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Dependence of the Atomic Level Green-Kubo Stress Correlation Function on Wavevector and Frequency. Molecular Dynamics Results from a Model Liquid.

V.A. Levashov
Department of Physics and Astronomy, University of Tennessee, Knoxville, TN 37996, USA.

We report on a further investigation of a new method that can be used to address vibrational dynamics and propagation of stress waves in liquids. The method is based on the decomposition of the macroscopic Green-Kubo stress correlation function into the atomic level stress correlation functions. This decomposition, as was demonstrated previously for a model liquid studied in molecular dynamics simulations, reveals the presence of stress waves propagating over large distances and a structure that resembles the pair density function. In this paper, by performing the Fourier transforms of the atomic level stress correlation functions, we elucidate how the lifetimes of the stress waves and the ranges of their propagation depend on their frequency, wavevector, and temperature. These results relate frequency and wavevector dependence of the generalized viscosity to the character of propagation of the shear stress waves. In particular, the results suggest that an increase in the value of the frequency dependent viscosity at low frequencies with decrease of temperature is related to the increase in the ranges of propagation of the stress waves of the corresponding low frequencies. We found that the ranges of propagation of the shear stress waves of frequencies less than half of the Einstein frequency, extend well beyond the nearest neighbor shell even above the melting temperature. The results also show that the crossover from quasilocalized to propagating behavior occurs at frequencies usually associated with the Boson peak.

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I. INTRODUCTION

In molecular dynamics (MD) simulations, the dependence of the generalized viscosity on frequency and wavevector is often studied using the transverse current correlation function (tccf) \( \langle \mathbf{j}(t) \mathbf{j}(0) \rangle \). It has been demonstrated for the tccf and the generalized viscosity that upon decrease of temperature toward the glass transition there occurs a significant increase in their values for small wavevectors, i.e., for the wavelengths larger than the lengths associated with the second coordination shell \( 6 \) \( 9 \) \( 11 \) \( 14 \) \( 17 \). However, there is not a large increase for the wavevectors associated with the first coordination shell and larger wavevectors \( 6 \) \( 9 \) \( 11 \) \( 17 \). Thus, results for the tccf differ from the results for the structural relaxation, which is often studied with the intermediate scattering function. Relaxation time for the intermediate scattering function, is usually determined (due to de Gennes narrowing) using the value of wavevector corresponding to the nearest neighbor distance. It has also been shown that properties of the tccf function can be modeled using kinetic and viscoelastic models if it is assumed that transport coefficients depend on the value of wavevector \( 17 \) \( 15 \) \( 16 \). These results suggest a non-local nature of the tccf and viscosity close to the glass transition temperature.

A different, but a closely related approach for understanding viscosity is based on the Green-Kubo expression and considerations of the macroscopic stress-stress correlation function \( \langle \sigma(t) \sigma(0) \rangle \). The Green-Kubo expression for viscosity corresponds to zero-wavevector \( \langle \mathbf{k} = 0 \rangle \) and zero-frequency \( \langle \omega = 0 \rangle \) limit of the expression for generalized viscosity \( \mu \). The Green-Kubo method is very common in MD simulations. However, the microscopic nature of the macroscopic stress correlation function is poorly understood. Sometimes it was assumed that the atomic level ssf is local and that there are correlations between the nearest neighbor atoms only \( 24 \) \( 26 \). This view contradicts the older and more recent results from the generalized hydrodynamics \( 8 \) \( 10 \) \( 16 \) \( 27 \) \( 28 \).

Previously we studied the microscopic nature of the macroscopic Green-Kubo ssf by decomposing it into correlation functions between the local atomic level stresses \( 29 \) \( 30 \). Our results explicitly demonstrate non-locality of the stress correlations. They also show that there is a relation between propagating transverse (shear) waves and viscosity. In this paper, we assume that the reader is familiar with the results presented in Ref.\( 29 \) \( 30 \).

Recently, the non-local correlated character of the particles’ displacements in model liquids was discussed in the context of the Eshelby field \( 31 \) \( 33 \). There has also been made observations concerning propagating longitudinal and transverse waves \( 33 \). Non-locality of the correlations is reflected in recent theories \( 34 \).

In order to get intuitive insight into the connection between the propagation of waves and the atomic level ssf we considered a simple model in Ref.\( 35 \). In this model propagating waves are plane waves, like in crystals. It is also assumed there that the atomic environment of every atom is spherically symmetric. We found that if an additional assumption concerning the decay of the stress correlation function for a given frequency is introduced, the atomic level ssf calculated within this toy
model qualitatively resembles the atomic level ssdfs that were obtained in MD simulations on a model liquid in Ref. [29, 30]. While it is clear that this model cannot be used to describe liquids, as in liquids vibrational eigenmodes are not plane waves, it still provides insights into the atomic level ssdf in liquids. The results that were obtained in Ref. [35] with respect to the Fourier transforms provide a guide for the present analysis.

In this paper, by performing the Fourier transform of the previous MD data [29, 30], we demonstrate how the atomic level ssdf method can be used to study properties of the stress waves and how these properties depend on frequency and wavevector. The results show that highfrequency stress waves are quasilocalized and temperature decrease does not strongly affect the degree of their localization. At the same time high-frequency viscosity exhibits only weak temperature dependence. However, the ranges of propagation of low-frequency stress waves significantly increase with decrease of temperature. This increase correlates with the significant increase in the value of low-frequency viscosity. Our data also show that the change from quasilocalized to propagating behavior happens in the range of frequencies associated with the Boson peak, as expected [33, 39].

In this paper, we introduce a wavevector $q$ that characterizes the length scales relevant to the atomic level Green-Kubo ssdf and to the propagating shear stress waves. It is important to realize that the wavevector $q$ is distinct from the wavevector $k$ in the tccf approach. All our results correspond to the case of $k = 0$, i.e., to the case of density fluctuations of very large wavelengths. This effectively means that we study shear stress waves in the absence of local density fluctuations. See Appendix A for more details.

The paper is organized as follows. In section II, we provide some details about our MD system. In section III, we describe how certain features in the ssdf can be separated. This separation is useful for further analysis. In section IV, we discuss some features of the ssdf. In section V, we describe how we apply the Fourier transforms. In section VI, we describe the results of the Fourier transforms at the lowest temperature that we studied. In section VII, we discuss the connection between our results and frequency dependent viscosity. In section VIII, we discuss the results of the Fourier transforms at several higher temperatures. We conclude in section IX. In Appendix A, we discuss the relation of our approach to the tccf approach. In Appendices B and C, we two particular features of the Fourier transforms.

II. DETAILS OF MD SIMULATIONS AND REDUCED UNITS

Details of our MD simulations were described in Ref. [29, 30]. Previously, when reporting results of MD simulations, we had not used reduced units. To be consistent, we also used non-reduced units in this paper. To make a comparison between our results and the results from other publications, we describe in Ref. [40] the relations between our units and the units of the corresponding Lennard-Jones potential.

III. SEPARATION OF THE SSCF INTO THE PDF-LIKE AND WAVE-LIKE PARTS

Some figures in this paper have several panels. In referring to these panels we use ($n$ – row, $m$ – column) notation.

Panel (1,1) of Fig. 1 shows the ssdf at 1500 (K) obtained on the large system of 43904 particles with $(L/2) \approx 41.21$ (Å) [29], where $L$ is the length of the side of the cubic box. The main features present in the ssdf are the stress waves and the pair distribution function pdf-like stripe structure. To understand the results of the Fourier transforms of this panel, it is convenient to make an approximate separation of the ssdf into the parts that correspond to the waves and to the pdf-like structure.

The implemented separation procedure is based on the following observation. Consider the zero-time cut of panel (1,1) of Fig. 1. This cut is shown as the lowest curve in Fig. 6 of Ref. [29]. Note the oscillating behavior for the distances beyond 10 (Å). Also note that for a particular maximum, which is beyond 10 Å, the distances from it to the nearest two minimums are approximately the same. Thus, for a particular maximum at $r$ the value of the ssdf at it, $f_{gr}(r)$, could be approximated as:

$$f_{gr}(r) \approx -\frac{1}{2} [f_{gr}(r - \Delta r) + f_{gr}(r + \Delta r)] \ ,$$

where $f_{gr}(r - \Delta r)$ and $f_{gr}(r + \Delta r)$ are the values of the ssdf at the left and right minimums nearest to this maximum. Finally, note that (1) can be used not only for the maximum/minimum values, but essentially for all $r$ beyond the third or fourth coordination shells. This, however, does not hold for nonzero times and those regions of $r$ that contain contributions from the waves, as can be seen in Fig. 2 (a).

Further, we assume that the pdf-like contribution to the ssdf satisfies (1), while the wave’s contribution does not. In our iterative numerical procedure we consider different times independently. We assume that for a particular time we know the distance dependence of the wave contribution on step $n$, i.e., $f_{w}^{n}(r)$. On the first step we assume that it is zero for all distances. Then we calculate the pdf-like contribution to the ssdf:

$$f_{gr}^{n}(r) = f(r) - f_{w}^{n}(r) \ .$$

Then, in accord with (1), we define:

$$f_{gr}^{n+1}(r) = -\frac{1}{2} [f_{gr}^{n}(r - \Delta r) + f_{gr}^{n}(r + \Delta r)] \ ,$$

where $\Delta r$ is the distance from the nearest maximum to its nearest minimum. $\Delta r$ is distance dependent (this depen-
FIG. 1. The sscf of the large (43904) system at 1500 K, the wave and pdf-like contributions to it, and their Fourier transforms. The notation \((n,m)\) in the text will be used to refer to the panels of the figure: \(n\)-for the rows and \(m\)-for the columns. One should not consider the values of \(q < 0.153 \text{Å}^{-1}\) and the values of \(\omega < 0.00025 \text{(fs)}^{-1}\).


FIG. 2. Separation of the 500 (fs) cut of the SSCF at 1500 (K) into the pdf-like and wave-like contributions. (a) 500 (fs) cut of the SSCF from panel (1,1) of Fig. 1. (b) pdf-like contribution. (c) Contribution from stress waves.

dence is weak in practice). Then we define contribution from the wave on step \( n + 1 \) as:

\[
 f_{n+1}^w (r) = f(r) - f_{n+1}^{gr} (r) .
\]  

Finally, we assume that the amplitude of the wave does not change significantly over the distance between the nearest maximum and minimum. It is indeed so according to Fig. 2. Thus, for convergence of the algorithm, we introduce the average amplitude of the wave, \( \bar{f}_{n+1}^w \), in which the averaging goes over the interval \((r - \Delta w, r + \Delta w)\):

\[
 \bar{f}_{n+1}^w (r) = \frac{1}{2\Delta w} \int_{r-\Delta w}^{r+\Delta w} f_{w}^{n+1}(\xi) d\xi
\]  

With this new value of \( \bar{f}_{n+1}^w (r) \) we go back to \ref{eq:4} closing the iteration loop.

The results of the described procedure are shown in Fig. 2 for temperature 1500 (K) and for the time-cut 500 (fs) cut of the SSCF from panel (1,1) of Fig. 1. The number of iterations was 1000.

Note that the procedure does not work for distances \( r < 10 \) (Å). This is so because for such \( r \) in the pdf-like contribution there is no periodicity assumed in the separation procedure. The value of \( r = 10 \) (Å) approximately corresponds to the inclusion of the 4th coordination shell. This distance could be associated with the medium range order distance \( 11 \) [41, 42]. It is of interest that viscoelastic continuous approximations appear to be valid at distances larger than 10 (Å), but not at smaller distances \( 16 \).

We applied the described algorithm to all times in panel (1,1) of Fig. 1. The results are shown in panels (1,2) and (1,3). Note again that the procedure does not work for \( r < 10 \) (Å). Figure 3 shows constant \( r \) cuts from panel (1,2). First peaks in the curves correspond to the compression wave, the second peaks to the shear wave. It would be useful to develop a method that would allow us separate contributions from the shear and compression waves.

IV. WHAT WE SEE IN PANELS (1,1), (1,2), AND (1,3)

The intensity in panel (1,1) of Fig. 1 shows the ensemble averaged atomic level stress correlation function between a central atom and atoms located inside the spherical annulus of radius \( r \) and thickness \( \Delta r = 0.2 \) (Å). This intensity is also normalized to the magnitude of the stress auto-correlation function at zero time \( 29 \) [30]. Figures 6 and 7 of Ref. [29], and Figures 2, 3 of this paper further clarify the scale of the correlations. For example, the magnitude of the stress correlation of a central atom with the maximum intensity annulus in the first coordination shell at zero time is \( \sim 0.35 \) of the stress auto-correlation function at zero time (width of the annulus is \( \Delta r = 0.2 \) (Å). Normalization to the atomic stress auto-correlation functions was also made.

FIG. 3. Constant \( r \) cuts of panel (1,2). The legends show the values of \( r \). The curves were shifted so that they do not overlap. The first peaks in the curves are due to the compression wave, while the second peaks are due to the shear wave. Note that the shear peaks are larger and broader. The scale on the \( y \)-axis, corresponds to the correlation between the central atom and atoms in the spherical annulus of radius \( r \) and thickness \( dr = 0.2 \) (Å).
Panels (1,1), (1,2), and (1,3) raise several questions. For example, as panel (1,1) shows the atomic level decomposition of the macroscopic shear sscf $S^{xy}S^{xy}$, it is reasonable to wonder why we see in it the pdf-like structure and the contributions from the longitudinal waves. Indeed, both of these features should be related to the density fluctuations and not to the shear sscf. The explanation can be related to the results presented in Ref. [46]. There it was shown that different components of the atomic level stress tensor on the same atom are correlated. On the other hand, the presence of correlations between the different stress components on the same atom questions the results and derivations in Ref. [43], as there, when the equipartition law is derived, it is assumed that different stress components on the same atom are independent. However, the derivations of the equipartition in [43] are based on the Taylor expansion and considerations of only those terms which are quadratic in atomic strain. Under this quadratic approximation, different stress components on the same atom are independent in the spherical (cubic) representation. Thus, the results presented in Ref. [46] can be related to the higher order terms in the Taylor expansion. This means that the density-density correlations which we see in panels (1,1), (1,2), and (1,3) can be related to higher order terms. These questions require further clarifications.

A. pdf-like contribution

It is reasonable to expect that the decay of the pdf-like contribution in panel (1,3) is related to the decay of the van Hove correlation function [48]. Figure 4 shows dependencies on time of the maximums in the pdf-like contribution to the sscf. For example, 12 Å maximum was found as the maximum value (for every time) of the pdf-like contribution in the interval of distances between 11 Å and 13 Å. Differences between the 10 Å, 12 Å and the other curves are likely to be caused by the stress waves: as intensities of the stress waves are larger at small distances it is likely that these higher intensities stimulate faster decay in the pdf-like stripes. The decay in the amplitudes of the stripes for $t < 200$ (fs) is likely to be due to the rattling cage motion. The decay for $t > 300$ (fs) is likely to be related to the particle diffusion. If we assume that the particles that diffuse away from the spherical annulus completely lose the correlation with the original state, while those that remain keep this correlation, then the magnitude of the remaining correlation should be proportional to the number of the particles remaining in the annulus. Since the rate of diffusion away from the spherical annulus should be proportional to the number of particles remaining in the annulus, the number of the remaining particles should decrease exponentially with time. Thus the pdf-like stress correlation function at large distances at $t > 300$ (fs) should decay exponentially with time. This behavior can be observed in the inset of Fig. 4.

B. Stress waves’ contribution

It is clear that panel (1,2) shows propagating shear and compression waves. Previously we argued that shear stress waves are related to viscosity [29, 30]. Thus, it is important to understand the features in panel (1,2). However, it is not clear how stress waves translate into the features observed in the sscf. The nature of these stress waves also remains obscure. These are complicated questions for liquids, as currently there is no accepted and convenient way to describe vibrational dynamics in disordered media and its coupling to diffusion [36, 39, 49-51]. In our view, it is possible that the atomic level stress correlation function that we consider here represents an alternative way to describe vibrational dynamics.

In order to gain at least some insight into the connection between the vibrational dynamics and the atomic level stress correlation function we considered in Ref. [35] a simple model. In this model vibrational modes are represented by plane waves, like in crystals. Of course, plane waves do not represent vibrational eigenmodes of liquids [40, 49, 49, 51]. However, in our view, considerations in Ref. [51] provide insight into the nature of the connection between the stress waves and the atomic level stress correlation function.
V. FOURIER TRANSFORMS OF THE SSCF

In our previous considerations, the atomic level scco, $F(t, r)$, is defined as a correlation function between a central atom and atoms inside the spherical annulus of radius $r$ and thickness $dr$. This definition naturally follows from the Green-Kubo expression for viscosity. For further analysis and in view of Ref.\[35\] we introduce:

$$f_p(t, r) \equiv \frac{1}{(4\pi r^2)^2} F(t, r), \quad f_r(t, r) \equiv r \cdot f_p(t, r), \quad (6)$$

where $f_p(t, r)$ is the atomic level stress correlation per pair of particles.

We define the Fourier transform over $t$ of $f_r(t, r)$ as:

$$\tilde{f}_r(\omega, r) \equiv \int_0^\infty f_r(t, r) \cos(\omega t) dt. \quad (7)$$

It was shown, in the framework of the model discussed in Ref.\[35\], that if vibrations are non-decaying plane waves, then $\tilde{f}_r(\omega, r)$ should, for every $\omega$, exhibit constant amplitude oscillations in $r$ with a wavelength determined by the dispersion relation $\omega(r)$. Since, for different $r$ the Fourier transforms over $t$ are independent, we transform $F(t, r)$ instead of $f_r(t, r)$. In the case of non-decaying plane waves, amplitudes of peaks in $\tilde{F}(\omega, r)$ should linearly increase with increase of $r$.

We define the Fourier transform over $r$ of $f_r(t, r)$ as:

$$\tilde{f}_r(t, q) \equiv \int f_r(t, r) \sin(qr) dr. \quad (8)$$

As shown in Ref.\[35\], $\tilde{f}_r(t, q)$ for non-decaying plane waves should exhibit constant amplitude oscillations in $t$ with a period determined by the dispersion relation.

Equation \ref{eq:7} can also be rationalized from a different perspective. It is natural to assume that the stress correlation function for a particular pair of atoms, i.e., $f_p(t, r)$, depends on the direction of the radius vector, $r$, from one atom to another. Let us define the three-dimensional Fourier transform of this stress correlation function as it is usually done:

$$\tilde{f}_p(t, q) \equiv \int f_p(t, r) \exp(-iqr) dr. \quad (9)$$

In isotropic cases, $f_p(t, r) \equiv f_p(t, r)$ and \ref{eq:9} could be rewritten as:

$$\tilde{q} \tilde{f}_p(t, q) \equiv \int [r f_p(t, r)] \sin(qr) dr. \quad (10)$$

It follows from \ref{eq:7} that the expression \ref{eq:10} is equivalent to the expression \ref{eq:8}. The expression \ref{eq:10} is similar to the expression that connects the pair distribution function, to the reduced scattering intensity $\tilde{p}$. The Fourier transform in time-space naturally follows from the formulas \ref{eq:7}. It was shown in Ref.\[35\], in the frame of the model considered there, that the Fourier transform of $f_r(t, r)$ over $t$ and $r$ should lead to the dispersion relation.

VI. RESULTS OF THE FOURIER TRANSFORMS AT 1500 K

Since $F(t, r)$ was obtained in MD simulations on systems of finite sizes with periodic boundary conditions there is a lower limit on the possible values of $q$ that we can consider. See Ref.\[35\] for details.

A. Time to frequency Fourier transform

The second row of Fig.\[4\] shows $\tilde{F}(\omega, r)$, i.e., time to frequency Fourier transforms \ref{eq:7} of $F(t, r)$ and contributions to it from the wave-like and the pdf-like parts. Panels (2,1), (2,2), (2,3) were obtained from the data in panels (1,1), (1,2), (1,3) respectively.

In panel (2,2) contributions from the shear and compression waves are mixed. For an analysis of the stress waves it would be very useful to find a way to separate contributions from these waves. Since the amplitude of the compression wave in panel (1,2) is significantly smaller than the amplitude of the shear wave, it is reasonable to assume that features in the upper panels of Fig.\[5\] are dominated by the shear waves.

It is useful to compare panel (2,2) of Fig.\[4\] of this paper with panel (1,2) of Fig.7 in Ref.\[35\]. Note, however, that panel (2,2) of Fig.\[1\] shows $t$ to $\omega$ Fourier transform of the function $r^2 \tilde{f}_p(t, r)$, while panel (1,2) of Fig.7 in Ref.\[35\] shows the Fourier transform of the function $r f_p(t, r)$. We show in this paper the Fourier transform of $r^2 \tilde{f}_p(t, r)$ because it is more directly related to the generalized viscosity and also because in Fig.\[5\] this $r^2$-scaling allows showing relative amplitudes of the peaks in $F(\omega, r)$ more clearly.

Figure \ref{fig:5} shows constant $\omega$-cuts of panel (2,2). If in panel (2,2) there were only shear waves, then, according to Ref.\[35\], for every $\omega$ in Fig.\[5\] the period of oscillations in $r$ would give the wavelength that corresponds to this value of $\omega$. If the scco were caused by non-decaying plane waves, then the amplitudes of the peaks in Fig.\[5\] would linearly increase with increase of $r$. However, the amplitudes of the peaks in Fig.\[5\] decrease with increase of $r$. This behavior suggests that the dynamic underlying the behavior of $F(t, r)$ is very different from the vibrational dynamics of non-decaying plane waves.

The lower panel in Fig.\[5\] shows the dependence of wavevector on frequency determined from the two upper panels. This dependence should primarily correspond to the dispersion relation for the shear waves. Indeed, the slope of the curves corresponds to the speed $c = 3$ (km/s), i.e., to the shear waves, according to panel (2,2) of Fig.\[1\].

Still, this picture should contain certain distortions due to the compression waves.

By comparing the scales on the $y$-axes in the upper panels of Figure 5 note that the amplitudes of the waves
FIG. 5. Constant $\omega$ cuts from panel (2,2). Legends show the values of $\omega$. The blue curves in upper two panels show the result from the large system with $(L/2) \approx 41.2$ (Å). Green curves show the results from the system with $(L/2) \approx 20.6$ (Å). We use the positions of the maximums and minimums, $L/\pi q$. Lower horizontal panel shows the dependence of $q$ on $\omega$. Different curves in the lower panel correspond to different selections of red squares used for the determination of wavelength. For example, the notation (2,4) corresponds to the selection of the second and the forth squares from the left to determine the wavelength.

for higher frequencies are significantly smaller than the amplitudes for lower frequencies. Note also that, even for the highest frequencies shown, the waves propagate over, at least, 5 interatomic spacings ($r_{nn} \approx 2.6$ (Å)). In this context the following comment is relevant. It can be seen in panel (1,2) of Fig. 1 and in Fig. 3 that the feature corresponding to the compression waves is not just smaller in the amplitude than the feature corresponding to the shear waves, but it is also narrower in $t$ and in $r$. Since it is narrower in $t$ its Fourier transform over time decays in a wider range of frequencies. Thus contributions from the compression waves to the higher frequency curves in Fig. 5 should be relatively larger than to the lower frequency curves. Because of the overlap of contributions from the compression and shear waves, we do not discuss here attenuation rates for different frequencies.

In considerations of the macroscopic (tcf) it is assumed that only transverse waves contribute to it [1–6, 8–10]. However, in view of the results discussed above, it is likely that compression waves also affect the tcf. Thus the results obtained from the analysis of the tcf can be distorted by the compression waves. While the distortions should not be very significant this issue deserves attention and clarification.

It follows from panel (2,2) of Fig. 1 that the main sickle feature vanishes at large distances because of the finite system size. This effect can also be seen in Fig. 9. Thus, periodic boundary conditions (PBC) affect the stress waves of small frequencies, i.e., $\nu \approx 0.0005 - 0.001$ (fs$^{-1}$). It is shown in section (C) that contributions from the shear and compression waves overlap in the main sickle feature.

Flattening of the main sickle feature at low temperatures in the region of frequencies between 0.001 and 0.002 (fs$^{-1}$) means that the stress waves of the lower frequencies can propagate over large distances. Frequency $\nu = 0.001$ (fs$^{-1}$) corresponds to the energy $h\nu \approx 4.1$ (meV). This energy approximately corresponds to the energy of the boson peak in metallic glasses [38, 39]. Thus significant increase of the propagation range with decrease of temperature happens in the range of frequencies usually associated with the boson peak. In a recent review [39] it was stated, on the basis of Ref. [36, 37], that: "There appears to be a growing consensus that the frequency of the boson peak corresponds to the maximum frequency at which transverse phonons can propagate in the disordered material ...", Our data are in agreement with this statement.

In panel (2,3) seemingly faster decay of the vertical stripes at large distances is misleading. Perceived behavior originates simply from the smaller amplitudes of the stripes at large distances at zero time. According to Fig. 4 at large distances all stripes decay at the same rate.

VII. FREQUENCY DEPENDENT VISCOSITY

It is known that frequency dependent viscosity, $\eta(\omega)$, exhibits on the decrease of temperature frequency dependent increase [10–12, 54]. The increase is the most significant for small $\omega$. Considerations of our results in this context provide additional insights into this phenomena.

It follows from the previous definitions of $\eta(\omega)$ [13, 10] and our definitions [29, 30] that:

$$\eta(\omega) = \frac{\rho_o}{k_b T} \int_0^{t_{\text{max}}} \left\{ \int_0^{R_{\text{max}}} F(t, r) dr \right\} \cos(\omega t) dt \quad (11)$$

Or:

$$\eta(\omega) = \frac{\rho_o}{k_b T} \int_0^{R_{\text{max}}} \tilde{F}(\omega, r) dr \quad . \quad (12)$$

Thus, for every $\omega$ in panel (2,1) of Fig. 1 the integral over $r$ gives $\eta(\omega)$. In principle, integration over a range of distances, $(r_1, r_2)$, should allow to estimate how this range contributes to $\eta(\omega)$. Figure 6 shows the results of integrations over all distances in panel (2,1) and similar panels at some other temperatures. The self contribution at $r = 0$ (Å) was not counted. We used the data from panel
FIG. 6. Dependencies of the frequency dependent viscosity on frequency for different temperatures. For better statistics, the averaging of the data over the window of frequencies, \(\omega \pm \Delta \omega\), with \(\Delta \omega = 0.00003\) (fs\(^{-1}\)), was done. The data show that on the decrease of temperature low-frequency viscosity significantly increases, while high frequency viscosity is not strongly affected.

(2,1) instead of (2,2) in order to avoid distortions of the data associated with bad separation of the ssf at small distances. However, we checked that use of panel (2,2) instead of (2,1) does not affect the results qualitatively.

We see that on the decrease of temperature, the rise in viscosity is the most significant for low frequencies. This is expected in view of the previous publications on the subject [6, 10, 12, 54]. However, our considerations explicitly demonstrate that this rise is associated with the character of propagation of low-frequency shear stress waves. In our view, this demonstration provides new insights into the previous discussions.

### A. Distance to wavevector Fourier transform

Panels (3,1), (3,2), (3,3) show \(r\) to \(q\) Fourier transforms of the ssf over \(r\) is similar to the transform of the pair density function, \(G(r)\), into the structure factor, \(S(q)\); if \(G(r) = 4\pi r \rho(r) - \rho_0\) then \(q[S(q) - 1] = \int_0^{\lambda_{\text{max}}} G(r) \sin(qr) dr\) [52]. Thus, knowledge of the general relations between \(G(r)\) and \(S(q)\) can help in guessing the roles certain features. This parallel allows us to relate the negative intensity near \(3\) (Å\(^{-1}\)) in (3,3) to the periodicity in \(r\) of the pdf-like contribution to the ssf (see (1,3)). The width of the 3 (Å\(^{-1}\)) feature in (3,3) is related to the extend of the pdf-like oscillations in \(r\).

Panels (3,1) and (3,2) show for how long in time stress waves, with a particular value of the wavevector, exist. If the stress waves were non-decaying plane waves, then, according to Ref. [35], for every \(q\) the amplitude of oscillations would be constant in time. Note that non-zero intensity for smaller \(q\) exists for larger times than nonzero intensity for larger wavevectors. Recall that in panels (3,1) and (3,2) the contributions from the shear and compression waves overlap.

Figure 7 shows constant \(q\)-cuts of panel (3,2) of Fig.6. Every particular \(q\)-cut was scaled to the maximum intensity in the sickle feature for this \(q\) and then shifted. The dependence of this maximum intensity on \(q\) is shown in the inset. Different curves correspond to the different values of \(q\) (in Å\(^{-1}\)) given in the legends. Upper curves correspond to the upper legends.

Note that the main sickle feature ends at \(q_{\text{wmax}} \approx 1.75\) (Å\(^{-1}\)), i.e., at \(\lambda_{\text{wmin}} = 2\pi/q_{\text{wmax}} \approx 3.6\) (Å). Thus, \(\lambda_{\text{wmin}} \approx 1.4d\), where \(d\) is the average distance between the nearest particles for the chosen value of the density and also the equilibrium distances between a pair of particles for our potential. The fact that the smallest possible wavelength of the shear stress waves is \(\approx 1.5d\) is in approximate agreement with the other results [7, 16]. In our data, the crossover in the main sickle feature happens at \(q_c \approx 0.5\) (Å\(^{-1}\)), i.e., at \(\lambda_c \approx 12.6\) (Å) or \(\approx 4.8d\). At larger distances, according to Ref. [16], ordinary hydrodynamics with \(q\)-independent transport coefficients is valid, while at smaller distances the situation is more
FIG. 8. The wave-like contributions to the sscf at different temperatures on the systems of 43904 and 5488 particles. Panels (1,1) and (1,3) are for the 43904 particle system. All other panels are for the 5488 particle system. Panels (1,1) and (1,2) are for \( T = 1500 \) (K). Panels (1,3) and (1,4) are for \( T = 2000 \) (K). Panel (2,1) is for 3000 (K), (2,2) is for 5000 (K), (2,3) is for 7000 (K), (2,4) is for 10000 (K). Note in panels (1,1) and (1,2) bright vertical stripes at \( r \approx 5 \) (Å). These stripes show that separation procedure used to produce wave-like contributions does not work for these distances. Note also that there are no such bright vertical stripes in the other panels. In the text we argue that this bright vertical stripe is the origin of the bonfire feature in the \((t,q)\)-sscf.

It is also possible to consider, from panels (3,1) and (3,2), wavevector dependent viscosity and thus study how different times contribute to it. However, in view of the discussion in Appendix (A), these considerations need more insights and we will not focus on them now.

### VIII. EVOLUTION OF THE DATA WITH TEMPERATURE

In this section we address the evolution of the sscf and its Fourier transforms with temperature. We also discuss size effects by comparing the data on the intermediate system of 5488 particles with \((L/2) = 20.06\) (Å), and on the large system of 43904 particles with \((L/2) = 41.21\) (Å).

The total sscfs for the two systems at different temperatures are shown in Fig.4,5 of Ref.[29]. Figure 8 of this paper shows wave’s contributions to the sscfs for different temperatures and systems in \((t,r)\)-space. The comparisons of panels (1,1) with (1,2) and (1,3) with (1,4) show size effects at low temperatures. It is clear that the finite size of the system affects propagation of the stress waves. It is also clear that the stress waves are more complicated.

There are two features in panels (3,1) and (3,2) that we discuss in sections (B) and (C). The first feature is a positive intensity that is centered at \( q \approx 1.15 \) Å\(^{-1}\) and extends in time from approximately 300 (fs) to 1700 (fs). We call this feature the bonfire. Another feature extends in time from 0 to 200 (fs) and in \( q \) from 0 to 1.3 (Å\(^{-1}\)). We call this feature the tongue.

It turns out, that both features originate from the interval of distances between \( \approx 2 \) (Å) and \( \approx 7.5 \) (Å). The shape of the tongue feature is affected by the position of the origin of the stress waves (they start from the first coordination shell and not from \( r = 0 \) (Å)). The bonfire feature is related to the famous splitting of the second peak in the pair distribution function which is associated with some local arrangements of particles which agglomerate into larger domains [42, 55, 56]. The bonfire feature is also present in Fig.7 though it is difficult to see it.

In panels (4,1), (4,2), and (4,3) show the Fourier transforms of \( f_r(t,r) \). One can guess in (4,1) and (4,2) broad dispersion curves associated with the stress waves. The dispersion, however, is not well pronounced.

### B. Fourier transform in time and space

Panels (4,1), (4,2), and (4,3) show the Fourier transforms in time and space of \( f_r(t,r) \). One can guess in (4,1) and (4,2) broad dispersion curves associated with the stress waves. The dispersion, however, is not well pronounced.
FIG. 9. The \textit{sscf} in (\(\omega, r\))-space for different systems and at different temperatures. The locations of panels correspond to those in Fig. 8. The scales on the z-axes are the same as in panel (2,2) of Fig. 1.

FIG. 10. The \textit{sscf} in (\(t, q\))-space for different systems and at different temperatures. The locations of panels correspond to those in Fig. 8. Note that panels (1,1) and (1,2) contain the \textit{bonfire} feature, while the other panels do not. From comparisons with panels in Fig. 8, it follows that the \textit{bonfire} feature is related to the bright vertical stripe at \(r \approx 5\) (Å).

pronounced at 1500 (K) than at 2000 (K). The results in the second row show gradual disappearance of the stress waves with increase of temperature.

Note in the results for 1500 (K) a bright vertical line at \(r \approx 6\) (Å). Note also that there is not a well pronounced line in the results for 2000 (K). Comparisons with the corresponding panels in Fig. 10 suggest that this vertical line is related to the \textit{bonfire} feature.
Figure 9 shows \( t \) to \( \omega \) Fourier transforms of the ss cf s. It follows from the comparisons of panels (1,1) with (1,2) and (1,3) with (1,4) that the main sickle feature vanishes at large distances because of the finite system size. In panel (2,2) of Fig.1 the same sickle feature extends to significantly larger distances. Thus PBC affect frequencies \( \nu < 0.004 \) (fs\(^{-1}\)).

Figure 10 shows the ss cf s in \((t, q)\)-space. Note that the Fourier transforms of the ss cf s in \((t, q)\)-space obtained on the large and on the intermediate systems exhibit different behaviors at small \( q \). These differences should be related to the finite size effects [53]. See Fig.1 for a wider range in time.

IX. CONCLUSION

We investigated the Fourier transforms of the atomic level Green-Kubo ss cf s obtained in MD simulations on a model liquid. These considerations demonstrate that the atomic level ss cf can be used to study how lifetimes and ranges of propagation of stress waves depend on their frequency and wavevector. It was also demonstrated that the crossover from quasilocalized to propagating behavior occurs at the frequencies usually associated with the Boson peak, confirming previous results [26, 29]. We found that the ranges of propagation of the shear stress waves for frequencies less than half of the Einstein frequency extend well beyond the nearest neighbor shell.

As temperature decreases the ranges of propagation of low frequency stress waves increase. Our results suggest that this increase is responsible for the increase in the value of low frequency viscosity. Thus, at high temperatures \((T > 2T_A, \text{ where } T_A \text{ is the potential energy landscape crossover temperature [57]}\)), stress waves of all frequencies decay on the length scales of 10 interatomic distances or less. As temperature is lowered, the increase in the ranges of propagation for the lower frequency waves is more significant than for the higher frequency waves. Our results suggest that this increase is related to the increase of low frequency viscosity and correspondingly viscosity in general.

The conclusions to which we arrived using our new method are expectable, in view of the other publications [6, 9–16]. However, in our view, investigations with this method compliment the results obtained with other methods.

Our data show that viscosity is related to the propagating stress waves. On the other hand, it was argued recently that at low temperatures relaxation of the shear stresses should become activated [58, 59]. It is of interest to study if viscosity at lower temperatures decouples from the shear stress waves, or if activated dynamics is causing decay of the stress waves, but viscosity remains related to them. For this it would be necessary to study a different system as the system that we studied crystalizes at relatively high temperatures.

Our results also suggest that the decay of the pdf-like part of the atomic level ss cf at large times is related to diffusion of particles.

The fact that we see compression waves in the shear stress correlation function should be related to the existence of correlations between the different components of the atomic level stresses on the same site [46, 47].

Our method has important shortcomings. For example, one would not suppose from our results, as they are the averages over many atoms and times, about the presence of force chains [60, 64] or chain-like displacements [65]. It appears that the spherical averaging that we perform also averages out the long range Eshelby field present in the system [31, 33]. It is unclear if it is possible to see dynamic heterogeneity [66] with our method. These shortcomings, however, are also present in the t ccf technique and in other approaches that rely on considerations of macroscopic quantities.

A separate question of interest is in what range of distances can the ss cf be modeled using viscoelastic approximations? The separation procedure that we used to extract the wave-like and the pdf-like contributions to the ss cf suggests that continuous approximation may not work for the distances smaller than 3 or 4 interatomic distances, but can work for larger ranges. This is in agreement with some other results [7, 15, 16, 41, 42].

X. ACKNOWLEDGMENTS

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Appendix A: On the connection between the transverse current correlation approach and our considerations

As we discussed in this paper the dependence of the ss cf on the wavevector \( q \) it is important to note that the wavevector \( q \) that enters into our considerations is distinct from the wavevector \( \mathbf{k} \) that usually enters into the discussions of the t ccf .

In derivations of the expressions for generalized viscosity through correlation functions the wavevector \( \mathbf{k} \) is related, in particular, to the density fluctuations. As we introduce the wavevector \( q \), it is not formally related to the density fluctuations. Standard considerations of the t ccf are as follows [11, 2, 13].

The transverse current, \( \mathbf{j}^\perp(t) \), and the transverse current correlation function, \( C(k, t) \) are defined as:

\[
\mathbf{j}^\perp(t) = \frac{1}{N} \sum_{i=1}^{N} m_i \mathbf{v}^\perp_i(t) \exp(i \mathbf{k} \mathbf{r}_i(t)) ,
\]

\[
C(t, k) \equiv \langle \mathbf{j}^\perp(t) \mathbf{j}^\perp(-k, 0) \rangle , \quad \mathbf{v}^\perp_i \equiv \mathbf{v}_i - \mathbf{k} \mathbf{v}_i .
\]
associated with the wavevector and frequency dependent viscosity \( [1, 2] \):
\[
\eta(\omega, k) = \frac{\rho_m}{k^2 C(\omega, k)} \left[ \omega \tilde{C}(\omega, k) + C(0, k) \right], \quad (A3)
\]
\[
\tilde{C}(\omega, k) = \int_0^\infty C(t, k) \exp(-i\omega t) dt, \quad (A4)
\]
where \( \rho_m = mN/V \) is the average mass density. The \textit{usual} viscosity corresponds to the limit of vanishing frequency and wavevector \( (\omega \to 0, \quad k \to 0) \). In this limit the following expression for viscosity in terms of the \textit{tcvf} can be obtained:
\[
\eta = \beta \rho_m \lim_{\omega \to 0} \lim_{k \to 0} \text{Re} \int_0^\infty \frac{C(t, k)}{k^2} e^{-i\omega t} dt, \quad (A5)
\]
where \( \beta = (\hbar T)^{-1} \).

Wavevector and frequency dependent viscosity can also be expressed through the correlation function of the macroscopic stress tensor, \( \Pi_{xz}^{\omega,k}(t) \) \( [3, 4] \):
\[
\eta(\omega, k) = \left[ \tilde{N}(\omega, k) \right] / \left[ 1 - \left( k^2 \tilde{N}(\omega, k) \right) / (i\omega \rho_m) \right], \quad (A6)
\]
where
\[
\tilde{N}(\omega, k) = \left( \beta / V \right) \int_0^\infty \langle \Pi_{xz}^{\omega,k}(t) \Pi_{xz}^{\omega,k}(0) \rangle e^{-i\omega t} dt \quad (A7)
\]
and it is assumed that \( k \) is parallel to the z-axis. The expression for the stress tensor in (A7) is \( \Pi \):
\[
\Pi_{xz}^{\alpha\beta}(t) = \sum_{i=1}^N s_{i}^{\alpha\beta}(t, k), \quad (A8)
\]
where \( s_{i}^{\alpha\beta}(t, k) \) is the atomic level stress element:
\[
s_{i}^{\alpha\beta}(t) = \left[ m v_i^\alpha v_i^\beta \sum_{j \neq i} \frac{s_{ij}^{\alpha\beta}}{r_{ij}^3} \Phi_k(r_{ij}) \right] e^{-ikr_i}, \quad (A9)
\]
and
\[
\Phi_k(r_{ij}) = \frac{r_{ij}}{2} \left[ \frac{\partial U(r_{ij})}{\partial r_{ij}} \right] \left[ e^{ikr_{ij}} - 1 \over ikr_{ij} \right]. \quad (A10)
\]

In liquids at low temperatures the first term on the right hand side of (A9) is much smaller than the second term and can be neglected [1].

In the limit \( k \to 0 \) from (A6-A7) we get:
\[
\eta(\omega) = \frac{\beta}{V} \int_0^\infty < \Pi_{xz}^{\omega}(t) \Pi_{xz}^{\omega}(0) > e^{-i\omega t} dt. \quad (A11)
\]

For \( \omega = 0 \) expression (A11) is the standard Green-Kubo formula.

It can be seen from (A7-A8-A9-A10), that \( \tilde{N}(k, \omega) \) can be decomposed into contributions from different atomic level stress elements. In Ref.\([29, 30]\) we studied the properties of this decomposition for \( k = 0 \). Thus we introduced there a function \( F(r, t) \):
\[
\langle \Pi_{xz}^{\omega}(t) \Pi_{xz}^{\omega}(0) \rangle = \int_0^\infty F(r, t) dt. \quad (A12)
\]

In this paper, we investigate features of \( F(r, t) \) by performing the Fourier transform of \( F(r, t) \) into \( F(q, t), \) \( F(r, \omega), \) \( \tilde{F}(q, \omega) \).

We would like to emphasize that the wavevector \( q \) that we introduce in our present investigation is \textit{distinct} from the wavevector \( k \) that is usually introduced in consideration of the generalized viscosity. Formally all our results correspond to the case \( k = 0 \), i.e., to the case of very large wave lengths of density fluctuations. This limit effectively corresponds to the case when local density fluctuations are absent. See also discussion on the transverse current correlation function in Ref.\([33]\).

**Appendix B: The origin of the \textcolor{red}{\textbf{bonfire feature}}**

In order to understand the \textcolor{red}{\textbf{bonfire feature}} in panels (3,1) and (3,2) of Fig.\([1]\) we adopt an \textit{ad hoc} approach. In particular, in performing the Fourier transforms, we integrate from \( r = 0 \) (Å) up to some maximum value \( R_{\text{max}} \) and check how the value of \( R_{\text{max}} \) affects the Fourier image. The results are shown in Fig.\([11]\).

Panel (a) shows the results of the integration up to \( r = 20 \) (Å). Note the similarities and differences between the intensity in panel (a) and the intensity in panel (3,2) of Fig.\([1]\). For panel (b) \( R_{\text{max}} = 10 \) (Å). By comparing panels (a) and (b) note that the negative intensity region close to \( q = 0 \) (Å\(^{-1}\)) present in panel (a) is gone in panel (b). Similarly gone is the part of the \textit{sickle feature} that apparently originates from the part of the shear stress wave that we do not count when we integrate up to \( R_{\text{max}} = 10 \) (Å). However, the \textcolor{red}{\textbf{bonfire feature}} is still there, even though it is affected in the transition from (b) to (c). In panel (c) the negative intensity region around \( q = 0 \) (Å\(^{-1}\)) appears again. Thus, from the comparison of panels (a), (b) and (c) we conclude that the negative intensity close to \( q = 0 \) (Å\(^{-1}\)) is related to the spatial extent of the \textit{sccf} . Further note that the \textcolor{red}{\textbf{bonfire feature}} is still present in panels (c) and (d). The transition from panel (d) to panel (c) affects the \textcolor{red}{\textbf{bonfire feature}} very significantly. It follows from panel (a) of Fig.\([7]\) in Ref.\([29]\) that the region between \( 4 \) (Å) and \( 5 \) (Å) corresponds to the interval of the pair density function occurs. Overall, we conclude that the \textcolor{red}{\textbf{bonfire feature}} is related to the absence of periodicity in the \textit{sccf} for \( r < 10 \) (Å\(^{-1}\)). Comparison of panels (1,2) and (3,2) of Fig.\([1]\) also hints that this conclusion is correct. Thus, one may notice that the temporal extent of the \textcolor{red}{\textbf{bonfire feature}} in (3,2) corresponds to the temporal extent of the bright vertical stripes in the region \( r < 10 \) (Å). The last idea can also be tested on the \textit{sccfs} and their Fourier transforms at higher temperatures, as can be seen in Fig.\([8, 10]\). Thus a comparison of panels (1,1)
FIG. 11. Explanation of the bonfire feature in the \( r \rightarrow q \) Fourier transform of the sscf. See panels (3,1) and (3,2) of Fig.1. Different values of the upper cutoff, \( R_{\text{max}} \), in the Fourier transform are shown in the panels.

and (1,3) of Fig.12 shows that the bright stripe present in (1,1) at 5 (Å) is significantly less pronounced in panel (1,3). The comparison of the corresponding panels in Fig.10 shows that the bonfire feature is present in (1,1), but nearly absent in (1,3).

Appendix C: The tongue feature

In order to understand the tongue feature present in panels (3,1) and (3,2) of Fig.1 we again adopt an ad hoc approach. Thus we create several model sscfs and, by comparing the Fourier transforms of these sscfs, we demonstrate that the line shape of the tongue feature is affected by the behavior of stress waves at distances at which the stress waves appear.

It is shown in Ref.67 that in viscous liquids in spherically symmetric homogeneous cases the pressure profile far away from the origin is given approximately by:

\[
p'(r, \tau) = p'_o \frac{\tau}{r^{5/2}} \exp\left[-\frac{\tau^2}{4ar}\right], \tag{C1}
\]

where \( p'(r, \tau) \) is the deviation of pressure from its average value in the system caused by the wave, \( \tau \equiv t - r/c \), \( c \) - is the speed of the wave, \( r \) is the distance from the origin of the wave, and \( a \) controls the rate of the dissipation of the wave. We use the functional form (C1) to create the tongue feature. This does not mean that we assume that (C1) correctly describes the shape of the shear stress waves. However, we believe that this approach allows to determine the origin of the tongue feature.

Panel (1,1) of Fig.12 shows the pressure profile calculated using (C1) with the following values of the parameters: \( c = 30 \) (Å/fs), \( 4a = 2500 \) (fs²/Å), and \( p'_o = 1 \). We use (C1) to calculate the pressure profile for \( r > 2 \) (Å). We assume that for smaller distances the pressure is zero. For smoothness we also convolute the function with the Gaussian function of width \( \sigma_r = 0.50 \) (Å) along the \( r \)-axis and with width \( \sigma_t = 50 \) (fs) along the \( t \)-axis.

Panel (1,2) shows the Fourier transform of panel (1,1) into \((t, q)\)-space. We see in (1,2) the analogue of the tongue feature and also the sickle feature. Note, however, that close to the origin the tongue feature has a negative intensity. We found that this negative intensity could be removed by assuming that the wave starts not at \( r = 0 \) (Å), but instead at some finite distance. Thus, panel (2,1) of Fig.12 shows a pressure wave in which instead of \( \tau \equiv t - r/c \) we used \( \tau = t - (r - r_b)/c \) with \( r_b = 3.3 \) (Å).

As before we assumed that for \( r < 2 \) (Å) the pressure is zero and we convoluted the function with the parameters
given above. Panel (2,2) shows the Fourier transform of (2,1) in \((t, q)\)-space. Note that compared to panel (1,2) there is no negative intensity around the origin in panel (2,2). The comparison also could be made with panel (2,1) of Ref. [38].

Panel (3,1) shows a pressure profile which is a sum of two waves. One wave is exactly the same as in panel (2,1). The second wave also starts at 3.3 \((\AA)\) and has the following values of the parameters: \(c = 60 \,(\AA/\text{fs}), \)
\[ 4a = 1000 \,(\text{fs}^2/\AA) \] and \(\rho_o'' = 3\). We see in panel (3,2) that the second wave makes the “sickle” feature much more pronounced. Note also the increased intensity and the increased width of the positive intensity region around \(q = 0\,(\AA^{-1})\).

Thus we conclude that the line shape of the tongue feature results from the behavior of the stress waves near their origin.

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[39] If the parameters of the Lennard-Jones (LJ) potential are chosen in such a way that the distance at which the LJ potential is zero is the same as the distance at which our potential is zero then \(\sigma \approx 2.2245 \,(\AA)\). Let us also assume that the depth of the LJ potential is the same as the depth of our potential, i.e., \(\epsilon \approx 0.2558 \,(eV)\). With this choice of parameters, the minimum of the LJ potential occurs at \(r_{min}^{\text{LJ}} = 2^{1/6} \sigma \approx 2.4969 \,(\AA)\), while the minimum of our potential is at \(r_{min}^{\text{our}} \approx 2.6166 \,(\AA)\). If the radial density were the delta function located at \(r_{min}^{\text{our}}\), the corresponding first peaks in the structure function \(S(q)\) would occur (because of the spherical Fourier transform) at the first maximum of the function \(\sin(qr_{min}^{\text{our}})/(qr_{min}^{\text{our}})\), i.e., at \(q_{0}r_{min} \approx 7.73\). Thus, \(q_{0} \approx 2.95 \,(1/\AA)\). The reduced value of the wavevector associated with this peak is \(q_{0}^{\text{r}} \equiv q_{0}\sigma \approx 7.37\).

The characteristic time that follows from the chosen parameters of the LJ potential (assuming that \(m\) is the mass of an iron atom) is: \(\tau_{c} = (m/e)^{1/2} \sigma \approx 337.4 \,(\text{fs})\). Thus, characteristic frequency is: \(\tau_{c}^{-1} \approx 1/\tau_{c} \approx 0.00296 \,(1/\text{fs})\).

The particle number density in our simulations, expressed in terms of the reduced density of the LJ potential, is: \(\rho_{our} = \rho_{our} \sigma^{3} \approx 0.8634\). Here, in particular, we report about simulations made at temperatures 1500 \,(K) and 2000 \,(K). These temperatures expressed in the reduced units, \((T^* = k_{B}T/\epsilon)\), are: \(T_{our}[1500(K)] = 0.5053\) and \(T_{our}[2000(K)] = 0.6737\). The triple point of the LJ potential at which simulations are often done \([4]\) corresponds to \(\rho_{o}^{*} \approx 0.844\) and \(T_{o}^{*} \approx 0.72\,[23]\).

The potential energy landscape crossover temperature of this system is \(T_{A} \approx 2300 \,(K)\,[13]\). The mode coupling temperature is \(T_{MC} \approx 1150 \,(K)\), and the glass transition temperature is \(T_{g} \approx 950 \,(K)\).

The characteristic value of viscosity for the LJ potential
is: \( \eta^* = (m \epsilon)^{1/2} / \sigma^2 \approx 1.2355 \times 10^{-3} \) (Pa s). The value of viscosity that we obtained in simulation with our potential at 1500 (K) is \( \eta(1500(\text{K})) \approx 29.33 \times 10^{-3} \) (Pa s), while at 2000 (K) it is \( \eta(2000(\text{K})) \approx 10.74 \times 10^{-3} \) (Pa s). In reduced units they are \( \eta^*(1500(\text{K})) = \eta(1500(\text{K}))/\eta^* \approx 23.7 \) and \( \eta^*(2000(\text{K})) \approx 8.7 \). For comparison the value of viscosity for the LJ liquid at the triple point is \( \eta_{\text{LJ}}(\text{triple}) \approx 3.2 \) \([23]\).

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