High-frequency vibrational density of states of a disordered solid

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Abstract

We investigate the high-frequency behavior of the density of vibrational states in three-dimensional elasticity theory with spatially fluctuating elastic moduli. At frequencies well above the mobility edge, instanton solutions yield an exponentially decaying density of states. The instanton solutions describe excitations, which become localized due to the disorder-induced fluctuations, which lower the sound velocity in a finite region compared to its average value. The exponentially decaying density of states (known in electronic systems as the Lifshitz tail) is governed by the statistics of a fluctuating-elasticity landscape, capable of trapping the vibrational excitations.

(Some figures may appear in colour only in the online journal)

1. Introduction

The density of states (DOS) of a disordered system is a quantity which has been vividly discussed [1–4]. Lifshitz was one of the first authors to calculate the disorder-induced corrections to the DOS, for electron and phonon systems [5]. By a phenomenological argument, he explained the occurrence of an exponential tail of the DOS in the vicinity of the band-edges, which is related to the localization of the one-particle states. As the penetrability of an arbitrarily shaped barrier tends towards zero at large energies [6], one expects the high-energy eigenstates of a system with a random fluctuating potential to be localized. The DOS is then proportional to the probability of the occurrence of wells, capable of trapping the single-particle states, which for weak fluctuations (e.g. low impurity concentrations) can always be approximated by an exponential function. The energy dependence of the exponent depends crucially on the energy of localization, from which some substantial corrections from the set of maximally crossed diagrams in the perturbation expansion arise, similar to the energy–momentum relation of a wave in a potential well, if the length scale set by the disorder parameter is of the order of the localization length. Such corrections can be treated in a field-theoretical approach by means of an \(\epsilon\)-expansion of the nonlinear \(\sigma\) model in the vicinity of the localization energy [7, 8], or by self-consistent mode coupling expansion techniques [9], which lead to the potential-well analogy [10–13]. These corrections are important for the electronic problem, because the Lifshitz tail mostly consists of the ground states of such wells, for which the irreducible one-particle self-energy is energy dependent.

For phonons the situation is different, because one deals with fluctuating elastic constants instead of potentials, and localized states appear only at the upper band edge [14, 15] for positive values of \(\omega^2\) [16, 17], where \(\omega\) is the vibrational excitation frequency. There is an extended amount of literature concerning the cross-over from the Debye type acoustic wave regime to a regime of diffusive, random-matrix type vibrational excitations (‘boson peak’ [18–20]), which has been accurately described within the \(\sigma\)-model approach by two non-crossing techniques, namely the self-consistent Born [21–24] and coherent-potential approximations [15, 25–27]. Instead, here we are interested in the behavior at large positive frequencies. In the \(\sigma\)-model approach one finds that the one-particle self-energy on the localized side becomes frequency independent, and the system can be assumed to be described by a renormalized Ginzburg–Landau theory [28, 29]. The states with the highest values of \(\omega^2\) within a region of constant elastic modulus, bounded by a mismatch, carry wavenumbers of the order of the Debye wavenumber. Hence these states dominate the Lifshitz tail. If one assumes further the fluctuations of the elastic constants to be small and the localization length to...
scale with the inverse frequency, \( \xi \propto \omega^{-1} \), as in elementary wave mechanics [6], the DOS should be proportional to \( \exp(-\omega \xi - d) \). Such a behavior of the exponent is suggested by recent experiments [30, 31], in which a linear exponential decay of the DOS in the high-frequency region is found.

In the remainder of this paper we will reformulate the field theoretic approach within the Keldysh formulation of quantum dynamics, in order to include the instanton contribution, from which the tail of the DOS can be extracted.

2. Keldysh formulation for weakly disordered phonons

The quantum dynamics for the displacement field \( u(x, t) \) with spatially dependent elastic moduli will be calculated from the Keldysh partition function \( Z = \int D\phi \, e^{iS} \), where the action involves the classical (symmetric) and quantum (antisymmetric) linear combinations of the two fields acting left and right on the density matrix describing the initial state [32]. The reason for naming them classical and quantum is as follows: if the action is related to the Lagrangian of a simple non-relativistic one-particle system, the saddle-point solution of the field theory with \( u_q = u_+ - u_- = 0 \), \( u_{1\pm} = u_+ + u_- = u(t) \) yields Newton’s equation of motion for the particle’s trajectory \( u(t) \). Quantum corrections like the appearance of a phase or tunneling contributions arise from the finite expectation values and higher correlations of the antisymmetric \( u_+ - u_- \) linear combination, which is hence named the quantum component.

The action may involve arbitrary nonlinear terms, which in a classic kinetic approximation give rise to phonon thermalization, at least in three dimensions. For the vibrational spectra of disordered systems the anharmonic interactions are only important below the boson peak [33, 34]. For our purpose it is hence enough to approximate the Keldysh action to quadratic order in the displacement field,

\[
S = \int dx \left[ \frac{1}{2} \mathbf{u} \partial_t^2 \mathbf{u} + \mu \partial_t \mathbf{u} + \lambda \mathbf{u} \partial_t \mathbf{u}^\dagger + \mathbf{u} \partial_t \mathbf{u}^\dagger \right] + \int dx \mathbf{u}^\dagger \mathbf{G}^{-1} \mathbf{u}
\]

(1)

where \( x = (x, t) \), \( u_{ij} = \frac{1}{2} (\partial_i u_j + \partial_j u_i) \) is the usual strain-tensor, and \( \lambda \) and \( \mu \) are Lamé’s elastic constants, which are assumed to fluctuate due to the structural disorder of the material [21–25]. We use units in which the mass density equals unity. The Keldysh component \( G_k \) characterizes the actual state of the system, which can be determined from knowledge of the retarded and advanced Green’s functions and a proper initial condition. If one assumes that the disorder does not alter the thermalization process, the Keldysh component can safely be replaced by a thermal distribution. The formulation (1) ignores further initial correlations.

The advantage of Keldysh’s closed time-contour is the absence of vacuum contributions to the partition function; hence, one can average it directly over an arbitrary distribution of elastic constants. This average can be performed formally by characterizing the probability distribution through its irreducible correlation functions

\[
K(x_1, \ldots, x_n) = \frac{\delta}{\delta x_1} \cdots \frac{\delta}{\delta x_n} \ln \int d\mu P(\mu) e^{S(\mu)},
\]

(2)

which leads to an action of the form

\[
S_{\text{dis}} = S - S(\mu_i) = + \int_{x_1x_2} K_\mu(x_1, x_2) u_i^\dagger(x_1) u_j(x_2) u_i q_{lm}(x_1) u_j q_{mn}(x_2)
\]

\[
+ \int_{x_1x_2} K_\lambda(x_1, x_2) u_i^\dagger(x_1) u_j(x_2) u_j q_{ij}(x_1) u_j(x_2)
\]

\[
\times u_i q_{ij}(x_1) + \cdots.
\]

(3)

For the Gaussian theory the structure of the action is the same as for a dissipative quantum system [35], in which the spectral function of the heat bath is replaced by two classical strain fields. Therefore the calculation of instanton solutions in disordered systems exhibits a strong resemblance to the calculation of thermal activation of a system attached to a heat bath [35].

Within this paper we do not want to perform technically detailed calculations. We rather want to demonstrate the general procedure of mapping the disordered bosonic system onto a disordered fermionic one within the Keldysh prescription. Of course this mapping is only possible between the pair modes of the bosonic and fermionic systems.

In general, the theory can be represented in terms of two-point functions using the Faddeev–Popov transformation

\[
F \left[ u_i^\dagger(x_1) u_j q_{ij}(x_2) \right] = \int DQ_{\Delta} e^{i \text{Tr} \left[ \mathcal{L}(\Delta - a u^\dagger) \right]}
\]

\[
\equiv \int DQ_{\Delta} \Delta F \left[ c_{\mu}^\dagger q_{ijlm}(x_1, x_2) \right] \times e^{i \text{Tr} \left[ \mathcal{L}(\Delta - a u^\dagger) \right]}
\]

Here Greek indices represent the Keldysh degrees of freedom. The following mapping achieves the same causality structure for the composite phonons as for the electrons [35]:

\[
A_R = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad A_L = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}
\]

If the matrix \( A \) has the causality structure of a bosonic Green’s function, \( A_R \) has the c.s. (causality structure) of a fermionic Green’s function and \( A_L \) the c.s. of a fermionic self-energy. The advantage is now that the phonon nonlinear interactions are only important below the boson peak [33, 34]. Within this paper we do not want to perform technically detailed calculations. We rather want to demonstrate the general procedure of mapping the disordered bosonic system onto a disordered fermionic one within the Keldysh prescription. Of course this mapping is only possible between the pair modes of the bosonic and fermionic systems.
transformation. This could be the starting point to formulate the nonlinear σ model approach to phonon localization around the weakly disordered SCBA saddle-point [21].

However, as also discussed by Cardy [29], in the large energy regime, where the phonon energy is large compared to the energy-fluctuations set by the disorder potential, the semiclassical one-particle approximation of the partition function becomes valid. It is more convenient to discuss the Lifshitz tail in terms of the one-particle functions, as one avoids the complicated renormalization-group method.

One would agree that the simple one-particle saddle-point is sufficient, as long as one is interested in the deep localization regime, where knowledge of the mobility edge is lost. In contrast, the universal properties of vibrations in a glass in the vicinity of the mobility edge [16, 17], must be developed from (4) within the usual Keldysh nonlinear σ model.

3. Instanton solutions

In the remainder we will use the simple instanton picture in order to calculate the exponential dependence of the vibrational density of states at high energies.

The first step is to express the partition function as an infinite product of discrete frequencies $1 = \prod_\omega Z(\omega)$, where $Z(\omega) = e^{\omega(\omega)}$ and

$$s(\omega) = \int d^d x \left[ \omega^2 u_q^2 + g_{h} u_q^2 + 2\mu u_q^2 + \lambda u_q^2 \right] + \int d^d x \gamma \left( u_i(-\omega, x) u_i^\dagger(\omega, x) \right)^2. \quad (5)$$

If the frequency is sufficiently large, this field theory can be solved in the semiclassical saddle-point approximation by extremizing the action with respect to the classical and quantum components. In analogy to the theory of dissipative quantum systems there exist instanton solutions in which the expectation value of the quantum component $u_i = i v_i^q$ is finite and purely imaginary.

From the equation of motion (6) one deduces that these solutions describe a situation in which condensation of strain, $v_i = -v_i = -v_i^q$, leads to a finite region with a sound velocity substantially lower than the average. The states are hence trapped in this finite region. There is a second instanton equation describing the other possibility, where the sound velocity is raised. We seek an instanton ansatz where the saddle-point solution satisfies

$$\begin{align*}
(\omega^2 - (\lambda + \mu|v|) \nabla \sigma - \mu|v| \Delta v) & = 0 \\
\mu|v| & = \mu(1 - \gamma v_i v_j). \quad (6)
\end{align*}$$

Reinsertion of this equation of motion (6) into (5) yields a finite action $i\Gamma(\omega)$, and hence an exponential factor of the partition function $e^{-\Gamma(\omega)}$, which is calculated via formula (11).

In order to calculate the density of states and the Green’s function we have to expand (5) with respect to the fluctuations above this instanton saddle-point $u_i^\dagger = v_i + \delta u_i^\dagger$. The irreducible retarded Green’s function is by definition just the correlator of $(\delta u_i^\dagger \delta u_i^\dagger)$. Obviously, this quantity is proportional to the factor $e^{-\Gamma(\omega)}$ which is nothing other than the exponential Lifshitz tail. The action (5) expanded around the instanton saddle-point reads

$$s(\omega) = i\gamma e^{-\Gamma(\omega)} \left[ G_0^{-1}(\omega, x) - \nabla \left( \Sigma + v_i v_j \right) + \Sigma \right] \frac{1}{G_0^{-1}(\omega, x) - \nabla \left( \Sigma + v_i v_j \right)} \times \frac{e^{\frac{1}{2} f(\omega)}}{\delta G_0^{-1}(\omega, x) - \nabla \left( \Sigma + v_i v_j \right)} \frac{1}{\delta \Sigma} \left( G_0^{-1} + \nabla \Sigma \nabla \right) v + S_{\text{kin}}[v] = 0. \quad (8)$$

The next step is to use the bosonization-procedure (5) to express this action in terms of the pair modes $Q_R = (\delta u_i^\dagger \delta u_i^\dagger)$. Using a further saddle-point approximation in order to determine $Q_R$ yields the following set of equations:

$$\langle Q_R(\omega, x) \rangle = i\gamma e^{-\Gamma(\omega)} \left[ G_0^{-1}(\omega, x) - \nabla \left( \Sigma + v_i v_j \right) + \Sigma \right] \frac{1}{G_0^{-1}(\omega, x) - \nabla \left( \Sigma + v_i v_j \right)} \times \frac{e^{\frac{1}{2} f(\omega)}}{\delta G_0^{-1}(\omega, x) - \nabla \left( \Sigma + v_i v_j \right)} \frac{1}{\delta \Sigma} \left( G_0^{-1} + \nabla \Sigma \nabla \right) v + S_{\text{kin}}[v] = 0. \quad (10)$$

These equations have a rather simple interpretation: they allow for a self-consistent determination of the instanton solution in the Hartree-approximation and a further non-crossing approximation of the self-energy of the propagator, which has a frequency and spatially dependent sound velocity, due to the finite instanton amplitude. However, we are mostly interested in a discussion of the exponential factor and leave the numerical solution of this set of equations for future work.

From (8) it becomes clear that within this approximation the density of states is just the usual one in the SCBA approximation, multiplied by the instanton factor $e^{-\Gamma(\omega)}$. In order to estimate the power law satisfied by the exponent of the DOS we look for a spherically symmetric solution with longitudinal polarization in three dimensions.

The equation for the radial part reads

$$\begin{align*}
\omega^2 \Psi(r) + (1 - 4\gamma (\partial_r \Psi)^2) \Delta_r \Psi(r) & = 0. \quad (12)
\end{align*}$$

The frequency and the disorder parameter can immediately be scaled out according to $\phi = \frac{1}{\sqrt{\omega}} \Psi(\omega r)$, where the spatially dependent part $\Psi(y = \omega r)$ satisfies the reduced equation

$$\begin{align*}
\Psi(y = (1 - 4(\partial_y \Psi)^2) \Delta_y \Psi(y) & = 0. \quad (13)
\end{align*}$$

For numerical solution one replaces (13) by the first-order system

$$\begin{align*}
\partial_t \Psi(r) & = \phi(r) \\
\partial_r \psi(r) & = \Psi(1 - 4\phi(r)^2)^{-1} - 2r^{-1} \phi(r).
\end{align*}$$
which may readily be solved by means of a second-order Runge Kutta algorithm.

The phase can be estimated at large frequencies

$$f = \int d^d r \phi(r)(\omega^2 + \Delta^2)\phi(r)$$

$$= \frac{1}{\gamma} \int d^d y \Psi(y)(1 + \Delta_y)\Psi(y)$$

$$= \int_{q < q_{D,0}} \frac{d^d q}{(2\pi)^d} \Psi(q)(1 + q^2)\Psi(q)$$

$$\omega \to \infty : \frac{1}{\gamma} \left( \frac{\omega}{\omega_D} \right)^{4-d} \int \frac{d^d q}{(2\pi)^d} \Psi(q)\Psi(q)$$

$$= \frac{1}{\gamma} \left( \frac{\omega}{\omega_D} \right)^{4-d}.$$  \hspace{1cm} (14)

From (13) it is clear that there exists a localized exponential solution, as long as the state has a single point where the condensed dimensionless strain exceeds the critical value $\delta_{\nu}\Psi(y) = \frac{1}{2}$, which can always be imposed as a boundary condition. Note that $q$ is not the Fourier component of the spatial variable $r$, but of the frequency scaled variable $y = \omega r$.

In contrast to the electronic calculation, the phase is dominated by the small-distance behavior of the wavefunction. As a result we find the same power law of the exponent as predicted by the Lifshitz-argument.

Figure 1 shows a spherically symmetric transversely polarized solution $\tilde{u}(y = \omega r) = \Psi \tilde{\theta}$ to the instanton equation (5), together with its radial derivative $\phi(y = \omega r) = \partial_r \Psi(y)$. The one-particle states turn out to be localized, because they drop to zero faster than the spherically symmetric volume element (which is proportional to $r^2$). The corresponding localization length would be a number of the order of several wavelengths. The inset shows the density of vibrational states normalized to the Debye law, measured by Baldi et al [36] in vitreous silica. The red curve is a previous fit with the SCBA theory, which used the disorder parameter $\gamma = 0.99\gamma_c$ close to the critical value $\gamma_c = 0.1764$. The blue curve shows the Lifshitz tail with the same disorder parameter. The amplitude of the exponential has been adjusted so that the high-frequency density of states agrees with the measured one. It can be seen that the Lifshitz line fits the experimental data beyond the boson peak quite well. From a theoretical standpoint one may state that the boson peak and the strongly localized one-particle states are related, due to the fact that the SCBA collects a set of strong scattering processes which are analytic in $\gamma$, and the instanton approximation is obviously a resummation of scattering processes which are analytic in $1/\gamma$. In this sense the boson peak is the precursor of the localization transition.

The high-frequency deviation from the experimental data is not captured by the present phenomenological theory. A possible reason is that the localization is related to ‘singular’ points within the material, where the elastic constant drops to zero. To account for this, non-Gaussian effects have to be considered in order to prevent a negative elastic constant.

This corresponds to the fact that the SCBA cannot account quantitatively for the magnitude of the boson peak [36]. It has been shown recently, using the coherent-potential approximation (CPA), that indeed non-Gaussian distributions of elastic constants are able to account for the strong boson peak enhancement of the density of states in oxide glasses [27].

Let us make a general statement on the nature of the vibrational excitations in disordered materials. Below the boson peak these excitations are Debye waves due to the fact that for scales large compared to an interatomic distance the material is homogeneous and isotropic. For smaller length scales, i.e. higher frequencies, the boson peak indicates the breakdown of the wave concept [24].

The vibrational excitations are no longer described by the translational and rotational symmetry. Instead they can be characterized as random-matrix states, which obey the Gaussian-orthogonal-ensemble spectral statistics [15]. At even higher frequencies the localization transition occurs, which is a transition within the random-matrix scenario. This step-wise transition from wave physics first to diffusive motion and then to localization is well known for electrons in a random potential [37, 38].

4. Summary and conclusion

In this work we formulated the theory of weakly fluctuating elastic constants of a glass within the Keldysh-formalism, in order to propose an expression for the high-frequency behavior of the density of states of a glass. In three dimensions, where this approach is valid, it yields an exponentially decaying density of states. This exponential decay is expected by basic Lifshitz-like considerations, and has also been observed in recent experiments [30, 31]. If one measures this exponential decay in an experiment, it is therefore straightforward to extract the disorder parameter $\gamma$.\hfill

\hspace{1cm} Figure 1. Numerical solution to the instanton equation (5); for details see the text. Inset: the density of states, divided by the Debye density of states, against the frequency $\nu = \omega/2\pi$. Symbols: spectral data measured in vitreous silica [36]; red line: previous calculation in the self-consistent Born approximation (SCBA) using the disorder parameter $\gamma = 0.99\gamma_c$ with $\gamma_c = 0.1764$ [23]; red line: Lifshitz tail calculated according to the present theory with the same $\gamma$ value. For details see the text.
which can be compared with the values extracted from experiments and simulations for the description of the boson peak [21–25].

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