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Model calculations for vibrational properties of disordered solids and the "boson peak"

Walter Schirmacher[a,*], G. Diezemann[b], C. Ganter[a]

[a]Physik-Department E13, Technische Universität München, D-85747 Garching, Germany
[b]Institut für Physikalische Chemie, Universität Mainz Welderweg 15, D 55099 Mainz, Germany

Abstract

It is demonstrated that a disordered system of coupled classical harmonic oscillators with a continuous distribution of coupling parameters exhibits generally a low-frequency enhancement ("boson peak") of the density of states, as compared with the Debye law. This phenomenon is most pronounced if the system is close to an instability. This is shown by means of a scalar model on a simple cubic lattice. The force constants are assumed to fluctuate from bond to bond according to a Gaussian distribution which is truncated at its lower end. The model is solved for the density of states and the one-phonon dynamic structure factor $S(q,\omega)$ by applying the two-site coherent potential approximation (CPA). The results for the density of states are in very good agreement with a numerical evaluation of the same model. © 1999 Elsevier Science B.V. All rights reserved.

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The origin of the anomalous low-energy vibration spectrum of disordered solids is a puzzle with which scientists are concerned already for a long time [1]. In particular, it was found that the vibrational density of states (DOS) $g(\omega)$ exhibits an excess low-frequency contribution as compared to the Debye ω^2 law, which, when plotted as $g(\omega)/\omega^2$, appears as a maximum, the so-called boson peak. A corresponding low-temperature peak is observed in the temperature dependence of the specific heat if plotted as $C(T)/T^3$ [2].[1] Very recently, the present authors have shown [3] that a strongly disordered three-dimensional model of coupled harmonic oscillators with a continuous force constant distribution exhibits such an excess low-frequency DOS (boson peak) as a generic feature. This was achieved by comparing the results of a numerical diagonalization with those obtained by the single-bond coherent-potential approximation (CPA). In the present contribution we review the main results of this work. Furthermore, we show, how to obtain a CPA expression for the coherent one-phonon dynamic structure factor $S(q,\omega)$, and discuss the general trends of the experimentally observed boson peak phenomena. Our model consists of a set of coupled scalar harmonic oscillators placed on a simple cubic lattice with lattice constant $a = 1$. The oscillators are coupled by nearest-neighbour force

* Corresponding author. Fax: + 49-89-289-12473; e-mail: wschirma@physik.tu-muenchen.de.

[1] See Ref. [3] for an extended list of recent experimental and theoretical references.

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constants K_{ij}, which are treated as independent (quenched) random variables, distributed according to a density $P(K_{ij})$. The corresponding Hamiltonian has off-diagonal elements $\mathscr{H}_{ij} = K_{ij}$, diagonal elements $\mathscr{H}_{ii} = -\sum_{j\neq i} K_{ij}$ and eigenvalues $\lambda_i = -\omega_i^2$, where ω_i are the vibrational eigenfrequencies. For $P(K)$ we have chosen a truncated Gaussian $P(K) = P_0 \exp\{-(K-K_0)^2/2\sigma^2\}\theta(K-K_{\min})$. Here $\theta(x)$ denotes the step function, P_0 is a normalization constant, K_0 and σ denote the maximum value and the width, respectively. $\sqrt{K_0}$ serves as frequency scale (i.e. $K_0 = 1$ in our units). The lower cut-off $K_{\min}$ is introduced to allow for the study of strongly disordered systems with a reduced amount of negative force constants [4].[2]

In Fig. 1 we present the results of a calculation [3] of $g(\omega)/\omega^2$ obtained in CPA (lines) and by numerical diagonalization (symbols) for various values of $K_{\min}$ with $\sigma = 1.0$. For comparison the spectrum of the ordered lattice ($\sigma = 0$) is also shown. The excellent agreement of the CPA calculations with the numerical analysis (except for the immediate vicinity of the instability) indicates both the reliability of the CPA and the correctness of the procedure utilized for the elimination of finite-size effects in the numerical work [3]. It is seen that the position and strength of the boson peak is determined by the amount of negative force constants, which is controlled by the parameter $K_{\min}$. We found out that the system becomes unstable if $K_{\min}$ exceeds a critical value (-0.85 in CPA, -0.6 in the numerical analysis). Obviously, the boson peak plays the role of a precursor phenomenon of the instability.

As demonstrated in Ref. [3], as well as in earlier model calculations [2,4,5,10], the boson peak is associated with a strongly reduced mean free path, which is of the order of the wavelength. As shown in Ref. [3] by means of a statistical analysis of the spectral fluctuations, the states in the boson peak region are neither propagating nor localized, indicating a diffusive type of transport of vibrational energy. Such a diffusive motion of vibrational excitations has already been shown [6,7,11–13] to be typical for glasses and to be responsible for the temperature dependence of the thermal conductivity above the plateau region.

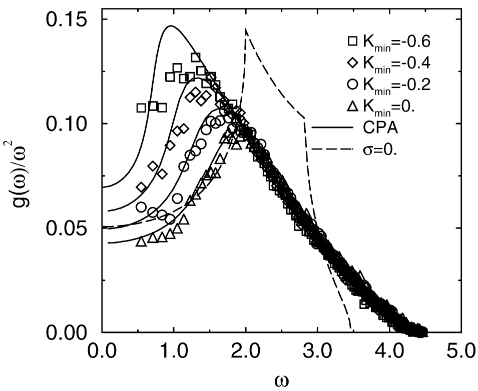

Fig. 1. Reduced DOS $g(\omega)/\omega^2$ versus frequency for force constant distributions with $\sigma = 1$ for several values of $K_{\min}$, calculated in CPA (lines) and by direct numerical diagonalization (symbols). The corresponding curve for the ordered lattice ($\sigma = 0.0$) is also given.

Experimentally the boson peak does not only show up in the vibrational DOS, which is proportional to the incoherent dynamical structure factor $S_{\mathrm{inc}}(\mathbf{q},\omega)$, but also in its coherent version $S_{\mathrm{coh}}(\mathbf{q},\omega)$. The one-phonon contribution to the latter quantity can be calculated in CPA. The central quantity in this theory is the self energy $\Gamma(\omega) = (v(\omega)/a)^2$. $v(\omega)$ is the frequency dependent complex sound velocity. The coherent one-phonon dynamic structure factor is given by [8]

$$S_{\mathrm{coh}}(\mathbf{q},\omega) = (n(\omega)+1)\frac{\omega}{\pi} \times \frac{2f(\mathbf{q})\mathrm{Im}\{\Gamma(\omega)\}}{[\omega^2 - f(\mathbf{q})\mathrm{Re}\{\Gamma(\omega)\}]^2 + [f(\mathbf{q})\mathrm{Im}\{\Gamma(\omega)\}]^2}, \quad (1)$$

where $n(\omega)$ is the Bose distribution and $f(\mathbf{q})$ is the structure function. On a simple cubic lattice it is given by $f_{\mathrm{sc}}(\mathbf{q}) = 2\sum_{i=x,y,z}(1-\cos(aq_i))$. To obtain a model expression that may be used for structurally disordered solids, we performed angular

[2] It should be realized that the Hamiltonian $\mathscr{H}$ can be stable (i.e. have no positive eigenvalues λ_i) for a restricted amount of negative off-diagonal elements K_{ij}. Physically this corresponds to a situation, where atoms are occasionally connected by negative force constants, but are still sitting at the bottom of a potential energy well.

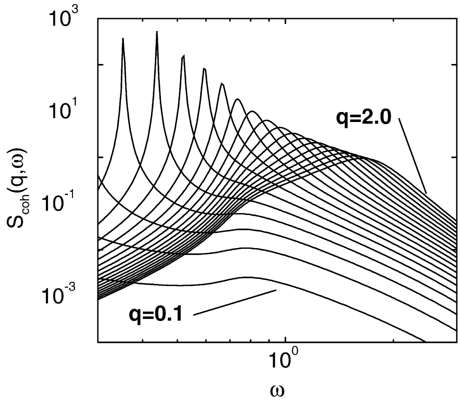

Fig. 2. Coherent dynamic one-phonon structure factor $S_{\mathrm{coh}}(q,\omega)$ ($\sigma = 1, K_{\min} = -0.6$) for wavenumbers ranging between 0.1 and 2.0 ($a = 1.0$).

averages of Eq. (1) with $f(\mathbf{q}) = f_{\mathrm{sc}}(\mathbf{q})$. We found out that the results cannot be distinguished from those obtained by using the angular averaged quantity $f_{\mathrm{av}}(q) = 6(1 - \sin(qa)/qa)$ in (1). The results for $\sigma = 1.0$ and $K_{\min} = -0.6$ are presented in Fig. 2. It is seen that the boson peak (as calculated in CPA) is a wavenumber independent phenomenon. The Brillouin peak is broadened considerably with increasing wavenumber.

At the end we would like to comment on a trend in the experimentally observed low-frequency vibrational spectra which was realized recently by Sokolov et al. [9]: They observe that the boson beak ist much more pronounced in "strong" glasses, i.e. network-type materials, whereas it is hardly visible in "weak" glasses, i.e. in materials, which are interacting via hard-core interactions. We find from our CPA investigations that the strength of the peak strongly depends on the atomic coordination number. If the atomic coordination is low, a single negative force constant renders the atomic arrangement much closer to an unstable situation than in the highly coordinated case. We therefore believe that the strong appearance of the boson peak in the network materials means that they are much nearer to an instability, i.e. much more frustrated than the weak materials.

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