Model of defect diffusion and development of the boson peak in an amorphous solid

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A schematic model is proposed for understanding the boson peak as a consequence of the coupling between slowly decaying density fluctuations and the transverse sound modes that develop in the amorphous solid. The present analysis demonstrates that the nature of the dynamics of defect densities in the disordered system plays a crucial role for the appearance of the peak. We compare the results for the dynamic structure factor with the scattering data of Sokolov *et al.*. [J. Non-Cryst. Solids **172-174**, 138 (1994)]. With the relaxation time for the defects becoming longer which is the case more appropriate for the strong glasses as compared to the fragile glasses where structural degradation occurs more easily, the boson peak become more pronounced. [S1063-651X(99)13503-7]

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I. INTRODUCTION

In recent years statistical mechanical models of fluids have been used for understanding the dynamics of supercooled liquids over a wide range of time scales. The self consistent mode-coupling [1,2] approximation for the memory function has been particularly useful in understanding the cooperative effects in a dense liquid. A characteristic feature of supercooled or glassy systems is observation of extra intensity in the neutron scattering [3,4] as well as in Raman scattering [5,6] at low frequencies, distinct from the quasielastic peak. This is usually referred to as the boson peak in the literature, and has been ascribed [7] to the coupling between the relaxational and vibrational motions in the supercooled liquid. The appearance of the broad peak in the low frequency spectrum of the amorphous solid takes place over a frequency range larger than that probed with simple mode-coupling models. The usual mode-coupling approximation for the memory function predicts a two step relaxation process involving the power law decay of correlation over intermediate time scales crossing over to the stretched exponential behavior in the long time, termed the α relaxation. In an extended version of the mode-coupling theory [8,9], there is a final exponential relaxation mode of the density correlation function restoring the ergodicity over the longest time scales. These final decay mode can be identified to be that of the defects or free volumes [10] in the amorphous system, resulting in a ergodic behavior in the supercooled system. In the present paper we describe an extension of the simple mode-coupling formalism to include the distinct vibrational modes that develop at low temperatures in the amorphous state for understanding the extra intensity appearing for the structure factor.

Following the formalism developed in Ref. [8], we use a Martin-Siggia-Rose-type field theory for computing the corrections due to the nonlinearities in the dynamical set of equations for the slow variables. In the first part of the analysis we keep the treatment general, using a rather standard form of the effective Hamiltonian that determines the equilibrium behavior in the system. We take into account the renormalization to the longitudinal viscosity due to the coupling of the density fluctuation with the transverse sound modes in the amorphous solid. We then analyze the model for a simple case where the wave vector dependence is ignored. We focus on the role of the slowly decaying defect densities which are inherent in the model, and effects of its coupling to the transverse sound modes arising from the solid like nature of the supercooled liquid. The paper is organized as follows: In Sec. II we give a brief description of the model studied in terms of the equations of hydrodynamics and consider the effect of the nonlinearities on the density correlation functions. In Sec. III we consider a simplified version of the model by reducing it to a schematic form, suppressing the detailed wave vector dependence. We demonstrate how the model can be used for an understanding of the boson peak phenomena as a consequence of the coupling of the vibrational motion with slowly decaying density fluctuations. We end the paper with a discussion of the results.

II. DESCRIPTION OF THE MODEL

The equations of fluctuating nonlinear hydrodynamics for the conserved variables density ρ and the momentum density $\vec{g}$ are standard in literature [11,8]. We note here that [12–14] the effective free energy functional used for constructing the dynamical equations has two parts: $F=F_K+F_U$. The kinetic part [15] $F_K=\int d\vec{x}\, g^2(\vec{x})/\rho(\vec{x})$, and the potential part F_U is assumed to have the simple form

$$F_U=\frac{1}{2}\int d\vec{x}\left[\frac{g^2}{\rho}+A\left(\frac{\delta\rho}{\rho_o}\right)^2+2B\frac{\delta\rho}{\rho_o}\vec{\nabla}\cdot\vec{u}+\lambda S^2+2\mu\left(s_{ij}-\frac{\delta_{ij}}{3}S\right)^2\right]. \tag{2.1}$$

The bulk and shear elastic moduli of the amorphous solid, respectively, are given by λ and μ, while A and B are the Landau parameters in terms of which the static structure factor for the liquid can be expressed. S is the trace of the strain tensor field s_{ij} in the isotropic solid, and is defined in terms of the gradient of the field $\vec{u}(\vec{x})$ as $s_{ij}=\frac{1}{2}(\vec{\nabla}_i u_j+\vec{\nabla}_j u_i)$.

$\vec{u}(x)$, which represents the local displacement field about the amorphous structure is being introduced as an extra slow mode [16,17] in the dynamical description for the amorphous solid, and is similar to a position variable. It is defined in terms of the nonvanishing Poisson bracket [14] with the momentum density, similar to the position variable, $\vec{g}$, i.e.,

$$\{u_i(\vec{x}),g_j(\vec{x}')\}=\delta(\vec{x}-\vec{x}')[\,\delta_{ij}-\vec{\nabla}^j_x u_i(\vec{x})]. \tag{2.2}$$

Using result (2.2) and following the same procedures as followed in obtaining the equations for ρ and $\vec{g}$, we obtain the equation of motion for $\vec{u}$:

$$\frac{\partial u_i}{\partial t}-\frac{g_i}{\rho}+\frac{\vec{g}}{\rho}\cdot\vec{\nabla}u_i+\Lambda_{ij}\frac{\delta F}{\delta u_j}=f_i\,. \tag{2.3}$$

The dissipation coefficient Λ_{ij} is approximated by the diagonal form $\delta_{ij}\Gamma_u$. T is the temperature. In the hydrodynamic approach the new bare transport coefficients referring to the dissipative parts in the dynamics of the new slow mode enter the theory as parameters. To keep the analysis simple, here we have ignored the energy fluctuations. The set of fluctuating equations thus obtained give the dynamics of the slow modes for the isotropic solid with elastic properties. In order to investigate the effects of nonlinearities in the hydrodynamics equations on the transport coefficients, a standard Martin-Siggia-Rose-type field theory is used for the study of statistical properties of a classical system. Here we will list a few results relevant for the present calculation, and for details we refer the reader to Ref. [8].

The amorphous solid is assumed to be isotropic over hydrodynamic length scales, and hence the correlations of the fields $\vec{u}$ or $\vec{g}$ are expressed in terms of longitudinal and transverse components, and denoted, respectively, with subscripts L and T in the following. It is straightforward to compute the correlation functions in the Gaussian theory by considering the linearized equations of the fluctuating hydrodynamics. Below we list the autocorrelation functions for the density field ρ and the displacement field $\vec{u}(\vec{x},t)$ obtained using the field theoretic analysis. The density autocorrelation function normalized with respect to its equal time value is denoted by $\psi(q,t)$, while that of the $\vec{u}$ field is denoted by $\phi_L(q,t)$ or $\phi_T(q,t)$. The Laplace transforms of these time correlation functions are

$$\psi^o(q,z)=\frac{z+iq^2\Gamma^o(q)}{z^2-\Omega_q^2+iq^2z\Gamma^o(q)}, \tag{2.4}$$

$$\phi^o{}_T(q,z)=\frac{z+iq^2\eta(q)}{[z+iD_Tq^2][z+i\,\eta_o(q)q^2]-c_T^2q^2}, \tag{2.5}$$

where the superscript o indicates the Gaussian expressions, and $c_T^2=\mu/\rho$ is the speed of transverse sound waves and $D_T=\mu\Gamma_u$. The bare transport coefficient $\Gamma^o=\frac{4}{3}\eta_o+\zeta_o$, where η_o and ζ_o are the shear and bulk viscosities respectively, Ω_q^2 refers to the microscopic frequency of the liquid state. For long times the longitudinal part of the $\vec{u}$ correlation function is given by the dominant pole,

$$\phi^o{}_L(q,z)=\frac{1}{z+iq^2\Gamma_u\alpha}, \tag{2.6}$$

where $\alpha=\lambda+\frac{4}{3}\mu$. The hydrodynamic modes are expressed in the pole structures of the correlation functions. The two poles [18] in expression (2.4) for the transverse component $\phi_T(q,z)$ are given by $z=\pm qc_T-iq^2(\Gamma_u\mu+\eta_o/\rho)$. These represent the two propagating transverse sound modes. The correlation function $\phi_L^o(q,\omega)$ has a diffusive pole given by $z=-iq^2\Gamma_u\alpha$, and represents the slowest mode. In addition to this there are two propagating longitudinal sound modes which are ignored compared to the long lived diffusive mode. In an ideal crystal with long range order, this diffusive mode is interpreted as the vacancy diffusion. In the present model of the amorphous solids this is interpreted as relaxation of the defects or free volumes in the supercooled liquids. This interpretation is given from the fact that the longitudinal part of the correlation function is related to $(\vec{\nabla}\cdot\vec{u})$, and in an ideal crystal without any vacancy would be equal to the negative of density fluctuation [19]. The difference between the two, i.e., $(\nabla\cdot\vec{u}+\delta\rho/\rho_o)$, is taken as a definition for the density of defects [20,13] in an amorphous solid, and this follows a linearized dynamics signifying simple diffusion [17] given by the above diffusive mode. The very long time scales of relaxation of the defects is related [12,14] to this diffusive pole, especially for systems having solid like behavior in the deep supercooled state.

The key quantity of interest for comparing results with the experiments are the scattering functions, the dynamic structure factor, or the density autocorrelation function. In Ref. [8] the following form for the Fourier-Laplace transform of $G_{\rho\rho}(\vec{x},t)$ normalized with respect to its equal time value is obtained in the small q and z limit:

$$\psi(q,z)=\frac{z+iq^2\Gamma^R(q,z)}{z^2-\Omega_q^2+iq^2z\Gamma^R(q,z)}. \tag{2.7}$$

Here $\Gamma^R(q,z)$ is the generalized longitudinal viscosity is equivalent to a memory function, and takes into account the cooperative effects in a dense fluid. The correction to the transport coefficient $\Gamma(q,z)$ in Eq. (2.7) due to the coupling of hydrodynamic fluctuations in a supercooled liquid [21] provides a feedback mechanism, resulting in the development of long relaxation times in the glassy dynamics. Using standard techniques [8,22], the renormalized transport coefficients or the memory functions are obtained as a functional of the hydrodynamic correlation functions in a self-consistent form. These involve the density correlation $\psi(q,t)$ and the correlation $\phi_L(t)$ and $\phi_L(t)$ of the $\vec{u}$ fields introduced for the amorphous solids as

$$\begin{aligned}\Gamma(q,t)=\Gamma_o+2\beta\int\frac{d\vec{k}}{(2\pi)^3}[B^2\{&u^4\phi_L(\vec{k},t)\\&+u^2(1-u^2)\phi_T(\vec{k},t)\}\psi(\vec{q}-\vec{k},t)\\&+A^2G_{\rho\rho}(\vec{k},t)G_{\rho\rho}(\vec{q}-\vec{k},t)].\end{aligned} \tag{2.8}$$

The model where only the second term with the density correlation functions is kept has been widely studied [1,23] with respect to idealized glass transition models. The coupling to the correlation of $\vec{u}$ fields refers to the distinct nature of the supercooled liquid developing solidlike properties near the glass transition. Here we have considered the simplest, or one loop, correction to the renormalized transport coefficients. Contributions from the coupling of many other correlation functions are present in the renormalization of the transport coefficients. However, the coupling of the density correlation functions are the dominant ones, and in the self-consistent mode coupling approximation we use the above expression for the renormalized density correlation function.

III. SIMPLE MODEL FOR THE DYNAMIC CORRELATION

In studying feedback effects on dynamics due to slowly decaying density fluctuations at supercooled states, [24,23], it has been generally assumed that the theory is not sensitive to the wave vector dependence. Ignoring the wave vector dependence in the integrals appearing in mode-coupling contributions to the renormalized transport coefficients, we obtain the following simple form for the equation of motion for the density correlation function $\psi(t)$ from Eqs. (2.7) and (2.8):

$$\ddot{\psi}(t)+\dot{\psi}(t)+\psi(t)+\int_0^t ds H[\psi(t-s)]\dot{\psi}(s)=0 \quad (3.1)$$

where the unit of time is chosen in terms of the inverse of the microscopic frequency Ω_0, and the bare transport coefficient Γ_o is chosen to be unity. The memory function $H[\phi(t)]$ is obtained as a functional of the hydrodynamic correlation functions in the form

$$H(t)=c_1 F[\phi_L(t),\phi_T(t)]\psi(t)+c_2\psi^2(t), \quad (3.2)$$

where c_1 and c_2 are dimensionless constants determined in terms of the wave vector integrals due to the mode-coupling vertex functions. If one makes a simple choice that the function $F(t)$ is frozen in time [say $F(t)=1$], a simple model of the form

$$H(t)=c_1\psi(t)+c_2\psi^2(t) \quad (3.3)$$

is obtained [12]. This has been widely studied in the literature as a *schematic* model for studying glass transition, especially the stretched exponential relaxation [23,25]. In the glassy phase $\psi(t)$ freezes into a nonzero value over the long time limit corresponding to the critical values

$$c_1^*=\frac{2\lambda-1}{\lambda^2}, \quad c_2^*=\frac{1}{\lambda^2}, \quad (3.4)$$

where $\frac{1}{2}\leqslant\lambda\leqslant 1$. The critical line is given by, $c_1^*=2\sqrt{c_2^*}-c_2^*$. In Fig. 1 we show the critical line separating the liquid (lower) from the glass (upper) phase in this simple model, and this is an indicator of the couplings c_1 and c_2 that give rise to the glassy phase where the appearance of the boson peak is more prominent.

Considering the model that follows from the equations of nonlinear fluctuating hydrodynamics and a proper time dependence for the functional $F[\phi]$, we are able to analyze the effect of the coupling of the sound modes as well as the slowly decaying defect density mode with the density fluctuations. Thus the function $F(t)$ is expressed as

$$F[\phi]=\phi_L(t)+f(\sigma)\phi_T(t). \quad (3.5)$$

Simple calculation from Eq. (2.8) yields $f(\sigma)=(12-14\sigma)/9(1-2\sigma)$, where $\sigma=(3\lambda-2\mu)/[2(3\lambda+\mu)]$ is Poisson's ratio. In general, formulation of the mode-coupling equations [14] for $\phi_T(t)$ and $\psi_L(t)$ are required for obtaining a fully self-consistent solution of the problem. As a first step, we approximate for the $\vec{u}$ correlation functions with the solutions obtained from the linearized dynamics, and analyze the coupling of the density fluctuations with $\phi(t)$'s driving the dynamics over time scales in which the solidlike behavior in the supercooled liquid persists. Apart from the propagating longitudinal and transverse sound modes, we have a decaying mode of defect density, and this is linked with the longitudinal correlation function ϕ_L. With this, the function $F(t)$ takes the form

$$F[\phi]=e^{-\delta t}+f(\sigma)\phi_T(t), \quad (3.6)$$

where δ now corresponds to the time scale of very slowly decaying defect density and $\phi_T(t)$ is the solution of Eq. (2.5) for the transverse sound modes. We solve for the density correlation function $\psi(t)$ by numerically integrating the integrodifferential equation (3.1) for suitable choices of the parameters involved.

We have used this model to fit the data of Sokolov *et al.* [6] for OTP. The value for the constants c_1 and c_2 are chosen so as to be close to the glassy phase. The long time limit of the density correlation function is subtracted out, and the resulting data are Fourier transformed to obtain the fit shown in the figure. In the simple model the density correlation function freezes to a nonzero value, and we replace this nondecaying part by the exponentially relaxing mode signifying the final decay process restoring the ergodicity. We approximate the time scale of relaxation of this mode to be same

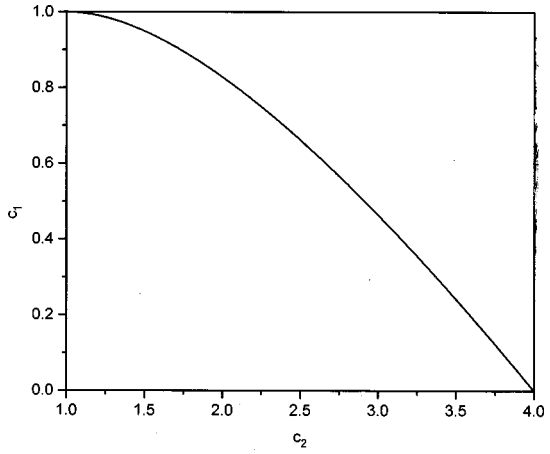

FIG. 1. The phase diagram showing the dimensionless parameters c_1 and c_2 for the ideal glass (upper) and the liquid phase (lower) in the simple model with $\delta=0$ (see text).

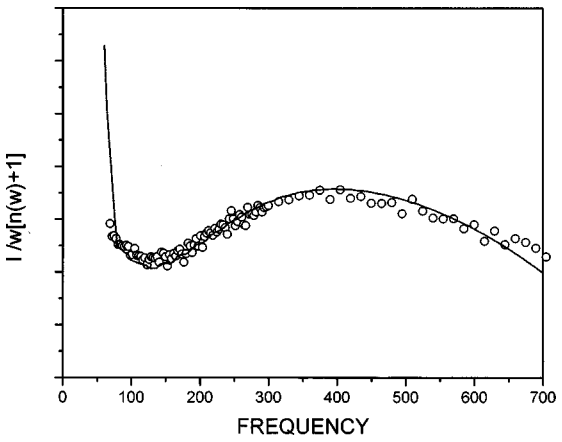

FIG. 2. The Raman spectra data (in arbitrary units) of Ref. [6] normalized with respect to the Bose factor $\omega[n(\omega)+1]$ (open circles) at $T=245$ K, vs the frequency in GHz. The solid line presents the result obtained from the present model for the normalized correlation function ψ.

[8,13] as that of the time scale of relaxation of the slowly decaying defect density given by δ. Thus the central quasielastic peak is fitted a Lorentian of width δ. Since the scattering results are expressed in the frequency space, in Fig. 2 we show the dynamic structure factor as a function of frequency.

The value of Poisson's ratio σ is taken to be 0.36. Figures 2–4 show different values of the quantity δ, which denotes the time scale for the decay of the defects density and plays a central role in the appearance of the peak on the shoulder of the quasielastic peak. As δ become very small, a secondary peak in the structure factor appear at the intermediate frequency determined by ω_o. However, this intermediate peak disappears in the shoulder of the quasielastic peak as δ become large, and the solidlike property of the supercooled liquid no longer persists. This is shown by the fit of the data of Sokolov *et al.* for higher temperatures in Figs. 3 and 4. The equations are solved for $c_1=0.56$ and $c_2=2.78$, corresponding to the value of $\lambda_o=0.6$ on the transition line for

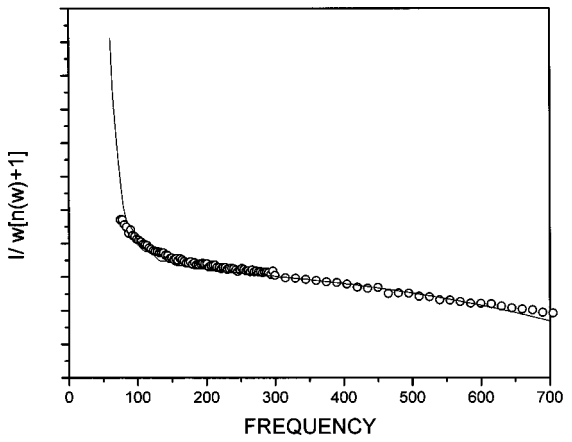

FIG. 3. The Raman spectra data (in arbitrary units) of Ref. [6], normalized with respect to the Bose factor $\omega[n(\omega)+1]$ (open circles) at $T=270$ K, vs the frequency in GHz. The solid line presents the result obtained from the present model for the normalized correlation function ψ.

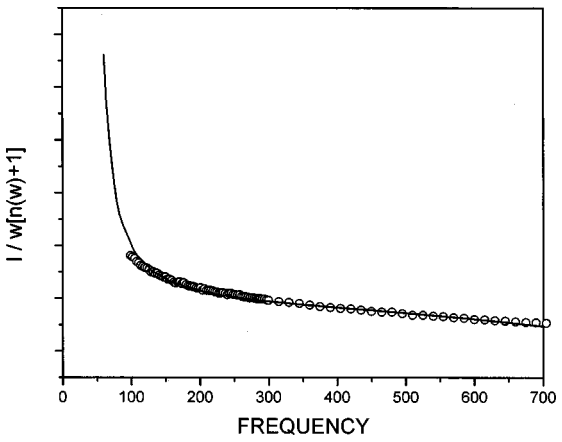

FIG. 4. The Raman spectra data (in arbitrary units) of Ref. [6] normalized with respect to the Bose factor $\omega[n(\omega)+1]$ (open circles) at $T=297$ K, vs the frequency in GHz. The solid line presents the result obtained from the present model for the normalized correlation function ψ.

Fig. 2, at $T=245°$. For the higher temperatures, i.e., $T=270°$ and 297°, the fit is obtained with lower values of $\lambda=0.57$ and 0.53, respectively. The values of the bare transport coefficients are kept same in all the three fits, and the only adjustable parameter used is δ. In Fig 5, we plot the quantity δ, giving the time scale of relaxation of defects vs the temperature, indicating that for lower temperatures the defect densities are long lived and the boson peak is more pronounced. Since here we have considered a simple model, dropping all wave vector dependences, this amounts to approximating the relaxation modes by single exponential relaxation. A fully wave vector dependent model should have a range of time scales for the relaxation coupling to produce the peak. These will be determined by the structural properties representing the short range order in the supercooled liquid, and the position of the peak signifies the dominant frequency coming from the coupling of density fluctuations with the sound modes in the amorphous solid.

IV. DISCUSSION

The coupling of the slowly decaying density fluctuations with the sound modes in an amorphous solid in the presence

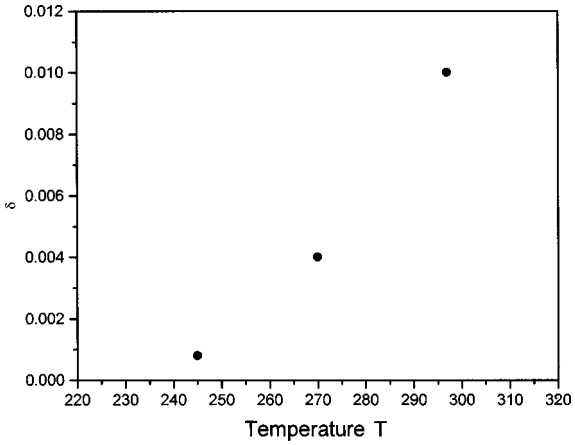

FIG. 5. δ values for the three different temperatures shown in Figs. 2–4.

of long lived defect densities gives rise to an extra intensity at an intermediate frequency following the quasielastic peak. The presence of the very slowly decaying defect density mode is crucial in the appearance of a local peak on the shoulder of the quasielastic peak, as shown in the present model. The increase of the quantity δ can be related to an increase of temperature. This can also be interpreted to represent systems which are more fragile. It is an experimental fact that in a strong glass [26], which has a tendency to form network type structures where structural degradation occur less easily, the defects or voids created are long lived; in the present analysis it is demonstrated that in those system the boson peak is clearly seen. In fragile glasses, where the traditional mode-coupling approach has been more successful, the defects relax more easily, and there the boson peak appears to be much less prominent. This feature of the present model, where *the qualitative dependence of the appearance on the boson peak with the decreasing fragility of the amorphous solid is being captured*, is a key result of the paper. Here we have used parameter values to demonstrate the appearance of the peak as well as its crucial dependence on the quantity δ related to the slow decay of defects in the amorphous solidlike structure. The solutions of the mode coupling equations are used to fit the data for boson peak in OTP, due to Sokolov *et al.*

The introduction of the displacement field $\vec{u}$ in the case of the amorphous solid requires reference to a rigid lattice, and the ergodicity restoring process in the system invalidates the existence of any such rigid structure. The crossover between these two situations requires a self-consistent treatment with possible dynamic connections between the elastic and viscous behaviors of the system. The longitudinal part of the local displacement field $\vec{u}$ is manifested through $-\vec{\nabla}\cdot\vec{u}$, which in the case of a strictly rigid lattice will be simply related to the density fluctuations, while the transverse part reflects the transverse sound modes present due to the solidlike nature of the system. Here $\vec{u}$ is not an order parameter in the amorphous solid, and translational symmetry is maintained over long length scales. $(-\vec{\nabla}\cdot\vec{u})$, which is well defined on time and length scales that are not too large, is used just to define the vacancy concentration $c(\vec{x},t)$. It is assumed that the equations for ρ, $\vec{g}$, and c constitute a reasonable set of equations incorporating vacancy diffusion at least below the mode-coupling transition temperature T_c.

Schematic models for the mode-coupling kernel have been used in the literature [27,28] to fit scattering data for the boson peak. The present work thus involves a realistic starting point for obtaining the mode-coupling equations used in the subsequent analysis, and does not take an *ad hoc* second correlator to analyze the data on the boson peak. Our model involves a set of equations governing the dynamics of the density fluctuations that are obtained from an extension of fluctuating hydrodynamics to complex systems. It takes into account the coupling to the transverse sound modes, which represent the vibrational modes developing in the amorphous solid, and the density fluctuations in the presence of a very long lived defect correlation. It demonstrates the role of long lived vacancies in an amorphous system in enhancing the boson peak. The present model involves splitting the correlation of the $\vec{u}$ fields into transverse and longitudinal parts, as applies for an isotropic system.

In the form of the mode-coupling theory used in the present work, the explicit temperature dependence of the peak is not captured. In order to obtain this explicit temperature dependence, the static or thermodynamic properties that appear in the mode-coupling integrals have to be used as inputs in the theory. This can actually be achieved by proper modeling of the vibrational properties in terms of temperature dependent parameters, presumably using computer simulation data on simple systems. Here we have described a simple model to demonstrate the crucial mechanism for the coupling of vibrational and translational motion in the supercooled system, giving rise to an extra intensity in the structure factor over the intermediate frequency range.

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