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Propagation and localisation of vibrational modes in 3-dimensional disordered systems: the binary force constant model

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Abstract. We consider a system of coupled harmonic oscillators on a cubic lattice. The force constants are supposed to take two distinct values at random according to a bond concentration x. The density of states (DOS) is evaluated both by numerical diagonalisation and in coherent-potential approximation (CPA). There is excellent agreement between the results of the two methods. Near the concentration, where the bonds with the larger force constants percolate, the DOS differs appreciably from the crystalline one and is anomalously enhanced at low frequencies as compared to Debye's ω^2 law ("boson peak"). These features are shared with models with continuous distributions of force constants. The mean free path near the strongest anomaly is comparable to the wavelength of the excitations. However, the level distance statistics reveils that the corresponding states are delocalised.

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1 Introduction

The anomalous low-frequency vibrational properties of disordered solids are still poorly understood, although they are subject to an enormous amount of theoretical and experimental investigations on this subject [1]. A controversal point in the discussion in this field is, whether the observed excess vibrational density of states (DOS) $g(\omega)$ (with respect to Debye's ω^2 law, "Boson peak") is due to localised or extended vibrational modes. This question is important, because it is related to the mechanism of thermal transport in the ~ 10 K temperature range [2]. Recently the present authors have shown that a model of coupled harmonic oscillators placed on a simple cubic lattice with spatially fluctuating force constants K produces a boson-peak like anomaly. As distribution of force constants $P(K)$ we took a Gaussian which was truncated on its negative side to avoid instability. The model was solved for the DOS both by numerical diagonalisation and using the single-bond coherent potential approximation (CPA). The results of both methods are in good agreement with each other. In the CPA one can extract a frequency-dependent sound velocity (acoustic index of refraction), which, at low frequency can be used to calculate the mean free path and the wavelength of acoustically propagating excitations. In the case of strong disorder our calculations

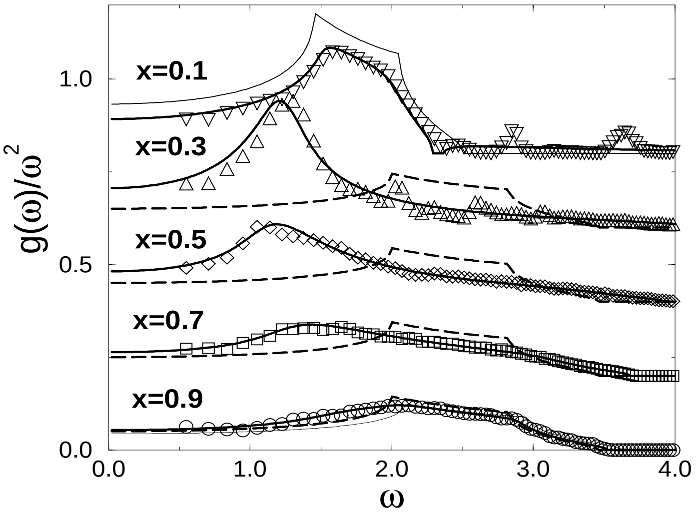

Fig. 1 Reduced DOS $g(\omega)/\omega^2$ vs. frequency for $K_2/K_1 = 0.1$ and concentrations $x = 0.1, 0.3, 0.5, 0.7$, and 0.9 (from top to bottom). Frequencies are given in units of the square root of the average force constant, which is normalised to unity. Symbols: diagonalisation results; thick lines: CPA; thin line: regular DOS for the K_2 sublattice ($x = 0.1$); dashes: regular DOS for the averaged force constants. (For clarity the curves for $x \leq 0.7$ have been shifted upwards.)

revealed that the mean free path is of the order (and even smaller) than the wavelength in the boson peak frequency range. According to the Ioffe-Regel criterion these modes may localise. An investigation of the level distance statistics, however, showed that in this regime the states are *delocalised*. This implies that the corresponding contributions to the thermal transport are of diffusive nature. These findings agree with simulation work on amorphous silicon [3] and a percolation model [4].

In order to find out how general these features are, i. e, whether they are typical for disordered solids, it is important to investigate different kinds of models. In the present contribution we consider a model with a binary distribution of force constants. We shall demonstrate, that at the concentration at which the larger of the coupling constants percolate the system is "most disordered", which means that the van-Hove singularities are completely washed out and the boson peak is most pronounced. As in the models with a continuous force constant distribution the states in the boson peak frequency range are shown to be neither propagating nor localised.

2 Model and results

We consider a set of coupled (scalar) harmonic oscillators with nearest- neighbour force constants K on a simple cubic lattice. K is a random number distributed according to the density $P(K) = x\delta(K - K_1) + (1 - x)\delta(K - K_2)$. As in ref. [5] we compare a nu-

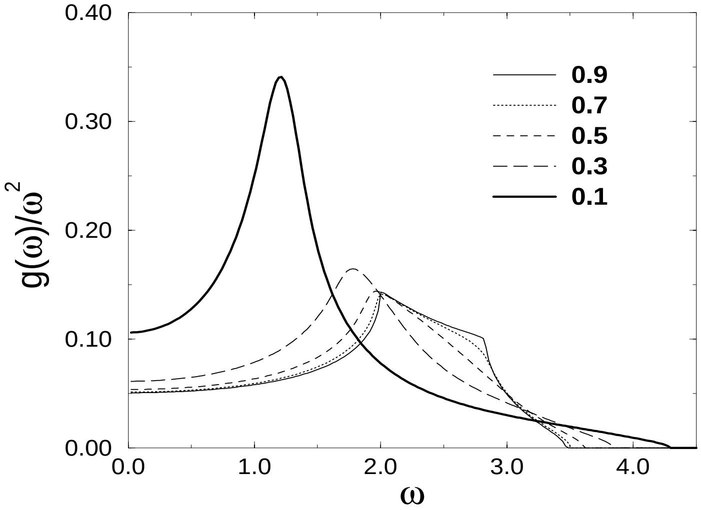

Fig. 2 Reduced DOS $g(\omega)/\omega^2$ vs. frequency for different ratios K_2/K_1 calculated in CPA

merical diagonalisation with CPA results. Except for the distribution all calculational details are as in ref. [5]. We normalise the K_i such that $< K >= xK_1+(1-x)K_2 = 1$. For the numerical work we consider only the case $K_2 \ll K_1$ and take the ratio as K_2/K_1=0.1.

In fig.1 we have plotted the DOS, divided by the frequency squared, i. e. $g(\omega)/\omega^2$ for five values of the concentration x. Clearly the diagonalisation results are in excellent agreement with those of the CPA. By comparing the curves corresponding to the different concentrations it is striking that for $x = 0.3$ the van-Hove singularities are washed out completely and a pronounced low-frequency "boson peak" appears, which, to our opinion is a benchmark for strong disorder. It is interesting to note that for $x = 0.1$ the DOS is not determined by the average of the force constants, which would be the prediction of the "virtual crystal approximation", but is given by the "host" force constant K_2 alone. The stiff force constants K_2 act obviously only as inert scattering centers.

In the following we are going to investigate the case $x = 0.3$ in more detail. In fig. 2 we have plotted $g(\omega)/\omega^2$ as calculated in CPA for different values of the ratio K_2/K_1. The change in the DOS from the regular to the strongly disordered case is most striking. In order to show that for $K_2/K_1 = 0.1$ and $x = 0.3$ the disorder is really strong we have calculated in CPA the mean free path given in terms of the frequency dependent complex sound velocity [5] $v(\omega)$ as $\ell(\omega) = |v(\omega)|^2/2\omega\text{Im}\{v(\omega)\}$ and compared it with the sound wavelength $\lambda(\omega) = 2\pi\text{Re}\{v(\omega)\}/\omega$ (see fig. 3). Comparing fig. 1 with fig. 3 it becomes clear that the boson peak coincides with the frequency where ℓ becomes comparable to λ. The Ioffe-Regel criterion now would suggest that the states beyond the frequency, where ℓ becomes equal to λ are localised. We have investigated this question by evaluating the level distance statistics. We deconvoluted the eigenvalues

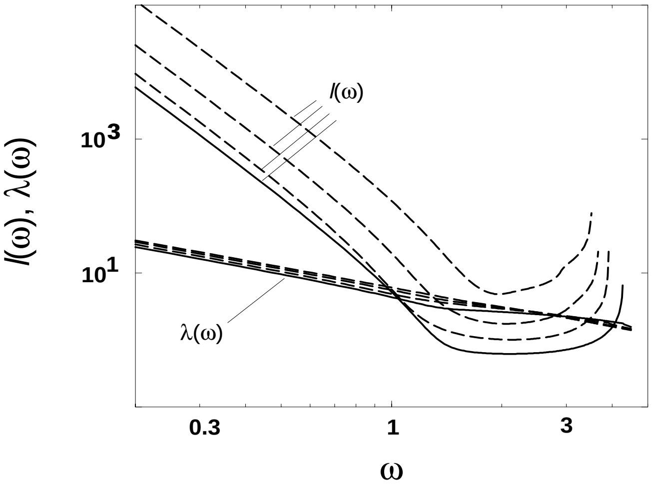

Fig. 3 Mean free path $\ell(\omega)$ and wavelength $\lambda(\omega)$ as defined in the text. Dashes: $x = 0.9, 0.7, 0.5$ (from top to bottom). Full lines: $x = 0.3$.

$\lambda_i = \omega_i^2$ using the averaged integrated DOS as described in [5] in order to obtain the normalised level distances s. The statistics of these distances was obtained by taking 11 samples of side length 14. The frequencies given in the legend of fig. 4 are the mid-points of frequency intervals of width $\Delta\omega = 0.5$ in which the statistics has been evaluated. It is clear from the figure that in the boson peak range $\omega \leq 1.75$ the GOE statistics is obeyed, i. e. the states are delocalised. Only at higher frequencies in the vicinity of the upper band edge the Poissonian statistics is approached, indicating the presence of a mobility edge near $\omega = 4$. This means that the boson peak in the binary force constant model is not associated with localized states.

3 Discussion

It is now interesting to discuss previous theoretical approaches for explaining the boson peak phenomenon in the light of the present findings and those of ref. [5]. The first was the so-called phonon-fracton model [6, 7]. Alexander and Orbach [6] considered a percolating system (in terms of our model $K_2/K_1 = 0$). They argued that for $x > x_c$, x_c being the percolation threshold, there should be a crossover from sound propagation (phonon regime) to localisation (fracton regime) at $\omega_c = 2\pi v/\xi$, where ξ is the correlation length and v the sound velocity. For $\omega > \omega_c$ the system should behave as for x_c, where a scaling analysis shows that the DOS varies as $g(\omega) \propto \omega^{\tilde{d}-1}$ with $\tilde{d} \approx 1.33$ (spectral or fracton dimension). As the states on the fractal percolation cluster are supposed to be localised it was conjectured that the boson peak in glasses is an indication of delocalisation-localisation crossover as it was supposed to occur in the percolation model. These ideas were qualified when extensive simulation work

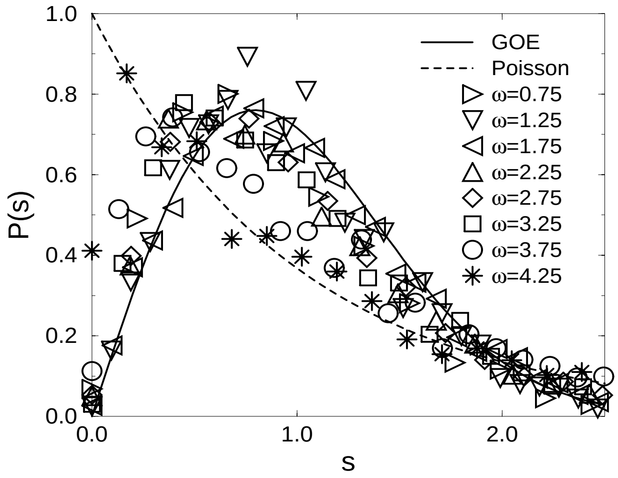

Fig. 4 Level distance statistics for $x = 0.3$ taken in frequency intervals of width $\Delta\omega = 0.5$ around the frequency values indicated. The boson peak is located at $\omega = 1.25$, where the statistics follows the GOE. The mobility edge is near $\omega = 3.75$.

for the percolation model [7] showed no sign of a boson peak near ω_c. Furthermore a recent numerical investigation of the same model performed by Kantelhardt et al. [4] reveiled that for $x > x_c$ an appreciable frequency range above ω_c exists, in which the states are still *delocalised* ("extended fractons"). Regarding the results of the present model it is remarkable that the boson peak, which is absent for $K_2 = 0$ appears for finite K_2. Obviously it is essential for the appearance of the boson peak that the "almost localised" vibrations are coupled to each other. However it is still reasonable to associate the boson peak with a crossover phenomenon, namely that between propagation and diffusion. In the propagating regime the states are forced to form a coherent wave due to the sum rule $\sum_j \mathcal{H}_{ij} = 0$ (see [5]),whereas in the diffusive regime this restriction is lost due to the disorder, which in turn produces the additional DOS. In the terminology of Allen and coworkers [3] one could call it a "phonon-diffuson crossover".

Another approach devised for the explanation of low-frequency vibrational anomalies in disordered solids is the soft potential model [8]. In this model atomic configurations in an effective quartic potential are considered with statistically distributed coefficients of the quadratic term, leading, at random, to very soft double and single-well configurations. The coupling of the modes corresponding to these local potentials to acoustic phonons are made responsible for the boson peak [9]. The model has the attractive feature that it accounts both for the T^{-1} behaviour of the low-temperature specific heat of most glasses and the boson peak. As emphasized in ref. [9] in the soft-potential scheme the boson peak is a quasi-harmonic phenomenon, i. e. essential for the boson peak is the softness of the renormalized potentials and not the anharmonicity. Within the present model and that considered in [5] the presence of very small (and even negative) force constants produce the boson peak. So this physical picture agrees to that of the soft potential model. One could call the present approach a "harmonic soft potential model". On the other hand it is difficult to fix the many adjustable parameters appearing in the soft-potential model in terms of microscopic physical quantities. The force-constants of the present model can in principle be related to interatomic forces. The force-constant distribution can then be related to the interatomic pair distribution function $g(r)$, as done in earlier effective-medium calculations [10].

A third approach which gives an explanation of the boson peak phenomenon is the mode-coupling theory of the liquid glass transition [11]. In this theory a transition from an ergodic phase (liquid) to a nonergodic one (glass) is described as a function of a control parameter. In the glass phase the density fluctuation spectrum reveals a characteristic maximum [12, 13], which appears, if compared to experimental data, in the boson peak range. As the physical mechanism of the liquid-glass transition is the formation of a cage by the particle interaction, the predicted peak can be interpreted as due to the vibrational modes of the particle motion inside the cages. The physical picture that the boson beak arises from the frozen disorder is the same as in the present approach, but it is difficult to make a more detailed comparison, because the present treatment is based on vibrational displacements, whereas the mode-coupling theory on density fluctuations.

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