Model Calculations for the Vibrational Anomalies of a Disordered Lennard-Jones Solid

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Abstract

Using a modified version of the 2-site coherent potential approximation (CPA), also known as effective-medium approximation (EMA), we calculate the density of states (DOS) of a disordered rare gas solid modelled by a Lennard-Jones potential. As input we use the radial pair distribution of a pertinent molecular-dynamics simulation of Rahman et al. The resulting DOS agrees well to that of the simulation. The DOS exhibits an anomalous enhanced low-frequency contribution ("boson peak"). We investigate and discuss the influence of the quenched structure on this anomaly and on the resulting sound velocity.

Keywords: disordered solids;low-frequency vibrational anomalies;boson peak;sound velocity

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The low-frequency vibrational anomalies of disordered solids, especially the enhancement of the density of states (DOS) with respect to the Debye law has attracted much interest in the last years. Recently the present authors have shown [1] by means of a model calculation that this enhancement is due to the frozen disorder. The model employed consists of a set of coupled harmonic oscillators placed on a simple cubic lattice with a continuous distribution of fluctuating force constants $P(K)$. Within this model only scalar force constants are considered. The DOS of the model was obtained both by numerical diagonalization and by applying the two-site coherent-potential approximation (CPA) or effective-medium approximation (EMA). There is good agreement between the results rendering the CPA as a reliable approximation scheme. For $P(K)$ a truncated Gaussian was chosen arbitrarily.

In the present contribution we consider a disordered rare-gas solid modelled by a Lennard-Jones (LJ) pairwise potential $\phi(r) = 4\epsilon[(\sigma/r)^{12} - (\sigma/r)^{6}]$ ("Lennard-Jones glass"). In order to allow for a coordination number $Z_k > 6$ we modify the CPA described in ref. [1] in the following way: Instead of a cubic lattice we consider a d-dimensional hypercubic lattice with $d = Z_k/2$ and replace the resulting diagonal Green's function by that corresponding to a half-elliptic spectrum of the same band width [2]. As force constant distribution we take $P(K)\mathrm{d}K = 4\pi\rho r^2 g(r)|\frac{\partial K}{\partial r}|\mathrm{d}r$. where ρ is the

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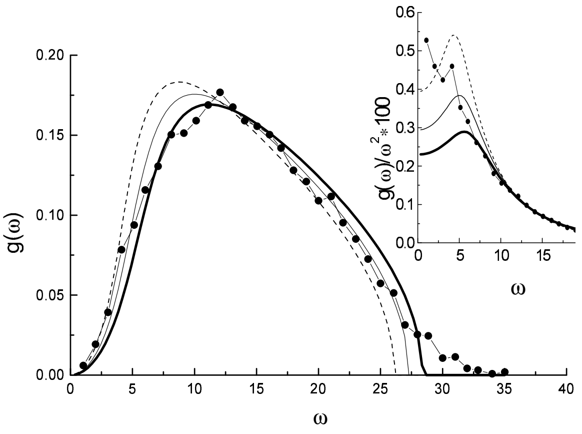

Fig. 1. Full circles: DOS evaluated by Rahman et al. [3] from their MD simulation at density $\rho = 0.95$. Thick full line: CPA calculation using Rahman's $g(r)$ for $\rho = 0.95$. Thin full line and broken line: CPA for $\rho = 0.85$ and 0.75. Insert: The same data, divided by ω^2.

particle density, r the interatomic separation, $g(r)$ is the radial pair-distribution and $K(r)$ is the averaged LJ force constant $K(r) = \frac{1}{3}\phi''(r) + \frac{2}{3}\phi'(r)/r$. For $g(r)$ we use the simulated data of a LJ glass of Rahman et al. [3] with an upper cutoff R_k as adjustable parameter. The coordination number is related to R_k by $Z_k = 4\pi\rho\int_0^{R_k} r^2 g(r)\mathrm{d}r$. As in ref. [3] frequencies are measured in terms of $\epsilon/M\sigma^2$ (M is the atomic mass), and lengths in terms of σ. Performing CPA calculations in the way described above, we found that for large R_k the DOS does no more depend on this parameter and agrees nicely with the DOS of Rahman et al. obtained by diagonalizing the quenched dynamical matrix.

In fig. 1 we show their data for a density $\rho = 0.95$ together with the CPA results for $\rho = 0.95$, 0.85, and 0.75. For these calculations we took $R_k = 8$ (We replaced $g(r)$ by unity in the range $r > 3$, where no computer data are available). The curve with $\rho = 0.95$ shows an overall agreement with Rahman's data. In the insert we show the DOS divided by ω^2. In this way the enhancement with respect to the Debye law ("Boson peak") becomes visible. It was shown in ref. [1] that this peak is due to the presence of very small and negative force constants. This is also true for the LJ glass. From our calculations (see the insert of fig. 1) we find that the "boson peak" becomes more pronounced and that the (average) sound velocity decreases with decreasing density. Such a trend is also observed with decreasing coordination number.

In this context it is interesting to discuss the dependence of the elastic properties of quenched Ne films as measured by the double-paddle oscillator technique [4] on thermal treatment. White et al. [4] find that the shear modulus G, as well as the transverse sound velocity v_t, increases whith annealing temperature. contrarily Classen et al. [4] find a *decrease* of G als well as v_t with annealing.

Although within our scalar model we cannot distinguish between longitudinal and transverse degrees of freedom, our CPA scheme provides a relationship between the sound velocity and the local atomic structure. To our opinion the different experimental results are due to different as-quenched local structures: In one case the as-quenched local coordination numbers (related to the peak height of $g(r)$) are smaller than that corresponding to the metastable equilibrium, whereas in the other case it is larger.

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