

Tracking shear mode dynamics across the glass transition in a 2D colloidal system

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Long-wavelength collective shear dynamics are profoundly different in solids and liquids. According to the theoretical framework developed by Maxwell and Frenkel, collective shear waves vanish upon melting by acquiring a characteristic wave-vector gap, known as the k -gap. While this prediction has been supported by numerous simulations, experimental validation remains limited. In this work, we track the dispersion relation of collective shear modes in a two-dimensional colloidal system and provide direct experimental evidence for the emergence of a k -gap. This gap appears at an effective temperature consistent with the onset of the glass transition and the vanishing of the static shear modulus. Our results not only confirm the predictions of the Maxwell-Frenkel framework but also highlight their relevance across continuous melting processes originating from low-temperature amorphous solid phases.

At low wave vectors, collective shear dynamics in solids are governed by propagating shear waves (phonons), with their speed determined by the static shear modulus [1]. In contrast, in liquids, these dynamics are dominated by diffusive processes governed by the finite shear viscosity [2].

Within the theoretical frameworks established by Maxwell and Frenkel [3], the transition between these two regimes is governed by the telegrapher equation:

$$\omega_T^2 + i\omega_T/\tau = v^2 k^2, \quad (1)$$

where ω_T is the frequency of collective transverse excitations, k the wave vector, v the instantaneous speed of sound and τ a characteristic relaxation time. In Frenkel's microscopic picture of liquid dynamics [4], τ represents the average time a particle takes to hop over potential barriers, that can be related to the lifetime of local atomic connectivity [5]. In contrast, Maxwell's theory [6] identifies τ as the Maxwell relaxation time, which characterizes the macroscopic viscoelastic response of the medium.

Equation (1), which arises in a wide range of physical systems [7], predicts that collective shear excitations in liquids exhibit a characteristic dispersion:

$$\text{Re}(\omega_T) = v\sqrt{k^2 - k_g^2}, \quad (2)$$

where $k_g \equiv 1/(2v\tau)$ is the so-called k -gap, characterizing the inverse length scale below which elastic response persists. In the long-wavelength limit, $k \ll k_g$, $\text{Re}(\omega_T) = 0$ and the dynamics are liquid-like (diffusive).

In the other limit, $k \gg k_g$, solid-like response and propagating shear waves emerge also in liquids. This reflects the high-frequency shear modulus that makes diving from a 10-meter platform so exciting, while swimming gently through water remains so relaxing.

Although the precise definition of τ in Eq. (1) remains debated, it is generally expected that k_g vanishes at the melting temperature, below which the standard wave-like dispersion $\omega_T = vk$, expected in solids, is recovered. Since the initial validation of these theoretical predictions in simulated molecular liquids and supercritical fluids [8], the k -gapped behavior described by Eq. (2) has been observed across a wide range of simulated systems, confirming its universality with respect to interparticle interactions (see [3] for a review).

On the other hand, experimentally verifying this mechanism in molecular liquids remains challenging due to limitations of current techniques in accessing the low-frequency, low-wave-vector regime. In [9], the emergence of a cutoff wave number k_g in the liquid-like phase of a two-dimensional Yukawa dusty plasma was observed and estimated as $k_g a \approx 0.16$ – 0.31 , where a is the 2D Wigner-Seitz radius. More recently, [10] reported the existence of a k -gap in the dispersion of collective transverse excitations in the liquid-like phase of athermal vibrated granular matter. However, in both cases, the disappearance of the wave-vector gap at the onset of solidification was not clearly demonstrated. Furthermore, whether this mechanism remains valid across a continuous glass transition from an amorphous solid to a liquid remains an open question.

Here, we study a two-dimensional colloidal glass composed of a binary mixture of super-paramagnetic polystyrene spheres confined to a flat water–air interface [11] (see the inset of Fig. 1(a) for an image of the experimental setup). The small and large particles have

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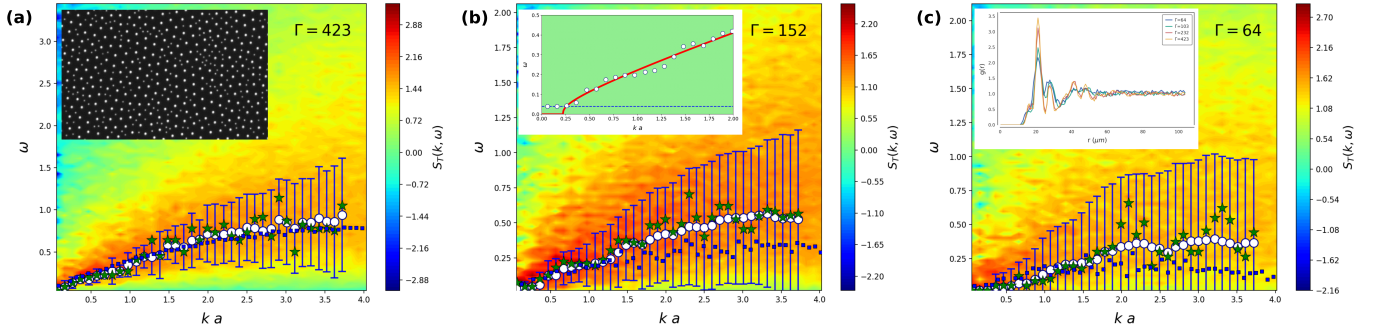


FIG. 1. (a) The dispersion of collective shear modes as a function of the normalized wave-vector ka for $\Gamma = 423$, deep in the amorphous glass phase. White symbols are the value of $\Omega_T(k)$ obtained by fitting the dynamical transverse structure factor $S_T(k, \omega)$ with a Lorentzian function and the error bars indicate the corresponding linewidth $\Gamma(k)$; green symbols are the position of the maxima in $S_T(k, \omega)$; blue symbols are the eigenvalues ω_j obtained diagonalizing the transverse sector of the dynamical matrix $\mathcal{D}(\mathbf{k})$. The background color is the absolute value of $S_T(k, \omega)$ in a logarithmic scale. The inset shows an image of the experimental setup. (b) Similar analysis for $\Gamma = 152$, slightly below the expected glass transition temperature, in the liquid phase. The inset shows an example of the fit of the dispersion relation using the k -gap equation (2), with the horizontal dashed line indicating the cutoff frequency ω_{\min} arising because of the finite size system. (c) Same analysis for $\Gamma = 64$, deep in the liquid phase. The inset shows the value of the pair correlation function $g(r)$ for the three values of Γ reported in this figure.

diameters of $2.8 \mu\text{m}$ and $4.5 \mu\text{m}$, respectively, with a mixing ratio of 45 : 55. The particles interact via a tunable magnetic dipole-dipole interaction, controlled by an external magnetic field and quantified by a dimensionless coupling parameter Γ , which serves as an effective inverse temperature. Particle positions are tracked over time using video microscopy and digital image analysis. The elastic properties of this experimental system have been thoroughly investigated [12], with evidence suggesting the onset of a glass transition at $\Gamma^* \approx 195$ [13].

Following [12], we define the particle displacement $\mathbf{u}_i(t) = \mathbf{r}_i(t) - \bar{\mathbf{r}}_i$, where $\bar{\mathbf{r}}_i$ is the average position of the particle $i \in [1, N]$ during a time interval $\Delta t \approx 18900$ s. We then derive the ‘dynamical matrix’, $\mathcal{D}(\mathbf{k}) = k_B T \langle \mathbf{u}_{\mathbf{k}}^* \mathbf{u}_{\mathbf{k}} \rangle^{-1}$, where $\mathbf{u}_{\mathbf{k}}$ is the Fourier transform of the particle displacement in terms of the wave vector \mathbf{k} and $\langle \cdot \rangle$ indicates time average. The eigenvalues $\lambda_j(\mathbf{k})$ of the dynamical matrix correspond to the squared eigenfrequencies of the system $\omega_j^2(k)$ and provide the spectrum of excitations in the harmonic limit [14].

From the mode analysis, we compute the transverse dynamical structure factor $S_T(k, \omega)$,

$$S_T(k, \omega) \propto \frac{k^2}{\omega^2} \sum_{\lambda} E_{\lambda, T}(\mathbf{k}) \delta(\omega - \omega_{\lambda}), \quad (3)$$

where

$$E_{\lambda, T}(\mathbf{k}) = \left| \sum_j (\hat{\mathbf{k}} \times \mathbf{e}_{\lambda}(j)) \exp(i\mathbf{k} \cdot \mathbf{r}_j) \right|^2. \quad (4)$$

Here, \mathbf{r}_i is the position of the i th particle, $\mathbf{e}_{\lambda}(j)$ is the eigenvector corresponding to eigenfrequency ω_j and $\hat{\mathbf{k}} \equiv \mathbf{k}/|\mathbf{k}|$. To improve the statistics, we average $S_T(k, \omega)$ over different $\hat{\mathbf{k}}$ orientations.

In Fig. 1, we present the experimental dispersion relation of collective shear modes as a function of the dimen-

sionless wave vector ka , where a is the average interparticle distance $\approx 22 \mu\text{m}$ (see inset of Fig. 1(c)), for several representative values of the control parameter Γ , spanning from deep within the amorphous solid phase (panel (a)) to the liquid phase (panel (c)). The background color map shows the absolute value of the transverse dynamical structure factor $S_T(k, \omega)$, plotted on a logarithmic scale to enhance contrast and visibility.

Blue filled symbols indicate the eigenvalues obtained by diagonalizing the dynamical matrix $\mathcal{D}(k)$. Green symbols mark the location of the maxima in $S_T(k, \omega)$ along constant- k cuts. Finally, the white symbols represent the dispersion relation $\Omega_T(k)$ extracted from Lorentzian fits to $S_T(k, \omega)$,

$$S_T(k, \omega) \propto \frac{\omega^2 \Gamma_T(k)}{(\omega - \Omega_T(k))^2 + \omega^2 \Gamma_T(k)^2}. \quad (5)$$

The error bars in Fig. 1 indicate the relative linewidth $\Gamma_T(k)$.

We first observe that the three independent methods yield consistent results for the dispersion of collective shear modes in the solid phase. On the other hand, the method based on the dynamical matrix becomes less reliable in the liquid phase, as the validity of the harmonic approximation is no longer guaranteed. More importantly, we note a progressive evolution in the form of the dispersion relation from panel (a), corresponding to $\Gamma = 423$, to panel (c), which corresponds to $\Gamma = 64$.

In the amorphous solid phase (panel (a), $\Gamma = 423$), shear modes exhibit a wave-like, propagating dispersion, consistent with predictions from elasticity theory and previously investigated in detail in Ref. [12]. In panel (b), at $\Gamma = 152$, slightly below the reported glass transition point $\Gamma^* \approx 195$, in the liquid phase, the dispersion relation undergoes a qualitative change at small wave

vectors ($ka < 0.5$), where the emergence of a gap, consistent with Eq. (2), becomes apparent.

This feature becomes even more pronounced in panel (c), deep in the liquid phase ($\Gamma = 64$), where the k -gap is clearly visible and larger than in panel (b). This trend supports the formation of a wave-vector gap in the dispersion of collective shear modes as the system transitions from the high- Γ amorphous solid to the low- Γ liquid. This transition is further corroborated by the behavior of the pair distribution function $g(r)$ shown in the inset of Fig. 1(c), which reflects the structural changes across the glass transition.

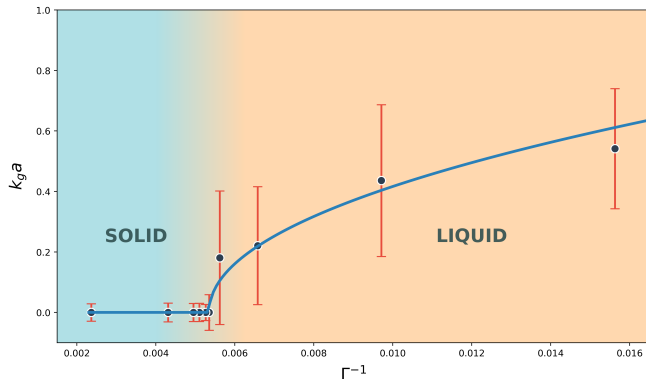


FIG. 2. The dimensionless k -gap $k_g a$ as a function of the control parameter Γ , playing the role of the inverse effective temperature. The solid line corresponds to the best fit, Eq. (6). The background color highlights the transition between the solid phase at large Γ to the liquid state at low Γ .

After validating the emergence of a gap in the dispersion of collective shear modes with decreasing control parameter Γ , we extended our analysis to a broader range of Γ values, spanning those shown in panels (a) to (c) of Fig. 1. For each case, we extracted the corresponding dispersion relation and fitted the low- k region using the k -gap equation (2). An example of this fitting procedure is presented in the inset of Fig. 1(b) for $\Gamma = 152$. We emphasize that due to the finite size of the experimental system, there exist lower bounds for both the wave vector and the frequency, denoted as k_{\min} and ω_{\min} , respectively. These cutoffs can be directly estimated and must be properly accounted for in the analysis. In the inset of Fig. 1(b), k_{\min} has been subtracted from the values on the y -axis, while ω_{\min} is indicated by a dashed horizontal line. Notably, this line aligns well with the frequencies observed in the low- k experimental data below

the k -gap.

This analysis allowed us to determine the wave-vector gap k_g as a function of Γ , or equivalently, the effective temperature $T_{\text{eff}} \propto \Gamma^{-1}$. The extracted values of k_g , along with their associated error bars from the fitting, are plotted in Fig. 2.

We find that the data are well described by the empirical relation:

$$k_g a = 5.62 (T_{\text{eff}} - T_{\text{eff}}^*)^{0.48}, \quad (6)$$

with $T_{\text{eff}}^* \approx 1/189$, which is in close agreement with the glass transition temperature previously reported based on the discontinuity of the static shear modulus, $\Gamma^* \approx 195$ [13].

Interestingly, the power-law exponent reported in Eq. (6) is not far from what one would expect from mean-field critical behavior. However, at this stage, we refrain from making any definitive claims regarding the universality of this exponent.

Nonetheless, we note that the extracted values of the k -gap fall within the range $k_g a \in [0, 0.8]$ for the Γ values explored. This range aligns remarkably well with values observed in other systems featuring vastly different interparticle interactions and particle sizes, suggesting a possible underlying universality, as originally proposed in [15].

In summary, we have presented experimental evidence for the emergence of a wave-vector gap in the dispersion relation of collective shear modes across the glass transition in a two-dimensional mesoscopic colloidal system. In the liquid state, the form of the dispersion is consistent with predictions from Maxwell-Frenkel theory, and the onset of the k -gap coincides with the glass transition, as independently determined from the vanishing of the static shear modulus.

Our findings not only provide direct experimental confirmation of existing theoretical predictions, but also suggest that this framework remains valid beyond conventional first-order solid-liquid melting, extending into continuous glass transition and 2D melting scenarios.

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