

Universal Sound Absorption in Low-Temperature Amorphous Solids

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A general elastohydrodynamic theory is developed based on the phenomenological assumption of a sharp decrease of shear relaxation time at large wave vectors $k > k_\xi$, where k_ξ is of order of inverse of several interatomic distances a . This theory describes the low-energy excitations of glassy and amorphous solids, which contribute to anomalous linear-in-temperature specific heat and limit phonon thermal conductivity. The ratio of the wavelength of the phonon, λ , to its mean free path, l , which is the universal property of sound absorption in glasses, is derived in this theory to be $\lambda/l = (2/3)(c_t/c_l)^2(k_\xi a)^3$, where c_t and c_l are transverse and longitudinal sound velocities correspondingly.

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Glasses at low temperatures ($T < 1\text{--}5$ K) show a remarkable universality in their sound absorption: “the ratio of the phonon wavelength, λ , to the phonon mean free path, l , has been found to lie between 10^{-3} and 10^{-2} in almost all cases, independent of chemical composition and frequency (wavelength) of the elastic wave which varied by more than nine orders of magnitude in the different experiments” [1]. The so-called standard tunneling model of glasses [2] assumes phenomenologically the existence of *independent local tunneling two-level systems* (tunneling TLS's) with wide distribution of tunneling splittings and relaxation times. The standard model had much success in describing several low-temperature properties of glasses (specific heat, thermal conductivity, etc.) (for a review, see [3]). The *coherent nature* of excitations was revealed by echo and spectral diffusion experiments, while the *tunneling aspect* of TLS's was never clearly demonstrated experimentally [4]. Most importantly, the understanding of the aforementioned universal absorption is beyond the scope of the standard model, because the density of tunneling TLS's has to be assumed *arbitrarily* for this model [5]. Moreover, the *independence of tunneling TLS's* implies large variation and nonuniqueness of density of TLS's in different materials, in contradiction to experimental observations [1].

In this Letter I construct a phenomenological viscoelastic theory, alternative to the phenomenological standard model, based on a single assumption that the relaxation time of structural shear modulus has a sharp threshold, becoming finite for large wave vector elastic perturbations of amorphous structure. This assumption is reminiscent of the Maxwell theory of viscoelastic liquids, with the important difference that here the relaxation time is assumed to be finite only for short-wavelength perturbations. The low-energy states of this theory (in addition to phonons), contributing an almost linear-temperature specific heat, are collective density modes with large wave vectors k and small frequencies. The same assumption leads naturally to the appearance (or

rather redistribution) of additional density of states at high frequencies (so-called boson peak). The universal dimensionless constant λ/l of amorphous condensed matter physics is derived in this phenomenological theory to be due to the weak coupling between phonons (small k) and relaxational collective (large k) modes.

There is more to be said about universality of λ/l ratio in glassy and other types of materials. It appears that at low temperatures only for pure crystalline solids and quantum liquids (He^3 and He^4) this universality is not applicable (for a few other exceptions, see the review [1]), while for a huge class of materials [1] (a large number of disordered crystals and polycrystals, some quasicrystals and metallic glasses, in addition to insulating glasses of various types) the ratio of λ/l falls into the same range 10^{-3} to 10^{-2} . Another insightful observation into the nature of universality of λ/l came from the irradiation experiments on crystalline silicon [6]. In a certain wide range of irradiation doses, the irradiated samples show the sound absorption properties very similar to glasses. It is illogical to believe that *independent tunneling* states are generated by irradiation in such a situation *at a fixed universal density*. It seems that a more general assumption is required to understand this remarkable universality, since neither amorphicity nor glassiness is a necessary condition for universality to appear.

Glassy and amorphous solids are solids in the sense that shear and compressional moduli are finite at low frequencies. The transverse and longitudinal phonons with long wavelengths are well-defined excitations, since the phonon mean free path is much longer than its wavelength, $l \sim 10^2 \lambda$. As the wavelength of phonon becomes comparable to the correlation length of frozen disorder [7], Rayleigh scattering of phonons by disorder as well as anharmonicity become important. Elastic Rayleigh scattering of phonons has strong dependence on the wavelength λ of phonon, $\lambda/l = 4\pi^2(\xi/\lambda)^3$, where ξ^3 is the “correlation volume” of disorder. Anharmonic three-phonon processes, being proportional to the phonon density of states (of high frequency or thermal phonon

involved), also become more relevant at higher frequencies (or shorter wavelengths). The corollary of such an argument is that at short length scales a combined effect of anharmonicity and disordered arrangement of atoms may produce finite shear relaxation. To be precise (rigorous justification of the assumption is the subject of microscopic theory and beyond the scope of this Letter), the main *phenomenological assumption* (PA) is the following statement: *structural shear relaxation time decreases sharply as a function of the wave vector k from infinity at small wave vectors [$k < k_\xi \equiv 1/\xi$, where ξ is the typical correlation length of disorder in glasses of order of several interatomic or intermolecular (or any elementary unit of structure) distances] to a finite relaxational time smaller or of the order of inverse phonon frequency for wave vectors $k > k_\xi$. This PA is firmly based on experiments [8] as well as motivated by various studies [9]. The main idea of this Letter is to construct a minimal hydrodynamic theory based only on macroscopic conservation laws and the PA.*

To derive the spectrum of density modes it is sufficient to use macroscopic conservation laws and constitutive relation for the stress tensor [10,11]. The conservation of the mass density ρ and the conservation of momentum density $g_i = \rho v_i$ are in a linearized approximation

$$\partial_t \rho + \rho_0 \operatorname{div} \vec{v} = 0, \quad (1)$$

$$\rho_0 \partial_t v_i - \nabla_j \sigma_{ij} = -n_0 \nabla_i \delta V_{\text{ext}}, \quad (2)$$

where v_i and σ_{ij} are local values of velocity and stress tensor fields, and $n_0 = \rho_0/m$ is an equilibrium number density. The constitutive relation between the stress tensor σ_{ij} and the strain tensor u_{ij} is assumed to be

$$\sigma_{ij} = \frac{2\mu}{1 + i/[\omega\tau_s(k)]} u_{ij} + \Lambda u_{ii} \delta_{ij}, \quad (3)$$

where Λ and μ are Lamé coefficients [11], and $\tau_s(k)$ is the shear relaxation time, dependent on the wave vector k of the periodic modulation. Notice that, if $\omega\tau_s(k) \gg 1$, the constitutive relation describes a solid body with a finite shear modulus. While for $\omega\tau_s(k) < 1$ the constitutive relation describes a viscoelastic medium with frequency- and wave-vector-dependent viscosity $\eta_k(\omega) = \mu\tau_s(k)/[1 - i\omega\tau_s(k)]$, but yet with a finite shear modulus $\operatorname{Re} \mu(\omega) = \mu(\omega\tau_s)^2/[1 + (\omega\tau_s)^2]$. Notice that for $k > k_\xi$, PA states $c_{l,t}k\tau_s(k) \sim 1$ and never $c_{l,t}k\tau_s(k) \ll 1$. Coupling between thermal variables (e.g., gradients of temperature) and mechanical variables is neglected

here. A particular functional dependence of $\tau_s(k)$ is not considered, since this dependence may vary among various materials. The only requirement is that $\tau_s(k)$ drops sufficiently fast at $k \sim k_\xi$. Substituting Eqs. (1) and (3) into Eq. (2), the longitudinal density correlation function can be calculated:

$$\chi_{\rho,\rho}(k, \omega) = \frac{n_0 k^2 / m}{-\omega^2 + k^2 (c_\infty^2 - \frac{c_\infty^2 - c_0^2}{1 - i\omega\tau_s(k)})}, \quad (4)$$

with $c_\infty^2 \equiv c_l^2 = (\Lambda + 2\mu)/\rho_0$ and $c_0^2 = \Lambda/\rho_0$. The longitudinal density fluctuation spectrum is given by $\operatorname{Im} \chi_{\rho,\rho}(q, \omega)$ and has long low-frequency ($\omega \ll c_l k$) tails, which can be approximately written as

$$\operatorname{Im} \chi_{\rho,\rho}(k, \omega) \cong \frac{n_0}{m} \frac{c_\infty^2 - c_0^2}{c_0^4} \frac{\omega\tau_s(k)}{1 + [\omega\tau_s(k)]^2 (c_\infty/c_0)^4}. \quad (5)$$

Therefore, due to shear stress relaxation, collective density modes at large $k > k_\xi$ have a *linear “bosonic” density of states at low frequencies*. This is a very general observation if there is any kind of relaxation for high- k modes. These modes of low frequency and short wavelength describe local anharmonic rearrangements of atoms, TLS’s of the standard model being a special instance, which are spatially correlated due to finite elastic bulk and shear moduli. Such atomic rearrangements are strongly inhibited in a crystalline solid due to the presence of short and long range order.

In a similar manner, the correlation function for the transverse part of momentum density $g_i^T = \rho v_i^T$ can be calculated [12]: $\chi_{g^T, g^T}(k, \omega) = (c_t^2 k^2 / \rho_0) / [\omega^2 - c_t^2 k^2 + i(\omega/\tau_s(k))]$, where the transverse sound velocity is $c_t = \sqrt{\mu/\rho_0}$. The correlation functions of density $\operatorname{Im} \chi_{\rho,\rho}(k, \omega)$ and momentum density $\operatorname{Im} \chi_{g^T, g^T}(k, \omega)$ can be used to calculate the interaction and kinetic energies correspondingly. For instance, the interaction energy is given by $\langle V_{\text{int}} \rangle = \int d\omega \int d^D k V(k) \operatorname{Im} \chi_{\rho,\rho}(k, \omega) \coth[\hbar\omega/(2k_B T)]$, where $V(k)$ is the interparticle interaction. The total energy, adding contributions from longitudinal and transverse degrees of freedom, can then be calculated in the harmonic approximation. The essential result for the linear temperature dependence of the specific heat, due to the interaction part of the energy, can be noticed already from the linear density of states [Eq. (5)]. Another technical way to calculate the specific heat is to use the expression for the entropy of interacting boson modes [13] derived using a diagrammatic approach:

$$S(T) \cong \frac{1}{\pi} \int_0^\infty d\omega \frac{e^{\beta\omega}}{(e^{\beta\omega} - 1)^2} \frac{\omega}{T^2} \sum_k \left[\operatorname{Im} \ln \left(\frac{n_0 k^2 / m}{\chi_{\rho,\rho}(k, \omega)} \right) + \frac{(c_\infty^2 - c_0^2) k^2 \omega \tau_s(k)}{1 + [\omega\tau_s(k)]^2} \operatorname{Re} \left(\frac{\chi_{\rho,\rho}(k, \omega)}{n_0 k^2 / m} \right) \right]. \quad (6)$$

Notice that the sum over k at low frequencies for the second term in Eq. (6) is approximately equal to $\sum_k \operatorname{Im} \chi_{\rho,\rho}(k, \omega)$ from Eq. (5). Thus, as inferred before from the interaction energy, the entropy and the specific heat have a linear temperature dependence. For the purpose of approximate estimates, the specific heat $C_V(T)$ after integrating over the range of wave vectors

$k_\xi < k < k_u$ (k_u is upper cutoff) with some typical relaxation time $\tau_{s,\text{typ}}$ is

$$C_V(T) \sim \frac{k_u^3}{3\pi^2} (k_B T \tau_{s,\text{typ}}) \left(\frac{A(c_\infty^2 - c_0^2)}{c_0^2} + \frac{6B}{c_l^2 k_u^2 \tau_{s,\text{typ}}^2} \right), \quad (7)$$

where A and B are numerical factors of order one. A crude estimate [$\hbar/\tau_{s,\text{typ}} \sim \hbar\Omega_{\text{BP}} \equiv k_B T_{\text{BP}}$, $k_B T_{\text{BP}} = 50$ K, $k_u \sim 10^7\text{--}10^8$ cm $^{-1}$, and a factor $(c_\infty^2 - c_0^2)/c_0^2$ as 0.3] gives the density of states from $\sim 10^{33}$ to $\sim 10^{36}$ (ergs cm 3) $^{-1}$ for $k_u \sim 10^8$ cm $^{-1}$, which is roughly of order of experimental values. In general, the two terms in brackets, coming from the longitudinal and transverse contributions, are comparable, but depend in different ways on parameters.

The PA straightforwardly implies an accumulation of additional density of states around frequency $\Omega_{\text{BP}} \approx c_0 k_\xi$, since $\tau_s(k)$ drops *sharply* to a finite value around k_ξ . This is the so-called boson peak observed in neutron and Raman scattering [14]. First, notice that the maximum of the spectrum $\text{Im} \chi_{\rho,\rho}(k, \omega)$ as a function of frequency ω is shifted downward from $c_\infty k$ if $\omega\tau_s \gg 1$ toward $c_0 k$ if $\omega\tau_s < 1$. Since it happens for the range of wave vectors k , large additional weight from the range of k wave vectors is redistributed from high frequencies to the frequency around the boson peak. Second, the shape of the intensity $\text{Im} \chi_{\rho,\rho}(k, \omega)$ is asymmetric with larger weight above the maximum. All these factors contribute to the additional density of states around Ω_{BP} : $\nu(\omega) \sim \sum_k \text{Im} \chi_{\rho,\rho}(k, \omega \sim c_0 k_\xi)$. The intensity of the boson peak varies depending on a detailed dependence of $\tau_s(k)$, which is beyond the framework of the phenomenological theory (but see Ref. [9]), and, indeed experimentally, intensity of the boson peak varies significantly among various glasses.

The calculation of sound attenuation [15] needs to take into account the wide distribution of relaxation times postulated in the PA. We are interested in the attenuation of a low-frequency phonon with frequency ω (to be distinguished from ω of the density spectrum) by slow relaxational density modes such that $\omega\tau_s(k) \ll 1$, and therefore a hydrodynamic description of attenuation is applicable. The simplest way to calculate the sound absorption is to use a classical expression for absorption due to finite viscosity [11]: $\gamma_l = \frac{\omega^2}{2\rho c_l^3} (4/3) \text{Re} \eta_k(\omega)$. The low-frequency phonon is absorbed by all collective modes with various large wave vectors k and corresponding relaxation times $\tau_s(k)$:

$$\gamma_l = \frac{2\omega\mu}{3\rho c_l^3} \sum_k \frac{\omega\tau_s(k)}{1 + [\omega\tau_s(k)]^2}. \quad (8)$$

Finally, the ratio of the wavelength λ_ω to the mean free path l_ω is remarkably simple:

$$\alpha \equiv \frac{\lambda_\omega}{l_\omega} = \frac{2}{3} F \frac{c_l^2}{c_t^2}, \quad (9)$$

$$F \equiv \frac{2\pi}{n_0} \int \frac{d^3k}{(2\pi)^3} \frac{\omega\tau_s(k)}{1 + [\omega\tau_s(k)]^2} \approx (k_\xi a)^3, \quad (10)$$

where $n_0 = 1/a^3$. The *small* numerical factor F is a constant, independent of frequency, since the integrand in the integral is essentially a δ function because of the sharp dependence of $\tau_s(k)$ on k . The constant F is very weakly dependent on the detailed functional form of $\tau_s(k)$ and other parameters. The simplicity of the main result—Eq. (9)—for the universal ratio gives insight into the origin of universality. This ratio depends only on the squared ratio of transverse and longitudinal sound velocities and the third power of the ratio of typical interparticle distance a to the scale of correlated disorder ξ . This result is equally correct for longitudinal and transverse phonons at low frequencies, because the damping is due to the coupling to short-wavelength density modes in inhomogeneous media. A more systematic calculation, using a kinetic equation formalism [11], gives the same result. The starting point for such a calculation is that the low-frequency phonon of frequency ω causes the variation of the density $\delta\rho_k(\omega)$ at this frequency, for all modes with various k 's and corresponding $\tau(k)$, and this, in turn, changes the phonon frequency $\delta\omega = \omega(\gamma_k \delta\rho_k)/\rho_0$, where γ_k is a Gruneisen parameter.

Straightforwardly, the thermal conductivity is $\kappa(T) \approx C_{v,\text{ph}}(T)c_0 l(T) \sim T^2$, where $C_{v,\text{ph}}(T) \sim T^3$ is the phonon-only specific heat, and for thermal phonons ($\hbar\omega \sim kT$ and also $kT \ll \hbar\Omega_{\text{BP}}$) the mean free path is $l(T) = \lambda_\omega/\alpha \sim 1/T$.

The density spectrum of Eq. (4) is essentially given by a *linearized* spectrum of the Navier-Stokes equation with viscoelastic moduli. Various nonlinear terms (e.g., of the type $v_j \nabla_j v_i$, due to the $\rho v_i v_j$ part of momentum density tensor) should be added to Eq. (2) [11] to treat nonlinear effects. The anomalous extended states, associated with low-frequency tails of high- k density modes [Eq. (5)], interact weakly with the small- k sound modes (consistently with Ref. [16]), since this interaction is described by the small coupling constant α of the sound absorption. Note also that echoes can arise from extended collective modes in inhomogeneous media, and examples are plasma and ferrites [17]. Therefore, various nonlinear effects observed in glasses (saturation, echoes, and spectral diffusion) are not *a priori* inconsistent with the picture of extended anomalous states. Nonlinear effects as well as long-time logarithmic specific heat (due to the bulk viscosity) will be addressed in the future.

In this paragraph, I compare briefly the elasto-hydrodynamic theory (EHT) proposed here with theories suggested in the literature. The main difference between the standard model [2] and the EHT is that for the former low-energy excitations are independent tunneling systems, while for the latter these excitations are extended modes of short-wavelength modulation of the density. The shortcomings of the standard model [18] were discussed convincingly, and the long-range dipolar interaction between local defects (e.g., TLS's) was suggested to lead to universality [5]. The EHT encapsulates the collective nature of anomalous states arisen due to the

interaction among the local defects (not necessarily two-level systems of any sort) by the elasticity fields, and therefore EHT is indeed somewhat similar in spirit to the earlier proposal [5]. A density spectrum, Eq. (4), was considered before [13] but with a very different assumption. The relaxation time τ_s was assumed to be *independent of wave vector k* and dependent on temperature. Such an assumption does not lead to linear frequency damping of low-frequency phonons as in Eq. (9) and does not explain the existence of the boson peak.

There are several directions to test experimentally the proposed theory. Direct observation of collective modes by inelastic neutron and x-ray scattering at large momentum k and small energies $\omega \ll \Omega_{BP}$ would be a most direct test. Another consequence, which follows from Eq. (4), is the broadening of sound waves close to the boson peak, $\Gamma_k \sim (c_\infty^2 - c_0^2)k^2\tau_s(k)$, proportional to k^2 if $\tau_s(k)$ is weakly dependent on k above the boson peak crossover ($ck_\xi\tau_s \sim 1$), which, it seems, was already observed experimentally [8,14]. Inelastic x-ray scattering [14] provides a direct support for the PA in vitreous silica, where strong phonon scattering occurs from the length scale $2\pi/q_{co} \sim 30 \text{ \AA}$ (to be associated with ξ), which is much longer than the size of elementary structural unit, SiO_4 tetrahedra. Since the anomalous low-frequency modes states appear as the low-frequency tails of high- k density modes around Ω_{BP} , it would be interesting to test experimentally if high- k modes are coupled relaxationally to the anomalous states. Experiments with nonlinear coupling of spectral diffusion type between the high- k modes and anomalous states would be desirable. Finally, echo experiments in their functional relationship to pulse amplitudes and time intervals should be able to differentiate between local and extended modes [17].

It is interesting to inquire further into the origin of universality. Equation (9) shows that the small universal number α is given essentially by the cube of the ratio a/ξ . Therefore, the universality of the a/ξ ratio and the sharp onset of shear relaxation are further important questions for the investigation. The smallness of the ratio a/ξ arises due to the presence of short-range correlation among atoms, which can be called medium-range structural order [19]. A close analogy of this dimensionless number to small numbers associated with various melting transitions can be noted.

In conclusion, the collective low-frequency density excitations, which are due to a finite shear relaxation time at short length scales, are shown to have the linear-in-frequency density of states, $\text{Im} \chi_{\rho,\rho}(k, \omega)$. These anomalous excitations contribute a linear-in-temperature specific heat and determine the inelastic mean-free path of phonons, $l_\omega = \lambda_\omega/\alpha$. The universality of the sound attenuation ratio $\alpha = \lambda_\omega/l_\omega$ is related to the cube of small ratio a/ξ of average interatomic distance a to the medium-range order length ξ .

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