starlike soft colloids, are in agreement with those of simulations of the Gaussian core model". Thus, those authors just noted a similarity of their findings (weak Stokes-Einstein violation) with similar observations for the GCM. This does not mean that block copolymer micelles "display equilibrium dynamics which match those of the GCM" in particular in the present density / temperature regime. Please revise these sentences related to the link with soft materials.

[**Reply**] We appreciate the valuable and detailed comment from the referee. We now understand that the GCM does not represent star polymers and dendrimers. We have taken this feedback into consideration and revised the sentences related to soft materials.

(3) 2) The authors write that their simulations are carried out in a "deeply supercooled" liquid (title, p.2 and p.7). For present-days computer simulations, "deeply" supercooled states are those close and below the experimental Tg, which became accessible thanks to the swap Monte Carlo method. The simulations carried out in this work cover instead a few decades in structural relaxation times, are limited above the MCT temperature and anyway fall short of experimentally relevant time scales. Please avoid this exaggerated wording.

[**Reply**] We completely agree with the referee. The present work studies the temperature regime above the dynamical transition point T_c, which is far above "deeply" supercooled regime. We have revised the word "deeply" to "close to the dynamical transition point T_c".

(4) 3) The MCT analysis suggests the presence of a two-step shear thinning with two distinct exponents depending on shear rate. The authors build their interpretation on the idea that these two regimes are actually found in the GCM and not in the conventional Kob-Andersen (KA) binary mixture. However, an unbiased comparison of the data in Fig. 2 (GCM) and Fig. 8 (KA) shows instead that the two models behave pretty much the same way. I attach a figure in which I digitized the data and superposed them (I did a little 5% scaling of the x axis, which does not change the exponents). The good collapse shows there is no qualitative difference between the curves obtained for the two models.
Honestly, I do not understand how the authors can claim the data in Fig. 1 and 2 are well described by $\dot{\gamma}^{-\nu_1}$ in this putative first shear thinning regime: this power law would not even cover a decade in relaxation times and the data clearly show a continuous curvature in this range... just look at the data: there is no consistent power law regime (linear in log-log) in this range of shear rates. One can fit a power law anywhere in this crossover range, but it will only work locally. This means there is no well-defined two-step shear thinning in the GCM. One observes instead a smooth crossover with what looks like a reasonable power law with exponent $\nu_2 < 1$ only at large shear rates. Contrary to what the authors write, the analysis of Fig.2 b gives no further evidence of a well-defined power law. It would work similarly (i.e. locally) also for the KA model, if one used an exponent $\nu_1 < \nu_2$.
The interpretation of the physical origin of the putative two regimes, as governed by "distortion" and "advection" respectively, in terms of the schematic MCT (p.6: "these results validate our expectation that the distortion induces the first thinning while the advection drives the second.") then becomes inconsistent because (i) there is no clear evidence of a well-defined intermediate power-law regime and (ii) the exponent predicted by MCT for the advection regime remains wrong - surprisingly, also for the GCM (!)

[**Reply**] We would like to express our sincere gratitude for the very important comment provided by the referee. After carefully re-examining our simulation data, we have realized that the referee's comment is entirely correct. Our initial claim about the two-step shear thinning was inaccurate, and it is, in fact, a crossover. The GCM and the Kob-Andersen(KA) model show very similar scaling behavior, as the referee correctly pointed out. In addition, we collected data on the SS model and the BKS model from published papers. Surprisingly, all of these glass models (GCM, KA, SS, and BKS models) exhibit very similar scaling behavior, as shown in Fig R1 (please also see Fig. 1 of the revised manuscript). The thinning exponent $\nu \sim 0.7$ of our theory's prediction is now consistent with the observations of all of these models. We are embarrassed that we correctly formulated the theory and presented all the numerical data, but unfortunately, we misinterpreted the data and reached a wrong conclusion. However, we have entirely modified the manuscript's conclusions and believe that the revised version is much stronger, more important, and surprising as it can explain all the shear-thinning data on not only the GCM but also the KA, SS, and BKS models. We cannot thank the referees enough for their valuable insights and feedback. Their comments have helped us to significantly improve the quality of our work.

(5) Finally, the authors mention at the very end (next to last line of conclusions) that the viscosity does not show any two-step shear thinning in the GCM (!!). This final observation throws the whole interpretation into confusion and it is difficult to accept if one has followed the line of reasoning of the authors (there should be little violation of Stokes-Einstein relation within MCT). This observation would instead make sense if one accepted that there is no two-step shear thinning, not even in the structural relaxation time.

[**Reply**] We have taken into account the referee's comment and decided to add data and discussion on the viscosity η to Figs. 7 and 8 of the revised manuscript. We found that although the viscosity demonstrates a single-step shear thinning behavior, its scaling behavior differs from that of the relaxation time. While it is commonly accepted that the viscosity is directly proportional to the relaxation time ($\eta \propto \tau_\alpha$), our observations show that this relationship does not apply to the GCM at high shear rates. This suggests that the shear modulus ($G = \eta/\tau_\alpha$) is dependent on the shear rate; as the shear rate increases, so does G in the GCM. Further analysis is necessary in the future to better understand this crucial observation.

(6) To sum up: I find the study interesting and well-motivated, but the interpretation of the data appears biased. The insistence on power laws is motivated the specific theoretical settings used by the authors, but there is little correspondence with the numerical data. The fact that MCT for sheared liquids does not work well for the GCM is by itself an interesting and somewhat surprising result, which needs to be addressed carefully and without bias. I hope that the authors will use this occasion to revise their manuscript and propose a clear and robust interpretation of the data.

[**Reply**] We express our sincere gratitude to the referee for their invaluable feedback, which has helped us to accurately interpret our simulation data and implement significant improvements to the manuscript. We hope that the manuscript is now suitable for reconsideration for publication.

(7) Minor points:
p.2: "The monatomic GCM exhibits slow glassy dynamics close to the dynamical transition point Tc without being intervened by unwanted crystallization."
→ The wording is clumsy: a liquid is not "intervened by crystallization", one may say perhaps "affected by crystallization"?
p.4: "In both cases of such the liquids and the GCM"
→ "of such liquids"

[**Reply**] We thank the referee for these points. We have fixed these points in the revised manuscript.

Reviewer 3

(1) The manuscript by Mizuno et al reports numerical and theoretical results concerning the shear-thining phenomena in supercooled glassy liquids. Using the Gaussian core model (GCM) in the deep supercooled regime (rho $=$ 2.0, T $\gtrsim 3 * 10^{-6}$), the authors performed non-equilibrium molecular dynamic simulations to analyze the effects of a steady shear flow on the system relaxation time and on the (long-time) self-diffusion coefficient. The simulation results indicate that the relaxation time presents two different scaling dependences on the shear rate which are associated with two different shear-thinning regimes. By properly normalizing the numerical results for all temperatures and shear rates, the authors establish a functional form for the relaxation time as a function of the shear rate with the corresponding estimated power-law exponents (Eq. 3 in the manuscript).

To rationalize their simulation results, the authors make use of two approximations. In the first place, they consider the distortion scenario according to which, the shear flow modifies the effective density along both the compression and decompression axis, which in turn facilitates the speeding-up of the dynamics of the particles. Using such an argument, the authors were able to determine the onset shear rate for the first shear-thinning regime. Secondly, the authors present a theoretical model based on a schematic version of the MCT equation adapted to take into account the shear flow effects. The authors generalized such an approach by including in the MCT memory kernel not only the advection term but also a distortion term, which follows their previous argumentation. In this way, they can reproduce the two shear thinning regimes and proportionate an explanation of the related two scalings exponents.

In my opinion, the manuscript is well written and correct within the applied methodology and interpretations, it is scientifically sound and the work presented gives some interesting insights for understanding the complex rheology observed in dense suspensions of soft colloids. In this way, I feel the results shown in this paper are worth to be published in "Communications Physics". Therefore, I recommend publication after the minor points detailed below have been addressed.

[**Reply**] We would like to express our gratitude to the referee for taking the time to evaluate our manuscript. After carefully considering the first and second referees' comments and re-examining our simulation data, we have come to realize that our original claims regarding the GCM and its two-step shear thinning were *not* accurate; instead, it is actually just a crossover. It is really embarrassing that we formulated the theory correctly, presented all the numerical data, and still misinterpreted the data to draw a wrong conclusion. However, we have entirely modified the manuscript's conclusions and believe that the revised version is much stronger, more important, and surprising as it can explain all the shear-thinning data on not only the GCM but also the other canonical models (KA, SS, and BKS models). Please also see our responses to the comments raised by the first and second referees.

(2) On page 4, 1st column, 1st paragraph, the second sentence reads "Since the dynamics accelerate as the density increases in the GCM, the direction along the compression axis, where b_ρ is positive and ρ_{eff} increases, contributes to the shear thinning". However, in note [20] one can read, "The present work focuses on fragile glass formers, where a decrease in effective density leads to shear thinning according to the distortion scenario." I think these are a little bit contradictory statements and should be made clearer. Moreover, the note [20] is indeed an important statement that should be included in the main text.

[**Reply**] We would like to express our gratitude to the referee for providing us with this valuable feedback. We completely agree with the referee's observation regarding the contradictory statements presented in the sentences. We would like to explain as follows.

In the case of the GCM, since the dynamics accelerate as the density increases, the direction along the compression axis, where b_ρ is positive and ρ_{eff} increases, contributes to the shear thinning. On the other hand, for the other fragile glass formers like the KA model, the dynamics speed up with decreasing the density, and the direction of the negative b_ρ along the decompression axis causes the shear thinning. This behavior is contrary to that of the GCM; however, in both cases, a minute but finite variation in ρ_{eff} commonly plays a crucial role in the shear thinning process.

We have revised the manuscript to clarify this point, as suggested by the referee. Moreover, we have moved the footnote to the Introduction section, as per the referee's recommendation.

(3) The Gaussian Core Model is not bonded and therefore, it is expected that at such high-density many particles will overlap at the same time. In more realistic systems many-body effects would arise, would the crossover between the different shear-thinning behaviors be hindered by accounting for these many-body effects? Please mention that in the manuscript.

[**Reply**] We express our gratitude to the referee for providing their valuable comment. In our original manuscript, we interpreted our results as per the referee's comment. However, after carefully considering the comments of the first and second referees and thoroughly examining our data, we have discovered that the GCM and other canonical glass formers, such as the KA, SS, and BKS models, exhibit very similar shear-rate dependence on relaxation time or viscosity. This suggests that the many-body effect mentioned by the referee is not evident in the relaxation time or viscosity data. Nonetheless, we acknowledge that this effect may still be observable in the microscopic dynamics. Previous studies have reported significant differences in equilibrium dynamics between the GCM and KA model. Therefore, we believe it is worthwhile to conduct further investigations into their microscopic dynamics in the shear-thinning regime. We have mentioned this point in the Conclusion section of the revised manuscript.

(4) Page 6, 1st column, last paragraph: I do not understand the term "dephrasing" in the sentence '... the advection and its resultant " dephrasing " of the nonlinear coupling of density fields...'. Is it a typo?

[**Reply**] We thank the referee for this comment. We have revised the word "dephrasing" to "decoupling" in the revised manuscript.

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have thoroughly revised their manuscript following the referees' comments and criticism. In particular, after a more careful analysis and interpretation of the data, the picture of a 2-step shear thinning for the GCM has been completely discarded in favour of a universal shear thinning mechanism. Indeed, the authors have included in their study results from other canonical glass formers and found a rather general behaviour for all of them. The theoretical formalism presented in the work remains the main novelty and is now well supported by the data analysis. The overall presentation of the work has also been revised: it is well written and provides a clear and convincing message.
Given all the above, I recommend the paper for publication.

Minor comment:
- typo on page 4, right column, last sentence: "The present systems show similar shinning exponents" -> thinning exponents.

Reviewer #2 (Remarks to the Author):

The authors have successfully taken into account all my comments and criticisms. The message of the paper is now clear, the interpretation is sound and the emerging physical picture is very interesting. I am thus happy to recommend its publication.

I only have some minor comments, which might be taken into account even at the proofs stage:

- abstract: the wording "destruction of the liquid structure" sounds a bit strange
- abstract: "larger thinning exponent" -> perhaps better "too large thinning exponent" (larger requires a "than", which is missing in the sentence)
- p.3: "which are sourced in published papers" -> sourced "from"

Reviewer #3 (Remarks to the Author):

The manuscript by Mizuno et al reports numerical and theoretical results on the shear thinning phenomenon in super-cooled glassy liquids. Using two different models of glass formers, the authors performed non-equilibrium molecular dynamics simulations and MCT-based analytical calculations to describe the effects of a constant shear flow on the system properties (relaxation time, viscosity, and self-diffusion coefficient). The authors made a major modification to the content of the original article by maintaining the same theoretical formulation and simulation data of the original but considering new simulation data from themselves and from other sources to improve the comparison between different systems. This allowed the authors to present a reformulation of their original interpretation and conclusions.

From the analysis of the simulation data, the authors conclude that all systems considered show a similar scaling dependence of the relaxation time with respect to the shear rate in the super-cooled regime. This scaling dependence is not adequately explained by the current sheared MCT model even in the case of the Gaussian core model, which would be expected to be the best testing model for MCT. Then, the authors propose a theoretical model based on a schematic version of the MCT equation taking into account the shear flow effects through the introduction of both advection and distortion terms, which allows to improve the theoretical prediction of the thinning exponents.

In my opinion the revised version of the manuscript is clearly written, it is scientifically sound and the work presented gives some interesting insights for the understanding of the complex rheology observed in dense suspensions of soft colloids. As mentioned above, the authors conducted a thorough review of the scope of their data and conclusions and I believe that they responded adequately to my requests and those of the other reviewers. In this way, I feel the results shown in this paper are worth publishing in "Communications Physics".

Response to reviewers

Please find attached a revised version of our manuscript titled “Universal mechanism of shear thinning in supercooled liquids” (COMMSPHYS-23-1216A).

We would like to express our sincere gratitude for the three referees’ reports. We are pleased that they recommend publication. Below we provide our response to each comment.

Yours sincerely,

Hideyuki Mizuno on behalf of the authors

Reviewer 1

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[**Reply**] We would like to thank the referee for taking the time to evaluate our manuscript. We are also pleased that they recommend publication.

(2) Minor comment: - typo on page 4, right column, last sentence: "The present systems show similar shinning exponents" → thinning exponents.

[**Reply**] We would like to thank the referee for the comment. We have addressed this spelling mistake.

Reviewer 2

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[**Reply**] We appreciate the valuable feedback provided by the referee. We have revised our manuscript in accordance with these comments.

Reviewer 3

(1) The manuscript by Mizuno et al reports numerical and theoretical results on the shear thinning phenomenon in super-cooled glassy liquids. Using two different models of glass formers, the authors performed non-equilibrium molecular dynamics simulations and MCT-based analytical calculations to describe the effects of a constant shear flow on the system properties (relaxation time, viscosity, and self-diffusion coefficient). The authors made a major modification to the content of the original article by maintaining the same theoretical formulation and simulation data of the original but considering new simulation data from themselves and from other sources to improve the comparison between different systems. This allowed the authors to present a reformulation of their original interpretation and conclusions.
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