Cutoff wave number for shear waves and Maxwell relaxation time in Yukawa liquids

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Because liquids cannot resist shear except over very short distances comparable to the atomic spacing, shear sound waves (i.e., transverse phonons) propagate only for very short wavelengths. A measure of this limit is the cutoff wave number \( k_c \), which is sometimes called the critical wave number. Previously \( k_c \) was determined in molecular dynamics (MD) simulations by obtaining the dispersion relation. Another approach is developed in this paper by identifying the wave number at the onset of a negative peak in the transverse current correlation function. This method is demonstrated using a three-dimensional MD simulation of a Yukawa fluid, which mimics dusty plasmas. In general, \( k_c \) is an indicator of conditions where elastic and dissipative effects are approximately balanced. Additionally, the crossover frequency for the real and imaginary terms of the complex viscosity of a dusty plasma is obtained; this crossover frequency corresponds to the Maxwell relaxation time.

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I. INTRODUCTION

Solids, unlike gases, can sustain two kinds of sound waves (phonons): compressional and transverse. The compressional wave is longitudinal, like a sound wave in air, with a periodic compression and rarefaction and wave propagation in a direction parallel to the collective particle velocities. The transverse wave has shear motion, with wave propagation perpendicular to collective particle velocities. The ability of a medium to sustain a shear wave requires an elastic response to a disturbance, where a particle tends to be restored to its equilibrium position after being disturbed.

A liquid, unlike a solid, exhibits elastic responses only for a limited time and over a limited distance. This is so because the potential landscape surrounding a particle in a liquid does not remain unchanged indefinitely as in a solid. When neighboring particles rearrange their positions irreversibly, there will also be an irreversible change in the confining potential landscape, and energy will be dissipated. Until a rearrangement occurs, a disturbance in the position of one particle can be restored elastically.

One approach used in the theory of liquids for characterizing the conditions for elastic vs dissipative motion is based on the propagation of the shear wave. Shear waves with a wave number less than a minimum wave number \( k_c \) cannot propagate [1]. Theoretical authors have referred to this minimum wave number as a “critical wave number” or a “cutoff wave number,” and we will adopt the latter term. Typically \( k_c \) has a value of \( k_c a \approx 1 \), within a factor of 3, depending on temperature and other parameters of the liquid. Here, \( a \) is a measure of interparticle distances in a liquid, defined as the Wigner-Seitz radius \( a = (3/4\pi n)^{1/3} \) for a number density \( n \).

The cutoff wave number has practical importance not only in simple liquids, but also in strongly coupled plasmas like those that we study here, and in supercooled liquids [2]. The cutoff wave number has been observed identified in molecular dynamics (MD) simulations including [3–5] for three-dimensional (3D) liquids and [6] for 2D liquids. A direct observation of the cutoff wave number has been made, to our knowledge, in one experiment [7], which used a dusty plasma. A dusty plasma is an ionized gas containing charged particles of solid matter [8–10]. Dusty plasma experiments have also been reported to characterize static viscosity [11] and viscoelastic effects [12,13].

Previous methods of determining \( k_c \) include examining a dispersion relation \( \omega versus k \) of the shear waves. By tracking particle positions and velocities in an MD simulation, one can compute a phonon spectrum, i.e., a graph of wave energy as a function of \( \omega \) and \( k \). In such a graph, energy is concentrated in a band in \( \omega-k \) space. Drawing a curve along the peaks of this band yields the dispersion relation. For a liquid, this dispersion relation curve does not extend down to \( \omega = 0 \) and \( k = 0 \) as it does for a solid, but instead ends at an intercept on the \( \omega = 0 \) axis. The previous method of determining \( k = k_c \) is by extrapolating the observed dispersion relation curve to this intercept, as has been done for liquids in three [5] and two dimensions [14]. We present in Fig. 1 an example of such a dispersion relation, as determined from the 3D MD simulation reported in this paper. The simulation method and parameters are described in Sec. II.

In this paper, we develop another quantitative method to determine the cutoff wave number \( k_c \). This method is based on identifying a negative peak in a correlation function and extrapolating to the condition where this peak vanishes. We present this method and demonstrate its usefulness over a wide range of temperature using MD simulation data for a liquid strongly coupled plasma in Sec. III-D.

A strongly coupled plasma is a collection of free charges that have an interparticle potential energy that is greater than the thermal kinetic energy. Such a strongly coupled plasma can have the structure of a solid or a liquid, depending on whether the temperature is below or above a melting point [15]. It is common to describe a strongly coupled plasma by the dimensionless Coulomb coupling parameter

\[
\Gamma = \frac{Q^2}{4\pi \epsilon_0 Q_k B T},
\]

where \( Q \) is the charge, \( \epsilon_0 \) is the permittivity of free space, \( a \) is the Bohr radius, and \( T \) is the temperature. Physically, \( \Gamma \) is the ratio of the potential energy to the thermal energy.
where \( Q \) is the charge of the particles and \( T \) is the temperature. When Debye screening occurs, the electric potential of a point charge can be described as the Yukawa (Debye-Hückel) potential energy \( \phi = (Q^2/4\pi\epsilon_0r)\exp(-r/\lambda) \), where \( \lambda \) is a screening length. In this case, another dimensionless parameter is the screening parameter

\[
\kappa = a/\lambda.
\]  

Time scales in a strongly coupled plasma are characterized by the plasma frequency

\[
\omega_p = (nQ^2/\epsilon_0m)^{1/2},
\]  

where \( m \) is the mass of a particle.

We will use the Yukawa potential, which is used often to describe dusty plasmas [16] as well as charged colloidal suspensions. A Yukawa system can behave as a liquid, as has been studied in both simulations and experiments, of which we will mention a few. The melting point of a 3D Yukawa system has been determined in MD simulations [17] to be \( T_{\text{melt}} = 217.4, 440.1, \) and 1185 for \( \kappa = 1, 2, \) and 3, respectively. The complex viscosity \( \eta(\omega) \) has been characterized in 3D simulations [18] and a 2D dusty plasma experiment [19].

In addition to determining \( \kappa \), we will also use our simulation to determine the frequency-dependent viscosity of a Yukawa fluid. The frequency-dependent viscosity

\[
\eta(\omega) = \eta'(\omega) - i\eta''(\omega)
\]  
is a formalism to describe two characteristics of viscoelastic effects: the real part \( \eta' \) indicates dissipation while the imaginary part \( \eta'' \) indicates elasticity, i.e., energy storage. As we will discuss in Sec. III, there is a frequency \( \omega_{\text{cross}} \) for which the curves for \( \eta'(\omega) \) and \( \eta''(\omega) \) cross over, and the inverse of this crossover frequency is the Maxwell relaxation time \( \tau_M \). We will present results for the frequency-dependent viscosity and Maxwell relaxation time for a Yukawa fluid in Sec. III C.

II. SIMULATION

A. Method

We use the equilibrium molecular dynamic simulation method as described in [18]. To imitate an infinite liquid, the simulation has periodic boundary conditions, which is suited for studying the intrinsic properties of a substance. This differs from the approach of simulations in a bounded system [20]. In a system containing \( N \) particles, we integrate the equation of motion of a particle \( i \),

\[
m \frac{dv_i}{dt} = -Q \sum_{j \neq i} \nabla \phi_{ij},
\]  
where \( \phi_{ij} \) is the potential for a binary Yukawa interaction at the position of particle \( i \) due to particle \( j \). We use the resulting time series data for positions and velocities of particles to calculate the transverse microscopic current

\[
\tau_{xy}(k,t) = \sum_{j=1}^{N} v_{xj} e^{iky_j}
\]  
and the off-diagonal elements of the shear stress tensor

\[
P_{xy}(t) = \sum_{i=1}^{N} \left[ m v_{ix} v_{iy} - \frac{1}{2} \sum_{j \neq i} \frac{x_{ij} y_{ij}}{r_{ij}^3} \frac{\partial \phi(r_{ij})}{\partial r_{ij}} \right].
\]

When we present our results in Sec. III, it will be useful to know that in Eq. (7) the first term on the right-hand side arises from kinetic effects that are dominant for liquids at high temperatures, while the second term arises from potential effects that are dominant at low temperatures [21]. These potential effects include caging [22,23] and decaging [24] of particles.

We then calculate the transverse current autocorrelation function

\[
C_T(k,t) = \langle \tau_{xy}(k,t) \tau_{xy}^*(k,t) \rangle
\]
and the shear stress autocorrelation function

\[
C_q(t) = \langle P_{xy}(t) P_{xy}(0) \rangle.
\]

We can use Eq. (9) in the generalized Green-Kubo relation

\[
\eta(\omega) = \frac{1}{VkT} \int_0^\infty C_q(t) e^{i\omega t} dt
\]  
to calculate the complex viscosity, Eq. (4). Here, \( V = L^3 \) is the simulation volume. As in [18], we replace the upper limit in Eq. (10) with the time when the correlation function first crosses zero. Equation (10), which can be derived from expressions in [25], includes a complex exponential so that
it is a generalization of the usual Green-Kubo relation. The latter is known to predict viscosity values that agree with those obtained by hydrodynamic schemes for steady-state conditions.

**B. Parameters**

The simulated cell contains $N = 8000$ identical particles in a cube with sides of length $L \approx 32.24a$. This length defines a minimum resolvable wave number $2\pi/L \approx 0.195a^{-1}$. The time step is chosen as $\Delta t \leq (\pi/15)a_p^{-1}$. The simulation is run for an initial thermalization period of $10^5$ time steps, and then data are recorded for $10^4$ time steps. We repeat the simulation over a wide range of the Coulomb coupling parameter $\Gamma < \Gamma_{\text{melt}}$ and three values of the screening parameter $\kappa = 1, 2, \text{and } 3$.

**C. Dimensionless variables**

We report results that are in dimensionless units, as indicated by bars over the symbols. These are the length $ar{r} = r/a$, wave number $\bar{k} = ka$, correlation functions $\bar{C}(t) = C(t)/C(0)$, viscosity $\bar{\eta} = \eta/(m\omega_p a^2)$, and frequency $\bar{\omega} = \omega/\omega_p$. For some results we will also report frequencies normalized as $\omega/\omega_E$, where $\omega_E$ is the Einstein frequency, whose values are reported in Table I of [17]. We make time $t$ dimensionless by multiplying by the plasma frequency $\omega_p t$. Other dimensionless variables are $\Gamma$ and $\kappa$ as defined in Eqs. (1) and (2), respectively.

**III. RESULTS AND DISCUSSION**

**A. Dispersion relation**

A method used by previous authors to identify a cutoff wave number is the measurement of the dispersion relation. One way of doing this is to perform an MD simulation to record the positions and velocities of particles undergoing thermal motion and use these data to compute the power spectrum in the $\omega-k$ space. The peaks of this power spectrum correspond to the wave dispersion relation.

An example of a dispersion relation is presented in Fig. 1, based on our MD simulation. The previous method of determining $k_c$ is to extrapolate these dispersion relation curves to their intercept on the $\omega = 0$ axis.

**B. Cutoff wave number: Transverse current correlation method**

An example of the transverse current correlation function $C_T$ for a liquid at a temperature nearly twice as high as the melting point, for various wave numbers $k$. The hydrodynamic limit is at small wave numbers while at larger wave numbers there are oscillations. The hydrodynamic limit is at small wave numbers while at larger wave numbers there are oscillations. The vertical axis of (a) is normalized by the value of the correlation function at zero time, $\bar{C}(t) = C(t)/C(0)$.

![Figure 2](image_url)

**FIG. 2.** (Color online) (a) Example of transverse current autocorrelation function $C_T$ for a liquid at a temperature nearly twice as high as the melting point, for various wave numbers $k$. The hydrodynamic limit is at small wave numbers while at larger wave numbers there are oscillations. (b) Method of determining the cutoff wave number $k_c$. We measure the amplitude $A$ of negative peaks of $C_T$ in (a) for various values of $k$, and plotting these we extrapolate to find the value of $k$ where the amplitude just touches a zero value, i.e., where the curve for $C_T$ would barely have a negative oscillation in the absence of noise; we report this value of $k$ as our measure of $k_c$. The vertical axis of (a) is normalized by the value of the correlation function at zero time, $\bar{C}(t) = C(t)/C(0)$.

We also find that, when normalized, the values for the cutoff wave number fall on a universal curve (see Fig. 3). This curve is approximately $k_c = \frac{1}{4}(\Gamma/\Gamma_{\text{melt}})^{-4/3}$. To make the data fall on this curve, we normalized $k_c$ by $a$ and $\Gamma$ by the melting point $\Gamma_{\text{melt}}$.
As a validation, we compared our results for the cutoff wave numbers to those reported by Murillo [28] and by Hamaguchi and Ohta [5]. They both determined $k_c$ from a dispersion relation, which they obtained by different means. Murillo used a hydrodynamic method, while Hamaguchi and Ohta used an MD simulation. We find general agreement when comparing our values and theirs. This agreement is improved by dividing $k_c$ by a factor of $\sqrt{2}$, which is reasonable considering Murillo’s argument that the dispersion relation methods generate a value of $k_c$ that is larger than the true onset of waves by a factor of $\sqrt{2}$ [28]. Our method detects the onset of waves.

C. Crossover frequency: Complex viscosity method

For comparison, we present here another indicator of the balance of elastic and dissipative effects, characterized by a frequency instead of a wave number, and based on viscosity instead of waves. This is done by exploiting the frequency-dependent viscosity Eq. (4), which has a real part $\eta'(\omega)$ that corresponds to viscous dissipation, and an imaginary part $\eta''(\omega)$ that corresponds to elasticity, i.e., energy storage.

Here, we do not investigate the full dependence of $\eta$ on both $\omega$ and $k$ because our Green-Kubo method yields valid results only with $k = 0$ [25]. We also note that a dependence on only frequency is what is typically measured in experiments, for example using rheometers that agitate a liquid by rotating at a specified frequency $\omega$. Besides our Green-Kubo method, there exist other theoretical methods that have been used to generate the full frequency and wave-number-dependent viscosity, which we do not attempt to use [29].

In the theory of liquids, the viscoelastic approximation [30] predicts the real and imaginary parts of viscosity as

$$\eta'(\omega) = \frac{\eta}{1 + \omega^2 \tau_M}, \quad (11)$$

$$\eta''(\omega) = \frac{\eta \omega \tau_M}{1 + \omega^2 \tau_M^2}, \quad (12)$$

where $\eta$ is the usual static viscosity for $\omega \to 0$, and $\tau_M$ is the Maxwell relaxation time [30]. The real part $\eta'$ is larger at low frequencies, while the imaginary part $\eta''$ is larger at high frequencies. Inspecting Eqs. (11) and (12), we see that the real and imaginary parts are equal, i.e., their curves cross over, at $\omega = 1/\tau_M$.

This prediction for the viscoelastic approximation, a crossover of the real and imaginary terms at a physically significant frequency of $1/\tau_M$, leads us to inspect graphs of the complex viscosity from our MD simulation, produced using Eq. (10).

An example of the complex viscosity and the crossover of its real and imaginary terms is shown in Fig. 4. The data points are our simulation results. Note that the real part $\eta'$ diminishes monotonically with frequency, so that its curve crosses that of the imaginary part $\eta''$. In this example, for $\Gamma = 125$ and $\kappa = 1$, the crossover occurs at $\omega_{\text{crossover}} = 0.240 \omega_p$. We interpret $1/\omega_{\text{crossover}} = 4.17 \omega_p^{-1}$ as an empirical measure of $\tau_M$, for the parameters of Fig. 4.
Also shown in Fig. 4 is a fit of our MD simulation data to the viscoelastic approximation Eqs. (11) and (12). This fit, which has only two free parameters, \( \eta \) and \( \tau_M \), shows good agreement. The fit yields \( \tau_M = 4.15 \omega_p^{-1} \), for the parameters \( \Gamma = 125 \) and \( \kappa = 1 \) of Fig. 4. The fit is good, as indicated by an agreement within 0.5% when compared to \( 1/\omega_{\text{cross}} \) for the simulation data points.

We repeat this calculation of the crossover frequency \( \omega_{\text{cross}} \) for a wide range of \( \Gamma \) and \( \kappa \), and the result is shown in Fig. 5(a). By normalizing the axes differently in Fig. 5(b), as \( \omega/\omega_E \) vs \( \Gamma/\Gamma_{\text{melt}} \), we find that the curves for various screening parameters \( \kappa \) are similar.

The curves for \( \omega_{\text{cross}} \) in Fig. 5 have a peak. We verified that this peak nearly coincides with a minimum of viscosity. This minimum of viscosity is a well-known phenomenon for liquid strongly coupled plasmas. It occurs between low temperatures where the potential terms in Eq. (7) dominate and high temperatures where the kinetic terms dominate [21]. For the purpose of understanding viscoelasticity, only the high-\( \Gamma \) regime is of interest, which in Fig. 5(b) is for \( \Gamma/\Gamma_{\text{melt}} > 0.1 \). In this case the viscosity is dominated by potential effects, and particles can be caged for a finite time between their nearest neighbors, as is required for elasticity. This is the same range of \( \Gamma \) where we found oscillations in the transverse current correlation function that indicate the presence of shear waves, as indicated in Fig. 3.

Within the regime \( \Gamma/\Gamma_{\text{melt}} > 0.1 \) that is meaningful for viscoelasticity, we find a trend that the crossover frequency diminishes with increasing \( \Gamma \). We can suggest an intuitive explanation for this trend. For this relatively low-temperature range, viscous effects are dominated by the second term of Eq. (7), which arises from potential energy terms that can result in caging. Consider that \( \omega_{\text{cross}} \) indicates a balance of dissipative and elastic effects, and that dissipation involves the slipping of particles out of a cage. This decaging requires a displacement that is a significant fraction of the particle spacing. Since the particle has a finite velocity, characterized by the thermal velocity, this displacement requires a finite time. At a high frequency \( \omega \), there is too little time, \( 1/\omega \), for this to occur, and elastic effects will dominate. Only at a sufficiently low frequency \( \omega < \omega_{\text{cross}} \) will there be sufficient time for a particle to slip enough to decage. As the temperature is decreased (i.e., \( \Gamma \) is increased), the thermal velocity is reduced and the time required for slipping becomes longer. Thus, we generally expect \( \omega_{\text{cross}} \) to diminish as \( \Gamma \) is increased. This is the trend that is observed in our data in Fig. 5, for \( 0.1 < \Gamma/\Gamma_{\text{melt}} < 1 \).

**D. Cutoff wave number vs crossover frequency**

We have developed a quantitative method of measuring the cutoff wave number determined by extrapolating a negative peak in a correlation function to the point at which it vanishes, as an indication of a balance of dissipative and elastic effects as a function of \( \Gamma \). As another method of indicating a balance of dissipative and elastic effects, we also presented results for a better-known method, the complex viscosity, which has two terms that cross at a frequency that we find. We can combine these results from Figs. 3 and 5 into a single graph of \( \omega_{\text{cross}} \) vs \( k_c \), in Fig. 6.

In Fig. 6, the data points in the lower left-hand corner are for low temperatures near the melting point, i.e., high \( \Gamma \), while those in the upper right are for high temperature. Beyond the last data points in the upper right, the effects of elasticity vanish.

The significance of any given data point in Fig. 6(a) is that for its value of \( \Gamma \) and \( \kappa \), the data point marks a frequency \( \omega \) and a wave number \( k \) where elastic and dissipative effects balance. Elastic effects dominate for the parameter space \( k \) to the right and \( \omega \) above the data point, while dissipative...
effects dominate to the left and below the data point. For lower temperatures, i.e., for data points nearer the origin in Fig. 6, a greater portion of the $\omega$-$k$ parameter space is dominated by elastic effects, as compared to higher temperatures. This temperature dependence is the reason that the data points are distributed along a curve with a distinctive trend in Fig. 6.

All the data points in Fig. 6 are for a liquid. A crystalline lattice at zero temperature would appear at the origin in this graph. This is so because dissipation is lacking and elastic effects dominate, for all values of $\omega$ and $k$ in a perfect crystal.

To avoid confusion, we should mention that despite its appearance as a graph of a frequency vs a wave number, Fig. 6 is not a dispersion relation. The vertical axis $\omega_{\text{cross}}$ has no direct relation to waves.

IV. CONCLUSIONS

We have developed a method of determining the cutoff wave number for shear waves, by detecting the presence of an oscillation in the transverse current correlation function $C_T(t)$. This is a sensitive method of detecting the onset of shear waves, and it allows a determination of the cutoff wave number. This method is useful over a wide range of temperatures, even as hot as ten times the melting point for the 3D Yukawa liquid that we studied. We obtained the data for this test using an equilibrium MD simulation. We empirically found the temperature dependence, i.e., the dependence on the coupling parameter $\Gamma$, to be $k_c = \frac{1}{3}(\Gamma/\Gamma_{\text{melt}})^{-4/3}$.

For comparison, we have also presented results from another method of identifying a balance of the effects of viscous dissipation and elasticity: a determination of the crossover frequency for the real and imaginary parts of the frequency-dependent complex viscosity. This method, which in principle could be useful for experiments as well as simulations, relies on the meaning of the real part of the viscosity as an indicator of dissipation and the imaginary part as an indicator of elasticity. The inverse of this crossover frequency is an empirical measure of the Maxwell relaxation time, which is a well-known parameter in viscoelastic theory that we determined for a dusty plasma.

This crossover frequency method yields results over a wide range of temperature, although for the purposes of understanding the elastic part of viscoelasticity it is likely meaningful only for the same temperature range as our cutoff wave number method. That range is the one where the potential terms dominate the viscosity, which for our 3D Yukawa liquid is roughly $0.1 < \Gamma/\Gamma_{\text{melt}} < 1$.

Note added in proof. An agreement of the Green-Kubo method and a hydrodynamic method of computing the shear viscosity was demonstrated recently for Yukawa liquids by Mithen et al. [31]. For the hydrodynamic limit, they also confirmed the monotonic decay of the transverse current autocorrelation function $C_T$ for a Yukawa liquid.

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