Quasi-localized vibrations, boson peak and tunneling in glasses

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Abstract. In the analysis of the low frequency vibrations in glasses one regularly observes in computer simulations quasi-localized vibrations (QLV) which can be understood as soft localized vibrations hybridized with the sound waves. We show that such modes explain both the Boson peak (BP) and the two-level-systems (TLS), the two most prominent glassy effects at low energies or temperatures. The weak interaction between the soft local vibrations causes a vibrational instability. Stability is restored by anharmonicity resulting in a universal shape of the BP and TLS's.

1. Introduction

At low temperatures or low energies amorphous materials and glasses exhibit two typical glassy features: the two-level systems (TLS), causing e.g. a linear temperature dependence of the specific heat for \( T \lesssim 1 \text{ K} \), and the Boson peak, seen as a maximum in the inelastic scattering intensity around 1 THz. These two effects are usually described by separate models without any interrelation.

The TLS are an intrinsically quantum mechanical effect, observed when a group of atoms can tunnel between two adjacent minima of the potential energy, double well system. The ground state energy is then split by

\[ E = \sqrt{\Delta_0^2 + \Delta^2} \]  

where \( \Delta_0 \) is the tunnel-splitting in a symmetric double-well system and \( \Delta \) is the asymmetry between the two minima. The tunnel splitting can be written approximately as

\[ \Delta_0 = \hbar \omega e^{-\lambda}. \]  

where \( \hbar \omega \) is the zero-point energy for the motion in either of the two minima and \( \lambda = g \sqrt{mE_b d} / \hbar \) with \( g \) a geometrical factor, \( m \) the mass of the tunneling entity, \( E_b \) the barrier height and \( d \) the distance between the two minima. The condition that the TLS are observable, i.e. that the tunneling time is shorter than the experimental time, \( \lambda \) has to be of order 1. This condition can only be met if tunneling is a local process. The concentration of TLS is low, of order \( 10^{-6} \) per atom. In the standard tunneling model [1] one assumes a constant distribution of \( \Delta \) and \( \lambda \).

It has been shown that the Boson peak (BP) is due to an excess of harmonic vibrations over the Debye density of states of the sound waves. It corresponds to a maximum in \( g(\omega)/\omega^2 \)
but mostly only to a shoulder in the density of vibrational states, \( g(\omega) \), itself. The fraction of vibrational modes comprising the BP is of order \( 10^{-2} \). Since there is a continuum of frequencies the harmonic eigenmodes are extended. The BP-states, being harmonic, can be obtained from a dynamic matrix which will reflect disorder. This ansatz has been used by a number of authors [2]. Its weakness is that one has to use some extra conditions to obtain the needed soft vibrations while avoiding negative eigenvalues.

Our approach is different; we start from the quasi-localized vibrations which have been ubiquitously observed in computer simulations, e.g. [3]. Instead of using the exact harmonic eigenstates we use the sound waves and cores of these vibrations as basis as was implied in our earlier work on the soft potential model [4]. Here we exploit fully the interaction between the soft mode cores which will downshift the low frequency spectrum and even cause a vibrational instability. Invoking anharmonicity to lift the instability we find both a universal (harmonic) BP-excess-spectrum and TLS’s. Full details of the calculations can be found in Refs. [5-7].

2. Interacting Soft Vibrations

Let us assume we have randomly distributed soft oscillators with a broad distribution of frequencies. Including the interaction between these oscillators their total potential energy is

\[
U_{\text{tot}}(x_1, x_2, ..., x_s) = \sum_i k_i x_i^2 - \frac{1}{2} \sum_{i,j \neq i} I_{ij} x_i x_j + \frac{1}{4} \sum_i A_i x_i^4, \quad A_i > 0. \tag{3}
\]

Here \( x_i \) are the generalized coordinates describing the vibrations, \( k_i > 0 \) are the quasielastic constants of noninteracting oscillators and \( I_{ij} \) determines the weak bilinear interaction between the oscillators. To stabilize the system we have added in this equation the anharmonic terms, \( A_i x_i^4 \) (with \( A_i > 0 \)). Taking the elastic dipole interaction between the oscillators, the interaction strength is given by

\[
I_{ij} = g_{ij} J / r_{ij}^3 \tag{4}
\]

where \( g_{ij} \simeq \pm 1 \) accounts for the relative orientation of the HO’s, \( r_{ij} \) is the distance between HO’s and \( J \) the dipole strength which can be determined from the coupling of the oscillators to the sound waves in the glass. We assume that all oscillators are stable without interaction and have a broad distribution of quasielastic constants. To be specific we will use \( \Phi_0(k) = 3/2\sqrt{k} \) for \( 0 \leq k \leq 1 \) corresponding to a density of states of the noninteracting oscillators \( g_0(\omega) = 3\omega^2, \omega \leq 1 \). The exact harmonic vibrations can then be obtained by matrix diagonalization neglecting the anharmonic term. The result is a downshift of the low frequency part of the spectrum as shown in Fig. 1. This result is similar to the one obtained for random matrices without restrictions to ensure stability (\( k \geq 0 \)). Instead of introducing some not well understood

![Figure 1.](image)

Low frequency part of the distribution function of the renormalized quasielastic constants, \( \Phi(k) \), calculated as ensemble average by exact diagonalization of systems of \( N = 2197 \) oscillators for coupling strengths \( J = 0.07, 0.10, 0.15 \) (solid curve) and without interaction (dashed curve). The dotted line is an analytic approximation for \( J = 0.01 \).
restriction on the matrix elements we show that this harmonic vibrational instability creates a universal shape of the BP and additionally a realistic density of TLS’s.

Stability is restored by anharmonicity. The downshift of $\Phi(k)$ for small $k$ is caused by the action of higher frequency oscillators on soft ones. One can show that there is a critical frequency $\omega_c = \sqrt{mk_c} \propto J$ below which the spectrum is strongly affected by the interaction. In a good approximation one can eliminate the high frequency motion from the low frequency one by an adiabatic approximation. Thus one obtains for the low frequency oscillator an effective quartic potential.

$$U_{\text{eff}}(x_1) = \frac{1}{2}(k_1 - \kappa)x_1^2 + \frac{1}{4}A_1x_1^4$$  \hspace{1cm} (5)

where $\kappa$ is the effect of the interaction and $k_1 - \kappa$ is the renormalized quasielastic constant whose spectrum is shown in Fig. 1. For negative values of $k_1 - \kappa$ stability is restored by moving the oscillator to either minimum where one regains a positive constant $2(\kappa - k_1)$. Apart from the factor 2 the negative part of $\Phi(k)$ is thus reflected onto the positive part. It is remarkable that this effect is due to the anharmonicity but the actual value of it does not enter the result.

This harmonic approximation plus stabilization so far gives a finite value of $\Phi(k = 0)$ which corresponds to a vibrational spectrum $g(\omega) \propto \omega$ for $\omega \lesssim \omega_c$. Such a spectrum is highly unstable against perturbations. In our system such a perturbation is inherent. Changing the origin of the oscillators to either minimum in Eq. (5) implies a static displacement which via the bilinear coupling, Eq. (3) translates into a force on the other oscillators. The effective potential energy then reads

$$U_{\text{eff}}(x) = -fx + \frac{1}{2}kx^2 + \frac{1}{4}Ax^4.$$  \hspace{1cm} (6)

Together with the anharmonicity this additional force shifts the lowest part of the density of states upwards, creating the seagull singularity, $g(\omega) \propto \omega^4$ for $\omega << \omega_c$, of the soft potential model [8]. A detailed calculation [5] gives for the excess vibrational spectrum the universal shape function total DOS at $T = 0$

$$g_{\text{tot}}(\omega) = \frac{12}{\pi}n_0M\Phi(0)\frac{\omega^2}{\omega^*} \left( \frac{\omega}{\omega^*} \right)^{\frac{3}{2}} \int_0^\infty \frac{dt}{1 + (\omega/\omega^*)^6} \int_0^\infty (3 - 2t^2)^2 \int_0^\infty \frac{dt}{1 + (\omega/\omega^*)^6}$$  \hspace{1cm} (7)

Where $\omega^*$ is approximately the BP frequency. Fig. 2 shows the excess density of vibrational states in the usual reduced representation. This BP-shape is in good agreement with experiment

![Figure 2.](image)

The Boson peak: the reduced density of states $g_{\text{tot}}(\omega)/\omega^2$ of the excess modes given by Eq. (7).

[6]. The theory can also be used to explain the pressure dependence of the BP [7].

Eq. (5) represents a two well potential which will give rise to TLS’s. Apart from logarithmic corrections, already found in the soft potential model [4] the distribution of the standard tunneling model is recovered. Our theory, however, allows us to estimate the magnitude of
the constants of this model. We can explain for instance the low number of observable TLS’s and the low value of the so called tunneling strength $C$. These values are small because only a small fraction of double-well systems allow tunneling in observable times due to the exponential dependence of the tunneling element, Eq. (2).

3. Discussion
We propose a mechanism of formation of the Boson peak and simultaneously of TLS’s in glasses and other disordered systems from weakly interacting QLV’s stabilized by anharmonicity. Whereas anharmonicity is essential in creating the atomic structures supporting the Boson peak, the vibrations forming the peak in the inelastic scattering intensity or the reduced density of states are essentially harmonic.

In the unified approach developed in the present paper the densities of tunneling states and of excess vibrational states at the Boson peak frequency are interrelated. The form of the Boson peak appears to be universal and independent of the initial assumptions about the interaction strength, or the initial distribution function of oscillator frequencies, or of the anharmonicity strengths. Relating experimental TLS’s and Boson peak parameters we are able to show that our theory is consistent and that the instability crossover frequency $\omega_c$ lies in the Boson peak region. Due to the weakness of the interaction $I$ the universal reconstruction of the spectrum in our theory occurs only in the low frequency range.

The theory presented in this paper deals with the effects of soft modes produced by disorder which can be expected to have a broad frequency distribution. In the literature the term Boson peak is rather loosely defined. Often it is used for any low frequency maximum in the reduced DOS. In particular in plastic crystals soft oscillators are present even before disorder. Consequently disorder only broadens their sharp density of states. Depending on the strength of this broadening our theory will apply more or less to such cases. The same applies to TLS which also can be present before disorder.

4. Conclusion
In summary, we have shown that the same physical mechanism is fundamental for such seemingly different phenomena as the formation of two-level systems in glasses and the Boson peak in the reduced density of low-frequency vibrational states $g(\omega)/\omega^2$. In this way two of the most fundamental properties of glasses are interconnected.

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