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Universal sound absorption in amorphous solids: A theory of elastically coupled generic blocks

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ABSTRACT

Glasses are known to exhibit quantitative universalities at low temperatures, the most striking of which is the ultrasonic attenuation coefficient Q^{-1}. In this work we develop a theory of coupled generic blocks with a certain randomness property to show that universality emerges essentially due to the interactions between elastic blocks, regardless of their microscopic nature.

1. Introduction

Starting with the pioneering study of Zeller and Pohl [1], experimental work over the last 40 years has shown conclusively that the thermal, acoustic, and dielectric properties of virtually all amorphous materials are not only qualitatively different than those of crystals, but also show a truly amazing degree of quantitative universality [2–4]. The theoretical interpretation of the low temperature data on amorphous materials has for the last four decades been dominated by the "tunneling two state system" (TTLS) model. This model gives an attractive qualitative explanation of the characteristic nonlinear effects observed in ultrasonic and dielectric absorption (saturation, echoes, hole burning). In addition it predicts a frequency and temperature dependence of the ultrasonic absorption $Q^{-1}(\omega,T)$ (here defined as $l^{-1}\lambda/2\pi^2$ in terms of the phonon mean free path and wavelength) which appears to be in fairly good agreement with the experimental data; in the present context we note that Q^{-1} is predicted to be independent of ω and T, and the *same* within a factor of 2 in two regions of the parameter space, in which, at least in terms of the model, the physics is very different, namely the high frequency "resonance regime" (k_BT, $\hbar/\tau \ll \omega$, where τ is a characteristic relaxation time of the thermally excited TTLSs) and the low frequency "relaxation regime" ($\omega \ll \hbar/\tau \ll k_BT$). This prediction appears to agree reasonably well with the data (see Figures 2 and 3 in ref. [4]). Finally, at the cost of introducing a fairly large number of fitting parameters, the model can reproduce most of the experimental data in the low-temperature regime reasonably quantitatively.

Nevertheless, there are a number of problems with the TTLS model. First, while in a few cases (such as KBr-KCN solutions [5]) it is possible to make a plausible identification of the "two level systems", in most amorphous materials their nature remains a matter of conjecture. Secondly, the model as such says nothing about the behavior at intermediate temperatures (1 K–30 K) which also shows a very strong degree of qualitative (though not quantitative) universality. A third difficulty relates to the striking *quantitative* universality and small numerical value of the quantity $Q^{-1}(\omega)$; whether observed directly or inferred (in the "resonance regime") from the coefficient of the $log(T/T_0)$ term in the ultrasound velocity shift, this quantity has the value $(3\pm2)\times10^{-4}$ for almost all non-metallic glasses measured to date [4]. While the TTLS model contains enough independent fitting parameters to "explain" this numerical result, the explanation requires a degree of statistical coincidence between these parameters which has no obvious basis in the model, and is prima facie nothing short of amazing. Finally, the model in its original form neglects the fact, which is emphasized below, that as a result of interaction with the strain (phonon) field, the TTLSs must acquire a mutual interaction [6]; while there exist theoretical approaches which take this feature into account and even use it [7] to attempt to account for the small universal value of Q^{-1}, it is not obvious that at this end of the process the TTLS structure is preserved, so that a question of self-consistency may arise.

In [8,9] the conjecture was made that if we start from a very generic model in which at short length scales there is a nonzero contribution to the stress tensor from some non-phononic degrees

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of freedom whose only necessary feature is that their spectrum is not harmonic-oscillator-like, and take into account their phonon-mediated mutual interaction, we will recover at long length scales a picture which reproduces most, if not all, features of the experimental data below 1 K. The goal of the present paper is quite modest: To attempt a somewhat more quantitative justification of this conjecture with respect to one specific feature, namely the (near) frequency independence and small universal value of Q^{-1} in the regime k_BT, $\hbar/\tau \ll \omega$ (i.e. what in the TTLS model is known as the high frequency resonance regime). We do not attempt to discuss here the behavior of Q^{-1} in other regimes (except for $\hbar/\tau \ll \omega < k_BT$), non-linear effects or (except briefly at the end of Section 2.2) the intermediate-temperature behavior.

We believe that it is one of the strengths of the present work that our result does not rely on adjustable parameters, or the existence of *other* microscopic (unmeasurable) universal ratios to explain the observable one [10–14] (though cf. [7]). The only two inputs on which our outcome depends sensitively are the ratios c_l/c_t and χ_l/χ_t (cf. below for details of the notation) both of which are observed *experimentally* to vary little between different amorphous systems (also cf. Appendix-A). Our third input r_0, which is the size of a "microscopic amorphous block" (defined below) only enters into our equations logarithmically.

The organization of the paper is as follows: In Section 2 we define our model and introduce the central object of our study, namely the dimensionless stress-stress correlation, whose thermally-averaged imaginary part is the measured ultrasonic absorption Q^{-1}. In Section 3 we carry out a real-space renormalization calculation of the *average* of $Q_m^{-1}(\omega)$ over the frequency ω and the starting state m (for details of the notation see below) and show that it vanishes logarithmically with the volume of the system and, for experimentally realistic volumes, has a surprisingly small value, ~0.015. In Section 4 we argue on the basis of a calculation up to second order in the phonon induced interaction, that the functional form of $Q^{-1}(\omega)$ at $T=0$ should be $(\ln\omega)^{-1}$, and that when we combine this result with that of Section 3, the numerical value of Q^{-1} for experimentally relevant frequencies should be universal up to logarithmic accuracy and numerically close to the observed value 3×10^{-4}. In Section 5 we assess the extent to which our calculations are consistent with experiments in the (linear) resonance regime. In Section 6 we attempt to assess the significance of our results.

Throughout this paper we set $\hbar = k_B = 1$. The notation a denotes a "typical" atomic length scale. The suffix $\alpha = l,t$ denotes the phonon polarization (l = longitudinal, t = transverse).

2. Formulation of the problem

Consider a cube of an arbitrary isotropic amorphous material, with side L which is assumed large compared to "microscopic" lengths a such as the typical interatomic distance, but is otherwise arbitrary. We define for such a block the strain tensor e_{ij} in the standard way: If $\vec{u}(\vec{r})$ denotes the displacement relative to some arbitrary reference frame of the matter at point $\vec{r}$, then

$$e_{ij} = \frac{1}{2}\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right) \tag{1}$$

(Note that the anti-symmetric part of the tensor $\partial u_i/\partial x_j$ corresponds to a local rotation; since a spatially uniform rotation costs no energy, any terms in the Hamiltonian associated with this part will be of order higher than zeroth in the spatial gradients, and hence for the purposes of the ensuing argument irrelevant in the renormalization-group sense; we therefore neglect any such terms in the following).

Let us expand the Hamiltonian of the block in a Taylor series in the strain e_{ij}:

$$\hat{H} = \hat{H}_0 + \sum_{ij} e_{ij}\hat{T}_{ij} + \mathcal{O}\left(e^2\right) \tag{2}$$

where the stress tensor $\hat{T}_{ij}$ is defined by

$$\hat{T}_{ij} = \partial\hat{H}/\partial e_{ij} \tag{3}$$

Note that in general, in a representation in which $\hat{H}_0$ is diagonal, $\hat{T}_{ij}$ will have both diagonal and off-diagonal elements.

As usual, we can define the static elasticity modulus $\chi^{(0)}$, a fourth order tensor, by

$$\begin{aligned}\chi^{(0)}_{ij:kl} &\equiv V^{-1}\left(\partial\langle\hat{T}_{ij}\rangle/\partial e_{ij}\right)_{eq} \\ &\equiv V^{-1}\langle\partial^2\hat{H}/\partial e_{ij}\partial e_{kl}\rangle eq\end{aligned} \tag{4}$$

where $V = L^3$ is the volume of the block and the suffix "eq" denotes that the derivative is taken in the thermal equilibrium state (both sides of Eq. (3) are implicitly functions of temperature T). Since by definition the properties of an isotropic amorphous material must be invariant under overall rotation, symmetry considerations constrain $\chi^{(0)}_{ij:kl}$ to have the generic form

$$\chi^{(0)}_{ij:kl} = (\chi_l - 2\chi_t)\delta_{ij}\delta_{kl} + \chi_t\left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}\right) \tag{5}$$

where χ_l and χ_t are the standard longitudinal and shear elastic constants; in the approximation of an elastic continuum, these are related to the velocities c_l and c_t of the corresponding longitudinal and transverse sound waves (of wavelength λ such that $a \ll \lambda \ll L$) by[1]

$$\chi_{l,t} = \rho c^2_{l,t} \tag{6}$$

where ρ is the mass density of the material. Such an approximation however throws away all the effects of interest to us, as we shall now see.

2.1. The Stress-Stress Correlation Function

Let us separate out from the Hamiltonian, the purely elastic contribution $\hat{H}_{el}$, namely,

$$\begin{aligned}\hat{H}_{el}\left(e_{ij}\right) = \text{const.} &+ \int\frac{1}{2}d^3r\sum_{ijkl}\chi^{(0)}_{ij:kl}e_{ij}\left(\vec{r}\right)e_{kl}\left(\vec{r}\right) \\ &+ \frac{1}{2}\sum_i\rho\dot{\vec{u}}^2_i\left(\vec{r}\right)\end{aligned} \tag{7}$$

(where it is understood that the velocity is slowly varying over distances a, as above). Similarly we define the "elastic" contribution to the stress tensor $\hat{T}_{ij}$ by

$$\hat{T}^{(el)}_{ij} \equiv \sum_{ijkl}\chi^{(0)}_{ij:kl}e_{kl} \tag{8}$$

(In above, e_{ij} (and u_i) should strictly be treated as operators, but we prefer not to complicate the notation unnecessarily). Then quite generally, we have

$$\hat{H}\left(e_{ij}\right) \equiv \hat{H}_{el}\left(e_{ij}\right) + \hat{H}'\left(e_{ij}\right) \tag{9}$$

[1] Note that despite the notation χ has the characteristics of a "stiffness" (~ inverse susceptibility) rather than a "susceptibility".