PHYSICAL REVIEW B VOLUME 55, NUMBER 5 1 FEBRUARY 1997-I

Model investigation of the Raman spectra of amorphous silicon

M. Marinov and N. Zotov
Central Laboratory of Mineralogy and Crystallography, Bulgarian Academy of Sciences, Sofia 1000, Bulgaria

(Received 14 August 1996; revised manuscript received 17 October 1996)

A model for calculating the first-order Raman spectra of amorphous silicon (a-Si) without adjustable parameters is proposed. Calculations on the original 216-atom model of a-Si, generated by the algorithm of Wooten, Winer, and Weaire (WWW) are in very good agreement with experimental spectra and give further indication that the WWW cluster is a realistic model of moderately disordered a-Si. The TA-TO assignment of the low and high frequency bands is supported by direct numerical calculations of the phase quotient and the stretching character of the vibrational modes. The calculated participation ratios and correlation lengths of the vibrational modes indicate that the high-frequency TO-like modes are strongly localized on defects. The relative intensities of the TA-, LA-, and LO-like bands depend on the intermediate-range order, while that of the TO-like band mainly on the short-range order. [S0163-1829(97)10805-0]

I. INTRODUCTION

The investigation of the structure of the archetypal tetrahedral material amorphous silicon (a-Si) has been active area of research over the past 20 years. A large number of continuous random network models of its structure and potentials, describing the interactions in a-Si, have been proposed.[1–10] Although most of these models reproduce with reasonable accuracy the experimentally measured radial distribution function (RDF) of a-Si, they differ considerably in their bond length and bond angle distributions, strain energy, ring statistics, and medium-range order, in general.

On the other hand, it is widely accepted that the vibrational spectra of a-Si are considerably more sensitive than the RDF to changes in the structure and thus can be used as a test measure of the credibility of the structural models.[11]

The experimental Raman spectra of a-Si, measured by different groups,[11–17] have two main strong bands at about 150 and 480 cm^{-1}, which are commonly assigned as TA and TO bands. It has been shown, however, that the TA/TO intensity ratio, their linewidths and frequency positions depend on the method of preparation, deposition conditions, degree of structural disorder, as well as on the hydrogen content in the sample.[11,14,16,17] Furthermore, if the temperature of the sample during the Raman measurement is different from the substrate temperature during deposition, an additional disorder could be introduced due to extrinsic stresses.[18] The TA/TO intensity ratio and the TO linewidth (Γ_{TO}) in the spectra of a-Si increase with increasing degree of structural disorder.[11,14,15] On the contrary, the TA/TO intensity ratio in hydrogenated a-Si samples decreases and the Γ_{TO} increases with increasing hydrogen content.[16] Therefore, in comparing a theoretical Raman spectra to experimental ones, it is desirable to know in detail the method of preparation and residual hydrogen content. It should be noted also that the degree of structural disorder in a-Si is usually characterized only by the rms bond angle deviation $\Delta\theta$. The influence of the defect concentration (under and overcoordinated atoms, highly strained bonds and bond angles) on the calculated, as well as on the experimental, Raman spectra is not studied in detail.

The vibrational density of states (VDOS), Raman, and infrared spectra of a-Si have been calculated by several approaches.[11,15,19–25] Most of the calculations of the Raman spectra[11,15,21,22] are based on the model of induced polarizability, proposed by Alben *et al.*[21] However, the polarizability components in this model should be fitted in order to compare with experiment. Such a fitting procedure introduces additional uncertainties in the interpretation of the spectra. Besides that, the acoustic (optic) character of the vibrational bands of a-Si has been determined only by comparison with the vibrational density of states (VDOS) of crystalline silicon (c-Si).[22,26] The TA band is assigned as bond bending taking into account only that its frequency goes to zero when the bond bending to bond stretching force constants ratio (β/α) goes to zero.[21] The possible mixing[19] of this band with high-frequency bands is studied only on the basis of five- and six-membered rings with Bethe-lattice boundary conditions. The vibrational localization of the high-frequency modes in the Biswas, Grest, and Soukoulis (BGS) model of a-Si is traced to the presence of overcoordinated atoms.[20] Therefore, further studies of the type of the vibrational bands and their localization of a-Si, which is not only important for the interpretation of the vibrational spectra, but also for further understanding of the structure of a-Si and a-Si:H, are needed.

In the present work we study the vibrational modes of a-Si on the basis of the 216-atom model of a-Si, generated by the WWW algorithm,[7] which appears to be a rather realistic representation of the bulk homogeneous structure of a-Si (Ref. 10). In Sec. II we briefly define the phase quotient,[27] stretching character, participation ratio,[28] and correlation length of the vibrational modes, which are used for analysis of the vibrational modes. In Sec. III we present a model for calculating the intensity of the first-order Raman scattering of a-Si without adjustable parameters. The results are compared with experiment in Sec. IV and the conclusions are in Sec. V.

II. CHARACTERIZATION OF THE VIBRATIONAL MODES

One of the most difficult problems in the study of amorphous materials is the definition of appropriate quantities for

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characterization of their vibrational modes. A commonly used approach includes the calculation of the participation ratio and the phase quotient.

The phase relationship between vibrations of neighboring atoms can be assessed by the phase quotient (PQ), originally introduced by Bell and Hibbins-Butler:[27]

$$\mathrm{PQ}(\omega_p)=\frac{\Sigma_m \mathbf{u}_{\mathbf{p}}^{\alpha}\cdot\mathbf{u}_{\mathbf{p}}^{\beta}}{\Sigma_m |\mathbf{u}_{\mathbf{p}}^{\alpha}\cdot\mathbf{u}_{\mathbf{p}}^{\beta}|}, \tag{1}$$

where atoms α and β constitute the mth bond, $\mathbf{u}_{\mathbf{p}}^{\alpha}$ is the displacement vector of atom α when it vibrates in mode p, and the summation is over all nearest-neighbor bonds in the cluster. If the motion of the atoms α and β is roughly parallel for all pairs (acousticlike modes), then $\mathrm{PQ}(\omega_p)$ would have the value of $+1$. If, on the contrary, the motions are roughly antiparallel for each pair (opticlike modes), then PQ (ω_p) will be equal to -1.

The localization of the vibrational modes on the different atoms is determined by the participation ratio (PR) defined by (Ref. 28):

$$\mathrm{PR}(\omega_p)=\frac{(M_1)^2}{M_0 M_2}, \tag{2}$$

where $M_n=\Sigma_\alpha|\mathbf{u}_{\mathbf{p}}^{\alpha}|^{2n}$ for $n=0$, 1, and 2, α numbers the atoms, and the rest of the symbols are as defined in Eq. (1). If the mode p is delocalized and all atoms vibrate with equal amplitudes, then $\mathrm{PR}(\omega_p)$ will be close to $+1$. If, on the contrary, the mode is strongly localized, then $\mathrm{PR}(\omega_p)\to 0$.

The spatial characterization of the vibrational modes, especially of the localized modes, can be further analyzed if we define center of gravity $\mathbf{r}_{\mathbf{p}}$ of mode p

$$\mathbf{r}_{\mathbf{p}}=\frac{\Sigma_\alpha \mathbf{r}^{\alpha}|\mathbf{u}_{\mathbf{p}}^{\alpha}|^2}{\Sigma_\alpha|\mathbf{u}_{\mathbf{p}}^{\alpha}|^2} \tag{3}$$

and the corresponding root-mean-square deviation (rms) $L_c(\omega_p)$:

$$L_c^2(\omega_p)=\frac{\Sigma_\alpha|\mathbf{r}^{\alpha}-\mathbf{r}_{\mathbf{p}}|^2|\mathbf{u}_{\mathbf{p}}^{\alpha}|^2}{\Sigma_\alpha|\mathbf{u}_{\mathbf{p}}^{\alpha}|^2}, \tag{4}$$

where $\mathbf{r}^{\alpha}$ is the radius vector of atom α in the model, $L_c(\omega_p)$ represents in fact a correlation length beyond which the amplitude of the atomic vibrations decreases significantly. It should be noted that in structural models with periodic boundary conditions the maximal value of L_c will be about $a/2$, where a is the box edge.

In addition to the participation ratio, phase quotient, and the correlation length we introduce a parameter referred to as stretching character (S),

$$S(\omega_p)=\frac{\Sigma_m|(\mathbf{u}_{\mathbf{p}}^{\alpha}-\mathbf{u}_{\mathbf{p}}^{\beta})\cdot\hat{\mathbf{r}}^{\mathbf{m}}|}{\Sigma_m|\mathbf{u}_{\mathbf{p}}^{\alpha}-\mathbf{u}_{\mathbf{p}}^{\beta}|}, \tag{5}$$

where $\hat{\mathbf{r}}^{\mathbf{m}}$ is a unit vector parallel to the mth bond and the rest of the symbols are as defined in Eq. (1). The value of $S(\omega_p)$ will be close to $+1$ if mode p is predominantly of bond stretching type, and close to 0 if mode p is predominantly bond bending.

In structural models with periodic boundary conditions all parameters defined above must be calculated using the nearest image convention in order to determine the atomic separations.

III. CALCULATION OF RAMAN SPECTRA

The model most frequently used for calculating the Raman spectra of a-Si is that proposed by Alben *et al.*[21] A cylindrical symmetry of the individual bonds is assumed and each bond is treated independently as a homopolar diatomic molecule. Three different forms for the bond polarizability[15,21] are introduced: α_1, α_2, and α_3. In practical calculations some authors have used either combinations of these three forms (Refs. 11, 21, and 22) or only one of them.[15] An extensive discussion of α_1, α_2, and α_3 can be found in Ref. 11, where it has been shown that α_1, α_2, and α_3 all contribute to the polarized (HH) Raman spectrum, α_1 and α_2 contribute to the fully depolarized (HV) Raman spectrum, while α_3 contributes only to the fully polarized [HH-(4/3)HV] Raman spectrum. An additional assumption is that these three mechanisms of light scattering contribute incoherently to the spectra. These difficulties in the practical application of the model of Alben *et al.*[21] indicate that it is necessary to adopt such an approach for the calculation of the Raman spectra of a-Si, which naturally combines all three mechanisms of light scattering.

For this purpose, we express the bond polarizability as a sum of three components:[29] component, A_b^m, parallel to the bond, arising from the bonding electrons; component, A_n^m, parallel to the bond, which is due to the nonbonding electrons, and a transverse component A_{tr}^m. These quantities are defined as follows:

$$A_b^m=\sigma^m\frac{(r^m)^4}{\sqrt[6]{4^4\eta_\alpha\eta_\beta}};\qquad \sigma^m=\exp\left[-\frac{(X_\alpha-X_\beta)^2}{4}\right];$$

$$A_n^m=f_\alpha\eta_\alpha+f_\beta\eta_\beta;\qquad A_{\mathrm{tr}}^m=n_{\mathrm{df}}^m\frac{(X_\alpha^2\eta_\alpha+X_\beta^2\eta_\beta)}{(X_\alpha^2+X_\beta^2)}, \tag{6}$$

where η_α and η_β are the atomic polarizabilities of the atoms forming the mth bond, σ^m is the fractional covalent character of the mth bond, X_α and X_β are the electronegativities of the atoms α and β, f_α and f_β are the fractions of the valence electrons of the atoms α and β out of the bond region, and $n_{\mathbf{df}}^m$ is the number of degrees of freedom of the mth bond. This form of the bond polarizability has been already applied to the study of the Raman spectra of silicate glasses.[30–33]

The coordination of all Si atoms in the 216-atom WWW cluster with periodic boundary condition is 4. We assume also that all Si atoms are of the same electronegativity and atomic polarizability. In this case $\sigma^m=1$, $n_{\mathrm{df}}^m=0$, f_α and f_β are also zero, so that $A_{\mathrm{tr}}^m=0$ and $A_n^m=0$. As a result, the bond polarizability depends only on the polarizability of Si atoms, η, and on the bond lengths. In this particular case, the polarizability tensor of the pth vibrational mode[32] takes the form

$$\rho(\omega_p)=\frac{1}{V}\sum_m \frac{(r_m)^3}{2^3\sqrt{16\eta}}\{(\Delta\mathbf{b}_\mathbf{p}^\mathbf{m}\cdot\mathbf{r}^\mathbf{m})\hat{\mathbf{r}}^\mathbf{m}\hat{\mathbf{r}}^\mathbf{m} + 1/2(\Delta\mathbf{b}_\mathbf{p}^\mathbf{m}\hat{\mathbf{r}}^\mathbf{m}+\hat{\mathbf{r}}^\mathbf{m}\Delta\mathbf{b}_\mathbf{p}^\mathbf{m})\}, \tag{7}$$

where $\Delta\mathbf{b}_\mathbf{p}^\mathbf{m}=\mathbf{u}_\mathbf{p}^\alpha-\mathbf{u}_\mathbf{p}^\beta$ and r^m is the length of the mth bond. This form of the polarizability tensor is used in the present paper. In the calculations we have used for all Si atoms in the cluster the value[29] $\eta=2.988$ Å^3.

IV. RESULTS AND DISCUSSION

The original 216-atom model of a-Si (Ref. 7) with periodic boundary conditions (box edge $a=16.28$ Å) is generated with a Keating potential with ratio of the bond bending to the bond stretching force constant, $\beta/\alpha=0.285$. The rms bond-angle deviation from the ideal tetrahedral value is 10.8°, while the rms bond-length deviation from the Si-Si bond in c-Si is 2.7%.

We obtain the atomic vibrations of the model by direct diagonalization of the dynamical matrix with a Kirkwood-type potential:[34]

$$V=\frac{\alpha}{4}\sum_{l,i}[(\mathbf{u}^\mathbf{l}-\mathbf{u}^\mathbf{i})\cdot\hat{\mathbf{r}}_\mathbf{li}]^2 + \frac{\beta}{4}\sum_{l\{i,j\}}[(\mathbf{u}^\mathbf{l}-\mathbf{u}^\mathbf{i})\cdot\mathbf{h}_\mathbf{li}+(\mathbf{u}^\mathbf{l}-\mathbf{u}^\mathbf{j})\cdot\hat{\mathbf{h}}_\mathbf{ij}]^2, \tag{8}$$

where the first sum is over all atoms l and their four nearest neighbors i, the second sum is over all atoms l and pairs of distinct neighbors $\{i,j\}$, $\mathbf{u}^\mathbf{l}$ is the displacement of atom l from equilibrium, $\hat{\mathbf{r}}_\mathbf{li}$ is a unit vector parallel to the bond $l-i$, while $\hat{\mathbf{h}}_\mathbf{li}$ is a unit vector perpendicular to this bond.

We use this type of potential because the bond compression and the bond-angle distortion terms in this case are fully separated into bond stretching and bond bending contributions, while in the original Keating potential[35] such separation occurs only in the case of bond angle 90°. As a consequence, there is a significant bond stretching contribution in the bond bending force constant of the Keating potential for bond angles different from 90°. That is why in our case the β/α ratio is expected to be smaller than the above mentioned value of 0.285.

In order to perform realistic calculations of the Raman spectra of a-Si, using the Kirkwood-type potential, Eq. (8), the β/α ratio must be redefined. It has been chosen taking into account that (i) the VDOS, obtained from neutron scattering experiments,[36] indicates that the overlap between the TA-like band and the other vibrational bands of a-Si is small, which is consistent with the choice $\beta/\alpha\geqslant0.02$ (see below); (ii) the frequency position of the Raman active opticlike band of the model must be close to that of the corresponding band in the experimental spectrum; (iii) the force constants should be in accordance with the phonon dispersion relations[37] for the diamond-cubic structure of c-Si. The conditions listed above can be satisfied for $\alpha=125$ N/m and $\beta=5$ N/m ($\beta/\alpha=0.04$) and all the subsequent calculations are performed with these values. Since the rms deviation of the Si-Si bond length in the model is only 2.7%, we have neglected the dependence of the stretching force constant α on r in Eq. (8).

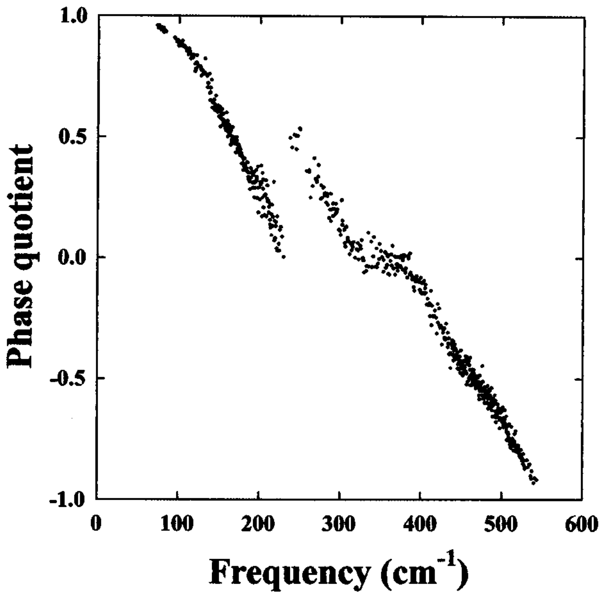

FIG. 1. Calculated phase quotient for the 216-atom model.

The phase quotient as a function of the vibrational frequency, calculated according Eq. (1) is shown in Fig. 1. It can be seen that the vibrations with frequency $\omega<180$ cm^{-1} are of predominantly acoustic character while the high frequency bands with $\omega>450$ cm^{-1} are of predominantly optical character. This is a first direct proof of the usual assignment of the acoustic-optic character of the vibrational bands of a-Si at about 150 and 480 cm^{-1} (Ref. 26). Besides that, the lowest frequency band approaches zero frequency when the bond bending force constant is small. This verifies the usual assignment of this band as originating from the TA band of c-Si (Ref. 21). The width of the TA-like band increases with increasing the β/α ratio due to the stronger interaction between stretching and bending modes. We have also studied the phase quotient for different values of the β/α ratio. Independently of how small this ratio is, the major peculiarities of Fig. 1 remain unchanged. However, for $\beta/\alpha>0.07$ the change of the character of the bands from acoustic at low frequencies to optic at high frequencies becomes continuous which indicates that in this case there is a mixing between the bands.

We have not performed special calculations to determine which modes are transverse or longitudinal in character and accept in the following discussion the commonly used assignment[22] that the bands at about 150 and 480 cm^{-1} are transverse in nature, while the bands in the range 250–450 cm^{-1} are longitudinal.

The modes below 225 cm^{-1} are bond bending in nature, while those above 225 cm^{-1} are predominantly of bond stretching character (Fig. 2). The later result confirms the usual interpretation that the TO-like band is of bond stretching character, a conclusion made on the basis of separate calculations[11] of the α_1, α_2, and α_3 contributions to the bond polarizability in the model of Alben *et al.*[21] As in the case of the phase quotient, Fig. 1, there is a jump in the stretching character (see Fig. 2) at about 225 cm^{-1}. Evidently there is

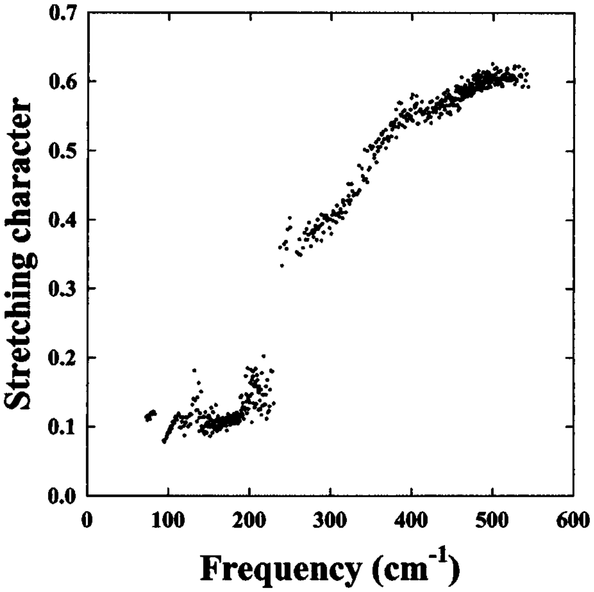

FIG. 2. Calculated stretching character of the vibrational modes for the 216-atom model.

no significant mixing between the TA-like band and the other bands for realistic values of β/α. This result is opposite to the interpretation of Nichols *et al.*[19] that the maxima in the central region in the vibrational spectra of a-Si are produced by splitting off of the bond bending and bond stretching modes when either one starts to mix with the others. From Figs. 1 and 2 it is evident that the vibrational modes of a-Si can be clearly subdivided into two bands: one below 225 cm^{-1} acoustic in nature and with bond bending character and one above 225 cm^{-1} bond stretching in character with acoustic nature for low frequencies and optic for high ones.

The participation ratio is given in Fig. 3. The modes below 400 cm^{-1} are fully delocalized and the atoms vibrate with almost equal amplitude like one giant molecule. The degree of localization above 400 cm^{-1} starts to decrease and for $\omega \geqslant 520$ cm^{-1} the modes become strongly localized. This result is in accordance with the calculations of Biswas *et al.*[20] performed with slightly different definition of the participation ratio. The localization of high-frequency modes is even more clearly seen from the frequency dependence of the correlation length, Fig. 4, which abruptly decreases from $L_c = 8.14$ Å for the delocalized modes to 3–5 Å for the strongly localized modes. As one can see, there are seven modes with $L_c \leqslant 5$ Å (Fig. 4). The calculated centers of gravity indicate that these modes are localized on seven structural defects well separated apart. Detailed geometrical analysis of the defects indicates that they represent single (type I) or double (type II) tetrahedra with angular distortions $\delta\theta$ larger that the average rms deviation of the model $\Delta\theta = 10.8°$. The correlation length for the defects of type I increases, while L_c for defects of type II decreases with increasing $\delta\theta$ (Fig. 5). In both cases the distorted tetrahedra contain long (longer than 2.4 Å) Si-Si bonds as well. As it was indicated above the stretching force constant in our calculations is not a function of the bond length and all atoms are fourfold coordinated (as an upper limit of the nearest-neighbor distance we consider the value of 2.63 Å). Nevertheless, high frequency localization is observed, which indicates that this phenomenon is not related only to coordination defects (five-coordinated atoms),[20] but, similarly to electron localization,[38] to large local deviations of the tetrahedral angles.

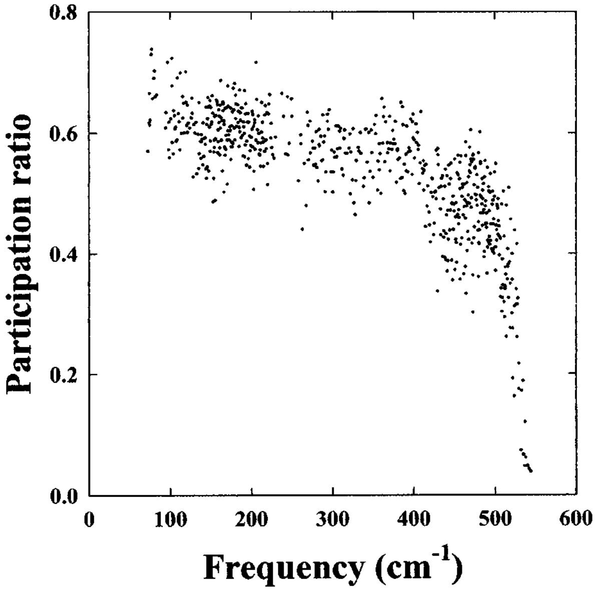

FIG. 3. Calculated participation ratio for the 216-atom model.

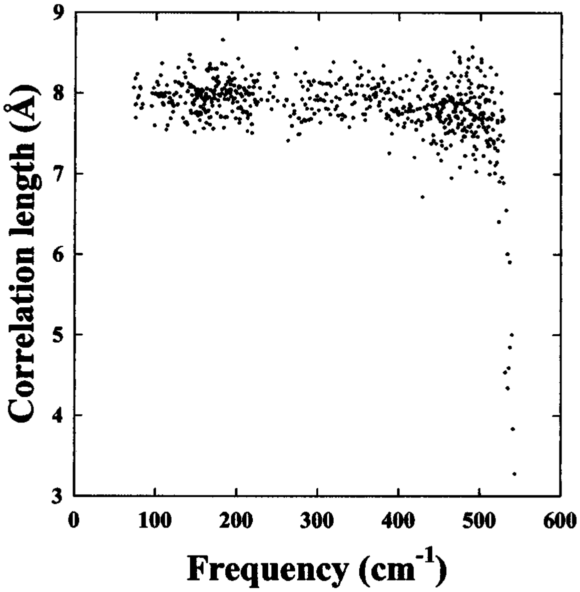

FIG. 4. Calculated correlation length of the vibrational modes for the 216-atom model.

The calculated VDOS, Fig. 6, is in good agreement with that obtained by Winer[22] and Biswas *et al.*[20] with Keating potential. This indicates that the VDOS of a-Si is not very much sensitive to the type of the short-range potential used. All spectral lines in the calculated VDOS are additionally broadened by 17 cm^{-1} to account for the finite size of the WWW model. The calculated VDOS (Fig. 6) is very similar to the experimentally measured one by inelastic neutron scattering.[36]

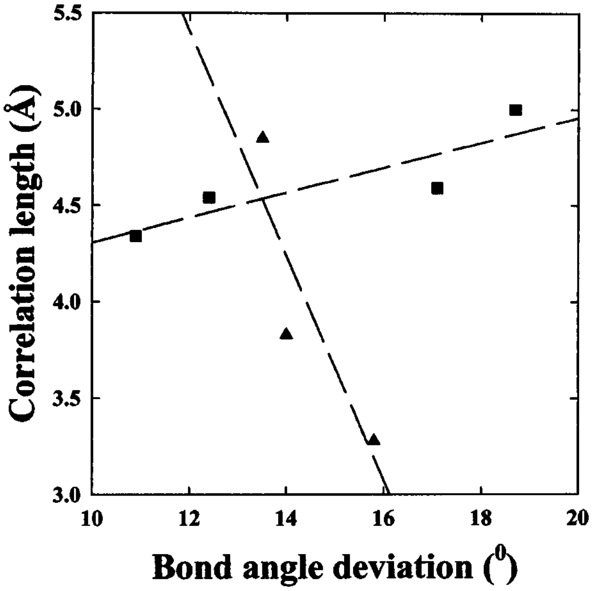

FIG. 5. Correlation length vs the bond-angle deviation $\delta\theta$ of defects of type I (■) and of type II (▲). The dashed lines are least-squares fit through the data points.

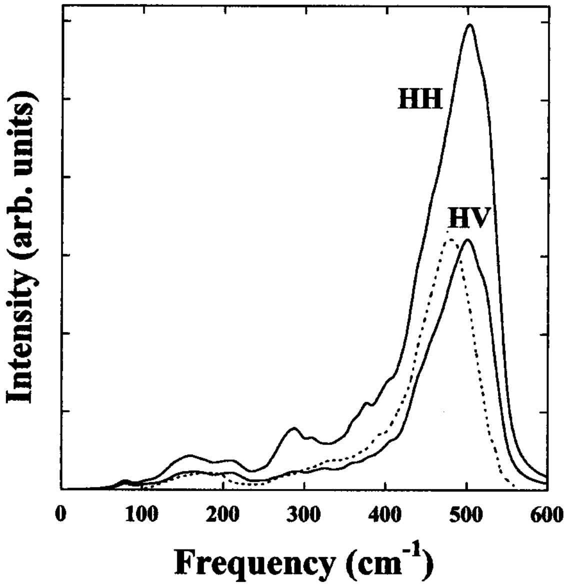

FIG. 7. Calculated HH and HV Raman spectra for the 216-atom model (solid lines) and the experimental HV Raman spectrum of the sample *a* (Ref. 11) (dotted line).

The calculated HH and HV Raman spectra for the 216-atom model are given in Fig. 7 (solid lines). The full width at the half maximum of the TO-like band in our HH Raman spectrum is 90 ± 10 cm^{-1}. It should be noted that for calculated Raman spectra Γ_{TO} depends also on the additional broadening (17 cm^{-1}) introduced to account for the finite size of the model. Taking into account that $\Delta\theta=10.8°$ one can see that the formula proposed by Beeman *et al.*[15] ($\Gamma_{\mathrm{TO}}=15+6\Delta\theta$), using several fully relaxed models with free boundaries, describes well the relation between Γ_{TO} and $\Delta\theta$ for the WWW model as well.

As has been pointed out in Sec. I, the experimental vibrational spectra of *a*-Si depend strongly on the method of preparation. In particular, Maley *et al.*[11] have reported that Γ_{TO} (measured at room temperature) of *a*-Si thin films, obtained by chemical-vapor deposition (CVD) and radio frequency (rf) diod sputtering vary from 78 to 120 cm^{-1} in dependence of the substrate temperature. Therefore our calculated spectra should be compared with experimental spectra of *a*-Si without hydrogen and with $\Gamma_{\mathrm{TO}}\cong90$ cm^{-1}. As one can see the calculated HV Raman spectrum reproduces very well all features of the experimental spectra of samples *a* and *b* of Maley *et al.*[11] ($\Gamma_{\mathrm{TO}}=78$ and 94 cm^{-1}, respectively). For comparison, the experimental HV Raman spectrum of the sample *a* is shown in Fig. 7 (dotted line). The TA/TO intensity ratio for the calculated spectrum is about 0.07, which is slightly less than that for sample *b* (TA/TO $\cong0.12$) and is close to that for sample *a* (TA/TO $\cong0.07$).[11] The difference between the frequency position of the TO-like band in the calculated and experimental Raman spectra of about 20 cm^{-1} can be eliminated by additional adjustment of the bond stretching force constant used in the model calculations. The average value of the calculated depolarization ratio is 0.5 and the depolarization ratio of only the TO-like band is 0.55, exactly as in the experimentally measured spectra.[11] The calculated depolarization ratios of the TA-, LA-, and LO-like bands are less than that for the TO-like band in qualitative agreement with the experimental spectra.[13]

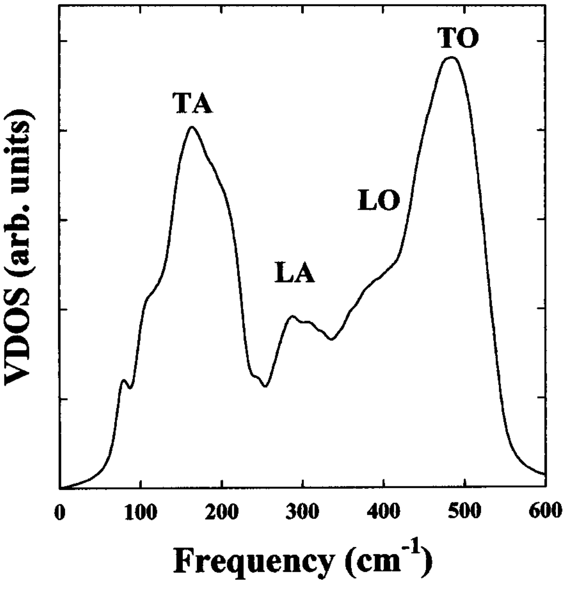

FIG. 6. Calculated VDOS for the 216-atom model.

To study the effects of intermediate-range order (IRO) we have calculated also the partial Raman spectra[39] of small clusters of equal size, but extracted from three different regions of the 216-atom model (and thus with different IRO and defect concentration). The comparison of these spectra, Fig. 8, indicates that the Raman intensities of the lowest-frequency acoustic modes in the TA-like band ($\omega<150$ cm^{-1}) and highest-frequency optic modes in the TO-like band ($\omega>490$ cm^{-1}) are independent on the IRO. In other words, they are sensitive predominantly to the short-range order. On the other hand, the modes with $-0.5<\mathrm{PQ}(\omega)<0.5$ (see Fig. 1) which are mainly in the

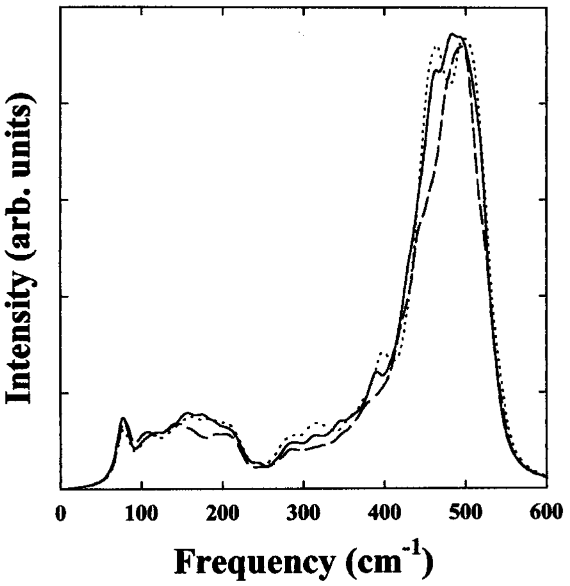

FIG. 8. Calculated partial HV Raman spectra for clusters of equal size extracted from different regions of the 216-atom model.

LA-LO region (150–450 cm^{-1}) depend on the IRO. Additional studies of the effects of IRO and defect concentration in structural models with similar rms bond-angle deviation are needed.

V. CONCLUSIONS

The character of the vibrational modes of a-Si on the basis of the 216-atom model of Wooten, Winer, and Weaire[7] was studied. The usual TA-TO assignment of the low and high frequency vibrational bands of a-Si, made by comparison with the VDOS of c-Si, is supported by direct numerical calculations of the phase quotient. The calculated participation ratios and correlation lengths indicate that the TA-, LA-, and LO-like modes are delocalized over all atoms, while the high frequency opticlike modes are strongly localized on defects (single or two interconnected tetrahedra with large angular distortions). The TA-like band, opposite to the other bands, is predominantly of bending nature. The jump in the stretching character and in the phase quotient at about 200 cm^{-1} indicates that the interaction of the TA-like band with the other vibrational bands of a-Si is small.

A model for calculating the Raman spectra of a-Si based on the bond polarizability approximation[28] without adjustable parameters is proposed. The calculated HH and HV spectra, as well as the depolarization ratio, are in very good agreement with the experimentally published spectra.[11] This is a confirmation that the 216-atom model of Wooten, Winer, and Weaire[7] is a realistic model of the structure of moderately disordered a-Si ($\Gamma_{\mathrm{TO}} \leqslant 90$ cm^{-1}). The high frequency modes in the TA-like band, as well as the LA- and LO-like bands are sensitive mainly to the intermediate-range order, while the TO-like band mainly to the short-range order. The results depend, however, to some extent, on the size, defect concentration, and the medium-range order of the structural model used. In this respect it would be interesting to extend the present calculations to larger models of the structure of a-Si generated, for example, by reverse Monte Carlo simulations.[8]

ACKNOWLEDGMENTS

We thank Professor F. Wooten for the coordinates of the WWW cluster. The financial support provided by the Spanish Interministerial Commission for Science and Technology for N. Zotov is highly appreciated. We thank also Professor M. F. Thorpe for the valuable remarks on the initial version of the manuscript.

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