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# Boson peak in amorphous silicon: A numerical study

Frank Finkemeier and Wolfgang von Niessen
*Institut für Physikalische und Theoretische Chemie, Technische Universität Braunschweig,*
*Hans Sommer Strasse 10, D-38106 Braunschweig, Germany*

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The low-frequency part of the phonon spectrum of $a$-Si is investigated for a microscopic model of this amorphous solid even for frequencies below 50 $\mathrm{cm}^{-1}$. This requires system sizes up to 64000 atoms. The variation of a model parameter allows us to generate structures with different degrees of disorder. The vibrational properties are calculated in a harmonic approximation to the Stillinger-Weber potential. The less disordered of our models already show an enhancement of the low-frequency vibrational density of states $g(\omega)$ (VDOS) compared to the crystal. For the more disordered structures additionally a peak in $g(\omega)/\omega^2$ versus $\omega$ is observed. The deviation from the Debye $\omega^2$ behavior is caused by at least two effects: a shift of the crystalline transverse acoustic peak toward lower frequency and a broadening of the same. Consequently the occurrence of the boson peak in the Raman spectrum of $a$-Si can be explained already in the harmonic approximation (at least in part) by a complex of phenomena of the low-frequency VDOS.

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## I. INTRODUCTION

Recent years have seen intensive investigation of the properties of the low-frequency vibrational modes of glasses. This is caused by experimental findings of some universally valid features of disordered solids. A prominent example is the so-called ''boson peak,'' a broad band that appears in Raman and neutron scattering spectra of glasses at about 20–120 $\mathrm{cm}^{-1}$ and has remained a phenomenon difficult to understand for quite some time. Related universal properties of amorphous solids are, among others, a linear term in the temperature dependence of the specific heat $C_p$ at temperatures $T<1\,\mathrm{K}$,[1] a bump in $C_p/T^3$ versus $T$ at higher temperatures[2] $(T=5-10\,\mathrm{K})$, and a plateau region in the thermal conductivity.[1] Neutron scattering clearly has revealed that an excess of modes in the low-frequency part of the vibrational density of states (VDOS) $g(\omega)$ compared to the crystalline behavior is the physical cause of the boson peak and its universality.[3]

The boson peak was observed in 1953 in the Raman spectra of silica glasses.[4] Since then numerous Raman experiments have shown the existence of the boson peak for different glass formers like alkali borate glasses,[5] alkali silicate glasses,[6] and halide glasses.[7] Even more basic results are obtained by inelastic neutron scattering experiments. With this technique the existence of the boson peak has been confirmed, e.g., for silica glasses,[8,3] borate glasses,[9] and metallic glasses.[3]

Two phenomenological approaches have been introduced to explain the appearance of the boson peak and the universality of the low-temperature properties of glasses. The first model is the one of two-level tunneling systems (TLS's) of Anderson, Halperin, and Varma[10] and of Phillips,[11] which can be extended to the soft potential model (SPM) of Karpov, Klinger, and Ignat'ev.[12] While the TLS model is able to explain effects only below 1 K, the SPM can be used to understand the occurrence of extra vibrational modes in the frequency range of the boson peak by the introduction of anharmonic soft potentials.[13,14] The second model—the fraction model of Alexander and Orbach[15]—introduces a different kind of excitation for amorphous solids, the fraction. Although quite successful in general this model has difficulties in explaining the excess VDOS at lower frequencies.[16]

There also exists another explanation for the boson peak, which involves scattering of acoustic phonons caused by fluctuations of the density of the amorphous material.[17] This aspect was brought into discussion because the SPM cannot explain differences in the excess vibrational contributions of strong and fragile glass formers, and shows that from the theoretical side also many question are not yet satisfactorily answered.

To clarify these questions by computer simulations, which are also used in the present work, is a difficult task, since the examination of low frequency vibrations is hindered by generic finite size effects. A molecular-dynamics (MD) simulation of an argon glass tried to give an explanation for the boson peak in terms of absorption of transverse phonons by resonant modes of finite extension.[18] Another MD simulation of amorphous silica interprets the boson peak in the framework of fragility.[19] Rather surprisingly two model calculations gave evidence of extra low-frequency excitations within the harmonic model. Schirmacher, Diezemann, and Ganter[20,21] showed the occurrence of the boson peak in a system of coupled harmonic oscillators on a cubic lattice. They used a microscopic model with 2744 atoms and in addition a mean-field approach. Nakhmanson and Drabold[22] were able to reproduce the excess specific heat of amorphous silicon ($a$-Si) by relaxing a MD model followed by a direct diagonalization of the dynamical matrix. Our work can be seen as a continuation of these investigations as it connects the two results into a consistent picture. We also reach much lower frequencies than previously achieved and obtain additional results.

Let us now consider the situation in $a$-Si with which we deal here. Although $a$-Si is not a typical glass, it possesses some glasslike properties, e.g., the low-temperature thermodynamic anomalies.[23–25] But it should also be mentioned that the linear temperature term in the heat capacity of *a*-Si was shown to be ruled by the existence of unpaired electronic states (dangling bonds).[26] On the other hand, these experiments were not sensitive enough to rule out TLS's in *a*-Si.[27]

Neutron scattering showed the occurrence of extra states at low frequencies in the VDOS for *a*-Si,[28] and for the structurally similar amorphous germanium.[29] Moreover, TLS's were found in thin films of *a*-Si and *a*-Ge by measurements of internal friction and shear modulus[30] and in bulk silicon, irradiated by neutrons and in this manner disordered, by examination of the variation of the low-temperature ultrasonic velocity.[31] It was also stated that the boson peak for hydrogenated amorphous silicon (*a*-Si:H) is found in the Raman spectrum[32,33] as a broad background signal with a maximum at 245 $\mathrm{cm}^{-1}$. This signal is found at too high a frequency to be the phenomenon generally designated as the boson peak. In a recent Raman experiment[34] on bulk silicon irradiated by neutrons the boson peak was observed at 114 $\mathrm{cm}^{-1}$.

Much confusion arises from the fact that *a*-Si can be prepared in numerous ways which lead to structurally different products with a large spread in physical properties. Maley, Beeman, and Lannin[35,36] have performed a systematical study of neutron and Raman spectra of *a*-Si samples with a varying degree of disorder. They find a strong correlation of the shape of the transverse optical peak and the bond angle distribution of the sample. In a recent review by Liu and Pohl[30] the focus is more on the low-frequency part of the spectrum. They describe *a*-Si:H films generated by hot wire chemical vapor deposition as the first amorphous solid without any significant low-frequency excitations. They show, on the other hand, the typical glassy behavior of electron beam evaporated *a*-Si. Perfect *a*-Si in the sense of a continuous random network (CRN) is believed not to possess extra modes at low frequencies while in real samples with an average coordination number less than 4 these excitations are present, although less numerous than in typical glass formers such as silica glasses. In a previous paper[37] we addressed this question among others. We dealt with the structure, the VDOS, and the localization of vibrational modes in *a*-Si. For a perfect CRN model of *a*-Si the VDOS was determined by direct diagonalization of the dynamical matrix using the Keating potential, and no extra low frequency states were found.

Although *a*-Si might not appear to be the most suitable system to study properties of an amorphous solid with respect to the low-frequency boson peak and thermodynamic anomalies at low temperatures, it still possesses rich and not yet understood dynamical properties in the low-frequency range. The reason why we choose *a*-Si as a model is its relative simplicity and the availability of good potentials. Only this simplicity allows us to examine structures of the size needed to inspect the low-frequency VDOS.

In the following sections we want to study the influence of disorder on the low-frequency part of the VDOS of a microscopic model of *a*-Si. The vibrational properties are calculated in the harmonic approximation to verify whether the introduction of disorder is sufficient to explain the appearance of the boson peak in the Raman spectrum of *a*-Si. Furthermore, we want to give a detailed description of how the disorder actually influences the shape of the calculated low frequency VDOS.

The content of this paper is as follows. Section II introduces the models and potentials used and gives details of the numerical procedures. In Sec. III we present the results of our calculations for the crystal and different disordered models of *a*-Si. Finally, we discuss our findings in Sec. IV and draw some conclusions.

## II. METHODS AND COMPUTATIONAL DETAILS

To generate the structural models we use the well-known bond switching model of Wooten, Winer, and Weaire[38] with periodic boundary conditions. Starting with the crystalline structure topological disorder is introduced via a bond switching algorithm. The structure is then first relaxed into a minimum of the Keating potential[39] with the help of a Monte Carlo method. The Keating potential applies a static neighbor list to determine whether an interaction between two atoms occurs. Only the use of this static neighbor list allows us to introduce disorder by switching bonds. Moreover it guarantees a fast convergence to a local minimum away from the crystalline coordinates. At this point the structures are still CRN's with purely fourfold coordination.

To introduce coordination defects, which are present in real samples of *a*-Si, a further relaxation using the Stillinger-Weber (SW) potential[40] is done via a conjugate gradient algorithm. In contrast to the Keating potential the SW potential has a natural cutoff of interaction. That causes a dynamical change of the neighbor list during the relaxation process. A consequence of this is a dramatic increase of computational costs compared to the first relaxation step. To assure that the structure arrives at a true local minimum in a reasonable time, a careful optimization of the conjugate gradient algorithm has to be performed. This task is extremely important, since the values of the low-frequency vibrations were found to be influenced strongly by the quality of the relaxation.[37] Although the use of empirical potentials could be seen as a shortcoming of this work, it is crucial to arrive at the system sizes needed to investigate low-frequency dynamics. At present we can calculate the low-frequency VDOS's of systems containing up to 64 000 atoms on a normal workstation (DEC Alpha 533au).

To characterize structures with different degrees of disorder we introduce the bond switching parameter $c_n$. It is just a parameter of our algorithm and has no direct physical meaning. A bond switching parameter $c_n=0.1$ results in a structure that is still very close to the crystalline case, whereas a value of $c_n=0.25$ gives a structure with a large amount of nonfourfold-coordinated atoms. The percentage of nonfourfold atoms is 2.5% for $c_n=0.1$, 9.5% for $c_n=0.15$, 18% for $c_n=0.2$, and 28% for $c_n=0.25$. The number of floating bonds is larger than the one of dangling bonds. Although a realistic value for the concentration of coordination defects (for dangling bonds 0.1–1.0%) appears to be much lower, it is up to now not possible to build a computer generated CRN with a lower amount of defects and a satisfactory reproduction of the radial distribution function at the same time. To achieve this, a new algorithm to introduce the disorder would be necessary. The bond switching model, which we use in this work, is accepted as the best algorithm available at this time. The model with $c_n = 0.2$ is believed to be in best accord with real samples of *a*-Si, which are far away from the crystalline structure. A more detailed description of these findings along with the respective radial distribution functions and bond angle distributions was published earlier by the authors.[37]

The VDOS is calculated via direct diagonalization of the dynamical matrix, obtained from a Taylor series expansion of the SW potential. The lowest eigenvalues are calculated using the Lanczos method[41] and then sorted into sample intervals with a width of 10 $\mathrm{cm}^{-1}$ to obtain the VDOS. Due to the finite size of the system a smaller sample interval width would lead to statistical noise in the graph of the VDOS. Another effect of the finite size of our simulation cell is a cutoff $\omega_{\min}$ for the smallest possible frequency of an acoustic phonon. Given the length of the simulation cell edge of 108.54 Å and a value for the transverse sound velocity $c = 3.67\times 10^3$ m/s from the literature,[42] we calculate $\omega_{\min} \approx 11\,\mathrm{cm}^{-1}$. As a consequence we do not show results for frequencies smaller than $\omega_{\min}$. To compare the VDOS's of different system sizes and degrees of disorder it is always normalized to unity.

The calculation shows that the boson peak appears at rather low frequencies. Thus for a proper investigation of the differences from the crystal, it is necessary to develop a phonon band structure code using the same model for Si and the SW potential. The VDOS of the crystal is computed by integrating over $2\times 10^7$ points of the irreducible Brillouin zone with a sample interval width of 2 $\mathrm{cm}^{-1}$. The reduced sample interval width compared to the calculations for *a*-Si is made possible by the larger number of collected eigenmodes.

## III. RESULTS

The SW potential is known to lead to a reasonable description of the VDOS of amorphous silicon although the peaks are shifted compared to experimental findings.[42] To demonstrate this, we have calculated the VDOS of crystalline silicon. It is shown together with the experimental curve for polycrystalline silicon of Kamitakahara *et al.*[28] in Fig. 1. To compensate for the shift of the absolute positions of all peaks, we used two different frequency scales. While this compensates the slightly higher values for the position of the transverse acoustic, longitudinal acoustic, and longitudinal optical peaks, the relative position of the transverse optical peak from our calculation is still at too high a frequency. However, the shape of the VDOS is reproduced well. From this we conclude that the SW potential can even be used to model the crystal, since the differences from experiment could be eliminated by an optimization of the parameters of the SW potential, which has been fitted to reproduce optimally several properties of silicon. Such an optimization would not change any of the qualitative implications of our work. This is why we used the original values of the parameters.

In Fig. 2 we present the low-frequency VDOS for a system size of 64 000 atoms for different degrees of disorder. Since the VDOS for frequencies larger than 100 $\mathrm{cm}^{-1}$ does not vary significantly on increasing the system size beyond 13824 atoms, we calculated the VDOS in this frequency range only for the latter system size to reduce computational costs. It can easily be seen that a shift of the transverse acoustic peak toward lower frequency occurs with increasing degree of disorder. The maximum of this peak is found at about 210 $\mathrm{cm}^{-1}$ for the crystal and at about 160 $\mathrm{cm}^{-1}$ for the most disordered structure. A second effect of the introduction of disorder consists in a decrease of the intensity and a

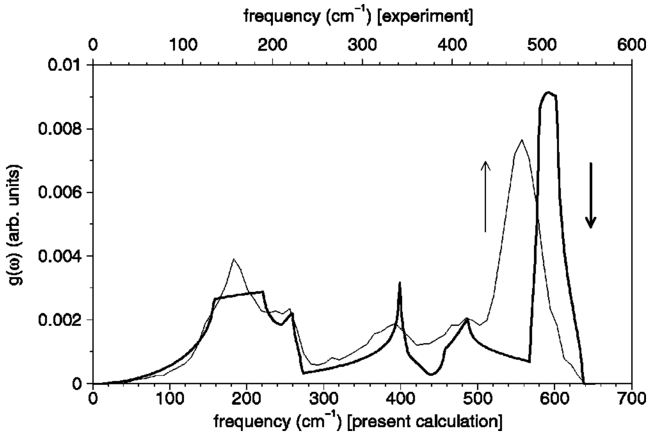

FIG. 1. Vibrational density of states $g(\omega)$ for crystalline silicon. Bold line, present calculation (bottom frequency scale); solid line, polycrystalline silicon from neutron diffraction (upper frequency scale) (from Ref. 28).

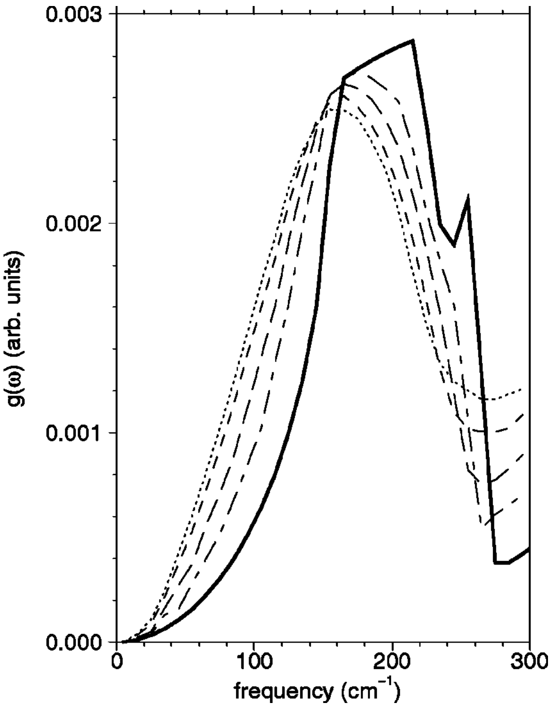

FIG. 2. Low-frequency part of the vibrational density of states $g(\omega)$ of *a*-Si for varying degrees of disorder. Bold, *c*-Si; dashed-dotted, $c_n = 0.1$; long dashed, $c_n = 0.15$; dashed, $c_n = 0.2$; dotted, $c_n = 0.25$.

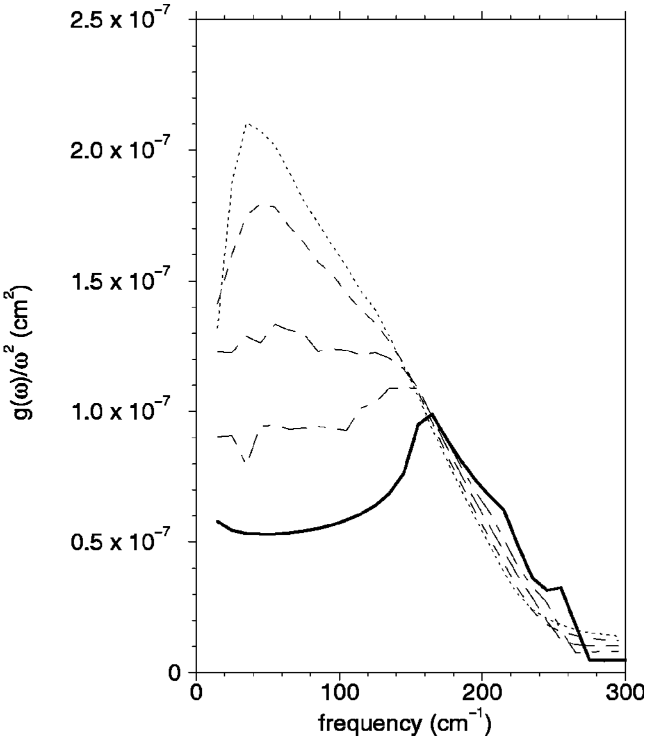

FIG. 3. Low-frequency part of $g(\omega)/\omega^2$ of $a$-Si for varying degrees of disorder. Bold, $c$-Si; dashed-dotted, $c_n=0.1$; long dashed, $c_n=0.15$; dashed, $c_n=0.2$; dotted, $c_n=0.25$.

simultaneous broadening of the transverse acoustic peak. Since the broadening of the transverse acoustic peak is not easily recognized from Fig. 2, we also give the values of the full width of the peak at half maximum $\Delta\omega_{hw}$. $\Delta\omega_{hw}=124\,\mathrm{cm}^{-1}$ for the crystal and $\Delta\omega_{hw}=155\,\mathrm{cm}^{-1}$ for the model with $c_n=0.25$. Summing up, it may be said that an enhancement of the low-frequency VDOS is a direct result of a shift and a broadening of the transverse acoustic peak.

This agrees in part with the result of Schirmacher *et al.*[20,21] They also observe the appearance of extra states in the low-frequency VDOS on introduction of disorder into a harmonic model. As a difference from our findings, the occurrence of extra states in their model seems only to be caused by a broadening and not by a shift of the VDOS. Our calculations support essentially the observations of Schirmacher *et al.*, but additionally it becomes evident that more realistic models of amorphous solids lead to a more complex interpretation of the low-frequency dynamics.

To obtain further information on the deviation of the VDOS of $a$-Si from the Debye behavior $[g(\omega)\sim\omega^2$ for $\omega\rightarrow 0]$ the results are plotted in a different way, i.e., we plot $g(\omega)/\omega^2$ against $\omega$ (Fig. 3). In the crystalline case we note the expected Debye behavior. The deviation from constancy at the lowest frequency for the crystalline curve is a consequence of the finite number of $k$ points $(2\times 10^7)$. We were able to show this by calculating the VDOS of the crystal for a varying number of $k$ points, where a reduction of this deviation with an increasing amount of $k$ points was observed. The fluctuations in the curves for $c_n=0.1$ and $c_n=0.15$ are noise due to the finite sizes and finite number of realizations.

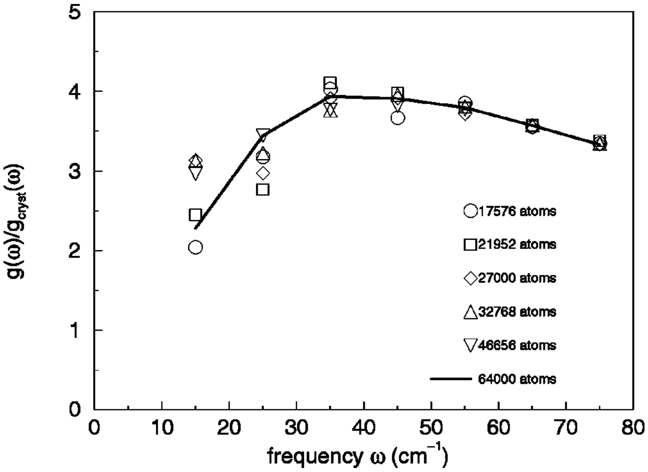

FIG. 4. Relative vibrational density of states $g(\omega)/g_{\mathrm{cryst}}(\omega)$ for models with $c_n=0.25$ at varying system size. $g_{\mathrm{cryst}}(\omega)$ is the vibrational density of states of $c$-Si.

For the least disordered structure with $c_n=0.1$ we observe an enhancement of the low-frequency VDOS over the crystalline value. For this structure the maximum in $g(\omega)/\omega^2$, which is already present in the crystal, increases slightly in intensity and is shifted toward lower frequencies. Nevertheless it can be seen that $g(\omega)\sim\omega^2$ as $\omega\rightarrow 0$ still holds. This is not true anymore for the more disordered models. Already the structure with $c_n=0.15$ shows a weak maximum in $g(\omega)/\omega^2$ at low frequencies. This maximum becomes stronger as the disorder is tuned to a higher value in the structure by choosing $c_n=0.2$. The most disordered structure ($c_n=0.25$) possesses a pronounced peak in $g(\omega)/\omega^2$. Here the VDOS exceeds the crystalline value by a factor of 4. Besides an increase of the intensity of the low frequency peak in $g(\omega)/\omega^2$, it can be seen that this peak is shifted slightly toward lower frequencies with increasing disorder: it is found at 55 cm$^{-1}$ for $c_n=0.15$, at 45 cm$^{-1}$ for $c_n=0.2$, and at 35 cm$^{-1}$ for $c_n=0.25$. We suggest that this peak in $g(\omega)/\omega^2$ should not be named the boson peak nor should the appearance of the boson peak be ascribed to only one of the described observations alone. We believe that the boson peak is generated by the complex of phenomena we observe in the low-frequency VDOS, when disorder is introduced.

To ascertain that the size of our systems is sufficient to describe properly the low-frequency part of the VDOS, we repeated the computation for a series of system sizes between 17576 and 46656 atoms. The results are shown for $c_n=0.25$ in Fig. 4, where the VDOS is shown relative to the crystalline value. It can be seen that the values of the VDOS fluctuate more strongly at low frequency, which is an expected effect of the finite system sizes. At a frequency of 75 cm$^{-1}$ a system size of 17576 atoms gives already a satisfactory description of the low-frequency peak, whereas there is still a strong dependence on system size at a frequency of 15 cm$^{-1}$. Bearing in mind that the cutoff $\omega_{\mathrm{min}}$ for the smallest possible acoustic phonon frequency in the largest of our models was calculated as about 11 cm$^{-1}$, this observation is expected. Nevertheless, the appearance of a peak in $g(\omega)/\omega^2$

at low frequencies is unambiguous even when the absolute values of $g(\omega)/\omega^2$ may have some uncertainty.

## IV. DISCUSSION AND CONCLUSION

In their experimental work Liu and Pohl[30] showed that the low-frequency vibrational properties of $a$-Si are strongly dependent on the sample preparation. They argue that a structure that could be described as a perfect CRN, would not possess an extra VDOS at low frequencies. This statement is supported by an earlier published work of the authors,[37] where a purely fourfold coordinated model of $a$-Si is examined and no extra low-frequency vibrations are observed. On the other hand Liu and Pohl reported the occurrence of extra low-frequency states for samples of $a$-Si with a significant amount of defects, i.e., dangling and floating bonds. In the present work we can confirm this observation, since we have shown that solely by introduction of defects into a purely harmonic model of $a$-Si extra states at low frequencies are generated compared to the crystal. If we tune the disorder of the model to a higher level, the number of defects increases and as a consequence the density of low frequency states is amplified. Because of this wide spread in the vibrational properties, we consider $a$-Si an interesting system to examine the low-energy phenomena of amorphous solids in general.

To our knowledge Schirmacher *et al.*[20,21] observed extra low-frequency vibrations in a purely harmonic model for the first time. Since we calculate all vibrational properties with the help of a harmonic approximation to the SW potential, our results have to be interpreted in the harmonic framework too. We go a step further by inspecting a well accepted model for a real amorphous solid. In addition to the observation of Schirmacher *et al.*, who found an increase of the low-frequency VDOS as a result of its broadening when disorder is introduced, our work shows that the enhancement of the low-frequency VDOS appears as a complex of several phenomena. (i) The transverse-acoustic peak is shifted toward lower frequencies in the amorphous structures compared to the crystal. (ii) A broadening of the same peak is observed as the disorder increases. (iii) In a plot of $g(\omega)/\omega^2$ versus $\omega$, one notes for the least disordered model a constant increase in $g(\omega)/\omega^2$ at low frequencies, whereas in the case of more disordered structures a pronounced peak in $g(\omega)/\omega^2$ evolves.

From neutron-scattering experiments it is known that the boson peak is a consequence of the existence of additional low-frequency vibrations. Thus our results suggest that the boson peak should be observable in sufficiently disordered $a$-Si. It could arise from the complex of phenomena described above. Moreover our investigation suggests that the boson peak is a phenomenon already explainable (at least in part) in the harmonic approximation. Thus it seems that the boson peak is not necessarily connected to anharmonic effects such as the existence of double-well potentials, etc.

In experiments on typical glass formers the boson peak is found in the region of 25–70 $\mathrm{cm}^{-1}$.[3] Since the absolute position of the calculated peak in $g(\omega)/\omega^2$ for our models is at about 40 $\mathrm{cm}^{-1}$, which is in very good agreement with this finding, it is tempting to identify this peak as the boson peak. However, as we already stated above, we believe the boson peak has to be explained as a consequence of several phenomena of the low-frequency VDOS. As mentioned above there is a Raman spectral value[34] for the boson peak in $a$-Si of 114 $\mathrm{cm}^{-1}$. This is somewhat higher than the typical experimental values in other systems. We have to note here that (i) Raman spectra are connected to the VDOS by an unknown phonon-photon coupling factor and (ii) the experiment investigates silicon disordered by irradiation with neutrons, which leads to an amorphous volume fraction of only 4%. The strong dependence of the properties of $a$-Si on the preparation method is well known. Our investigations have also shown that the value of the boson peak decreases in frequency with increasing disorder.

Our findings also support the observation of Nakhmanson and Drabold[22] of an excess specific heat for $a$-Si. Their model is very similar to ours and it is purely harmonic, but as it contains only 4000 atoms it is too small to resolve the shape of the low-frequency VDOS and identify its consequences. In addition to their work on pure $a$-Si the authors focused also on $a$-Si with voids, for which they found an extra peak in the low-temperature dependence of $C/T^3$, which they could trace back to a few localized modes below 11 $\mathrm{cm}^{-1}$. To discuss this problem we refer to our previous paper on the VDOS of $a$-Si and the localization character of the modes. We presented a small model of the same type as used in the present work (relaxation with the SW potential). By calculating the scaling behavior of the correlation length of low-frequency modes, we showed that the modes at the lower edge of the VDOS seem to be localized if one considers only small system sizes, but finally turn out to be delocalized when the system sizes are increased to 8000 atoms. The explanation for this phenomenon is the following. At small system sizes the modes appear to be localized. At larger system sizes long-wavelength phonons appear and couple with the localized modes so that they become extended. Thus these modes should be called resonant modes. Probably the same effect applies to the localized modes in voids in the work of Nakhmanson and Drabold. This points out the problems of finite-size effects.

To summarize; as long as coordination defects are explicitly included in our microscopic model of an amorphous solid ($a$-Si), the introduction of disorder is sufficient not only to increase the density of low-frequency states, but also to generate a peak in $g(\omega)/\omega^2$ in the harmonic approximation. We interpret the simultaneous presence of both phenomena as the boson peak. We want to stress that we do not deny the implications of anharmonic effects and we also do not claim that our model could explain any findings at very low temperatures, which have to be described with the help of the tunneling model.

## ACKNOWLEDGMENTS

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