PRL **100,** 137402 (2008) PHYSICAL REVIEW LETTERS week ending 4 APRIL 2008

Raman Scattering and the Low-Frequency Vibrational Spectrum of Glasses

Bernhard Schmid* and Walter Schirmacher
Physik-Department E13, Technische Universität München, D-85747, Garching, Germany

(Received 31 August 2007; published 2 April 2008)

We present a theory of low-frequency Raman scattering in glasses, based on the concept that light couples to the elastic strains via spatially fluctuating elasto-optic (Pockels) constants. We show that the Raman intensity is not proportional to the vibrational density of states (as was widely believed), but to a convolution of Pockels constant correlation functions with the dynamic strain susceptibilities of the glass. Using the dynamic susceptibilities of a system with fluctuating elastic constants we are able for the first time to describe the Raman intensity and the anomalous vibration spectrum of a glass on the same footing. Good agreement between the theory and experiment for the Raman spectrum, the density of states, and the specific heat is demonstrated at the example of glassy As_2S_3.

DOI: 10.1103/PhysRevLett.100.137402 PACS numbers: 78.30.Ly, 65.60.+a, 78.20.−e

Raman scattering is an indispensable tool for investigating the structure and dynamics of solids [1,2]. In crystalline materials Raman scattering mainly probes optical phonons, from which structural information can be extracted. In disordered solids a continuous band of excitations in the range of 10 to 100 wave numbers is observed, which is absent in single crystals, in which sharp lines of specific Raman-active optical modes are observed. The appearance of the continuous band in disordered solids, especially glasses, is obviously due to the breakdown of the crystal symmetries, which, for example, does not allow for a distinction between acoustic and optical modes in the low-frequency domain [3]. This spectroscopic peculiarity of disordered solids has been the object of a large number of investigations in the last three decades [4–9]. It is related to similar features in far-infrared absorption [10,11] and has proven recently to lead to important applications for optical amplifiers [12].

In spite of the large number of low-frequency Raman data the theoretical description of the spectra, especially their relationship to the experimentally measurable vibrational density of states (DOS) $g(\omega)$, remains an open question until today. According to an early treatment of this problem the Raman intensity $I(\omega)$ is related to $g(\omega)$ via [13]

$$I(\omega) = C[n(\omega) + 1]\frac{g(\omega)}{\omega} \tag{1}$$

where $n(\omega) = [\exp\{\hbar\omega/k_BT\} - 1]^{-1}$ is the mean boson occupation number. Equation (1) was derived assuming that the light couples to the displacements of spatially uncorrelated harmonic vibrations and a breakdown of the selection rules due to the disorder. For incoherent inelastic neutron scattering a relation for the intensity similar to (1) holds. However, the observed $g(\omega)$ differ in shape from the Raman spectra, divided by $[n(\omega) + 1]/\omega$, in all known cases. In order to be able to use an expression similar to (1) one introduced a phenomenological frequency dependent coupling coefficient $C \rightarrow C(\omega)$, which is essentially the Raman susceptibility $I(\omega)/[n(\omega) + 1]$, divided by the level density $g(\omega^2) = g(\omega)/2\omega$ [5]. Considerable effort has been invested in determining $C(\omega)$ from simulations [5] and from comparing Raman with inelastic scattering or specific heat data [6].

However, except for the numerical simulations for fractal networks [5] there exists no theory in the literature, which allows for calculating both the Raman intensity and the density of states leading to a realistic frequency dependence of $C(\omega)$. Two theories for $I(\omega)$, which appeared after Ref. [13], do not satisfactorily address this issue. The first one, due to Martin and Brenig (MB) [14], does not give a relation between $I(\omega)$ and $g(\omega)$ as it is based on a model with fluctuating elasto-optic constants combined with Debye-like acoustic waves. Moreover the theory does not agree with experiments [8]. The second [15] is based on the soft-potential model [16]. It is assumed that the amplitudes of quasilocal modes couple to the light. This leads to a formula like (1). However, this theory predicts a frequency-independent depolarization ratio, which is in disagreement with experiment [9].

A relationship between the Raman intensity and the vibrational DOS is highly desired because near the peak of the Raman intensity an anomalous enhancement of the DOS over Debye's ω^2 law is observed, which shows up as a peak in the reduced DOS $g(\omega)/\omega^2$. This feature has attracted enormous attention in the literature [17]. Both peaks have been called "boson peak" (BP) but their shape and position differ. The temperature dependent specific heat $C(T)/T^3$ exhibits a peak around ~10 K in many glassy materials [21], which can be traced as well to a BP in $g(\omega)/\omega^2$ and is therefore also called boson peak. In this temperature regime the thermal conductivity has a characteristic plateau or dip [22], which can be shown to be related to the BP of $g(\omega)$ [23].

Explanations of the boson-peak anomaly [17] include theories based on the soft-potential model [16] and on the model of fluctuating elastic constants [23–26]. All these studies revealed that the BP marks the lower frequency boundary of a band of irregular states produced by the frozen-in disorder, which hybridize with the Debye phonons. These states can either be quasilocal defect modes [16,25] or random-matrix type states [23–26].

In the present Letter we develop a new theory of Raman scattering in disordered solids. It is based on the assumption that due to the frozen-in disorder the elasto-optic (Pockels) constants fluctuate in space. The result of our theory is that the Raman intensity can be represented as a weighted sum over the imaginary parts of the longitudinal and transverse dynamic susceptibilities. The weight functions are the appropriate spatial correlation functions of the Pockels constants. This establishes a relationship between the anomalous vibrational spectrum of the solid and the Raman intensity. By this we present for the first time a theoretically founded tool for comparing Raman with inelastic neutron and x-ray spectra. We demonstrate this for the example of glassy As_2S_3 using a fluctuating-elastic constant model [23,26] for the boson-peak anomaly.

We start with the usual expression for the Raman intensity [1,2]

$$I_{ij}(\mathbf{q},\omega) \propto \int d^3\mathbf{r} \int dt e^{i[\mathbf{q}\mathbf{r}-\omega t]} \langle \Delta\epsilon_{ij}(\mathbf{r}+\mathbf{r}_0, t+t_0) \times \Delta\epsilon_{ij}(\mathbf{r}_0, t_0)\rangle, \qquad (2)$$

where i and j denote the Cartesian directions ($x \equiv H$, $y \equiv V$) of the polarization vectors of the incoming and scattered light, respectively, and $\Delta\epsilon_{ij}(\mathbf{r}, t)$ denotes the local fluctuations of the dielectric tensor. The latter couples to the vibrational degrees of freedom via the strain tensor $u_{ij} = (1/2)[\partial_i u_j + \partial_j u_i]$ as $\Delta\epsilon_{ij}(\mathbf{r}, t) = a_1(\mathbf{r})\sum_\ell u_{\ell\ell}(\mathbf{r}, t)\delta_{ij} + a_2(\mathbf{r})v_{ij}(\mathbf{r}, t)$ with $v_{ij} = u_{ij} - (1/3)\delta_{ij}\sum_\ell u_{\ell\ell}$ [27]. $a_{1,2}(\mathbf{r})$ are the local elasto-optic (Pockels) constants. These quantities are now assumed [14] to have disorder-induced fluctuations $a_{1,2}(\mathbf{r}) = a_{1,2}^{(0)} + \Delta a_{1,2}(\mathbf{r})$ with correlation functions $C_{1,2}(\mathbf{r}) = \langle \Delta a_{1,2}(\mathbf{r}_0 + \mathbf{r})\Delta a_{1,2}(\mathbf{r}_0)\rangle$. The constant terms $a_{1,2}^{(0)}$ produce the usual formulae for Brillouin scattering and Raman scattering from high-frequency optical modes [3]. From the fluctuating terms we obtain a new disorder-induced low-frequency contribution, in which we set $I_{ij}(\mathbf{q}, \omega) \approx I_{ij}(\mathbf{q} = 0, \omega) \equiv I_{ij}(\omega)$, and obtain [28]

$$\frac{I_{VH}(\omega)}{n(\omega)+1} = \alpha \int \left(\frac{d\mathbf{k}}{2\pi}\right)^3 \frac{1}{30} C_2(\mathbf{k})[2\chi_L''(\mathbf{k},\omega) + 3\chi_T''(\mathbf{k},\omega)], \qquad (3)$$

$$\frac{I_{VV}(\omega)}{n(\omega)+1} = \alpha \int \left(\frac{d\mathbf{k}}{2\pi}\right)^3 \Big(C_1(\mathbf{k})\chi_L''(\mathbf{k},\omega) + \frac{2}{45}C_2(\mathbf{k}) \times [2\chi_L''(\mathbf{k},\omega) + 3\chi_T''(\mathbf{k},\omega)]\Big), \qquad (4)$$

where α is a proportionality constant which involves the incident intensity, divided by the 4th power of the wavelength of the scattered light [1,28]. Here $\chi_{L,T}''$ are the imaginary parts of the longitudinal and transverse dynamic strain susceptibilities. $[n(\omega) + 1]\chi_L''(\mathbf{k}, \omega)$ is proportional to the coherent inelastic neutron and x-ray scattering law $S(\mathbf{k}, \omega)$ [26]. We now assume for simplicity that the k dependence of the correlation functions $C_{1,2}(k)$ is the same, i.e., $C_{1,2}(k) = f_{1,2}\tilde{C}(k)$ with $f_{1,2} = C_{1,2}(\mathbf{r} = 0) \propto \langle \Delta a_{1,2}^2\rangle$ and $(1/8\pi^3)\int d^3\mathbf{k}\tilde{C}(k) = \tilde{C}(\mathbf{r} = 0) = 1$, and we define $\tilde{\chi}_{L,T}(\omega) = (1/8\pi^3)\int d^3\mathbf{k}\tilde{C}(k)\chi_{L,T}(\mathbf{k}, \omega)$. We obtain

$$I_{VH}(\omega) = \frac{1}{30}\alpha f_2[n(\omega) + 1](2\tilde{\chi}_L''(\omega) + 3\tilde{\chi}_T''(\omega)) \qquad (5)$$

$$I_{VV}(\omega) = \alpha f_1[n(\omega) + 1]\tilde{\chi}_L''(\omega) + \tfrac{4}{3}I_{VH}(\omega), \qquad (6)$$

from which we get the depolarization ratio

$$\rho(\omega) = \frac{I_{VH}(\omega)}{I_{VV}(\omega)} = \left[\frac{4}{3} + 15\frac{f_1}{f_2}\frac{1}{1 + \frac{3}{2}\tilde{\chi}_T''(\omega)/\tilde{\chi}_L''(\omega)}\right]^{-1}. \qquad (7)$$

The value and the spectral shape of $\rho(\omega)$ is controlled by the ratio of the mean-square fluctuations of the longitudinal and transverse Pockels constants and the ratio of the two susceptibilities.

If one would take the dynamic susceptibilities of Debye-type plane waves $\chi_{L,T,0}''(k, \omega) \propto \frac{k}{v_{L,T}^2}\delta(k - \frac{\omega}{v_{L,T}})$ ($v_{L,T}$ = longitudinal/transverse sound velocity) and assume a Gaussian correlation function

$$\tilde{C}(k) = [4\pi\xi^2]^{3/2}\exp\{-k^2\xi^2\} \qquad (8)$$

one would obtain $\tilde{\chi}_{L,T,0}''(\omega) \propto (\xi^3\omega^3/v_{L,T}^5)e^{-\omega^2\xi^2/v_{L,T}^2}$. Inserting this into expressions (5)–(7), leads just to the results of MB [14] for $I_{ij}(\omega)$ and $\rho(\omega)$. We conclude that the MB theory describes the Raman spectrum of a *Debye solid* with spatially fluctuating Pockels constants. Obviously the disorder-induced distortions of the plane waves introduced by MB only enter into the prefactor of the spectrum but do not affect its frequency dependence.

In order to describe the vibrational excitations of a disordered (non-Debye) solid we apply the model developed in [23,26]. The model is based on the assumption that as a result of the disorder the shear modulus G of the material has a spatial variation $G = G_0 + \Delta G(\mathbf{r})$, where the fluctuations $\Delta G(\mathbf{r})$ have a correlation function $C_G(\mathbf{r}) = \langle \Delta G(\mathbf{r}_0 + \mathbf{r})\Delta G(\mathbf{r}_0)\rangle$. This model can be solved in self-consistent Born approximation (SCBA), which appears as a saddle point of a matrix-valued effective field theory (nonlinear σ model [29,30]). The short-range correlated version of this model [i.e., $C_G(\mathbf{k}) = \text{const} \propto \langle \Delta G^2\rangle$ for $k < k_D$, where k_D is the Debye wave number] has already been applied successfully for describing the anomalous vibrational properties of glasses [23,25,26]. Quite recently the inclusion of longer-range correlation has been shown [31] to lead to an interesting scaling behavior of the Brillouin linewidth and a reinforcement of the boson-peak anomaly.

In SCBA the disorder enters via a complex self-energy function $\Sigma(\omega)$ [32], which obeys the self-consistent equation

$$\Sigma(\omega) = \gamma \int_{|\mathbf{k}|<k_D} \frac{d^3\mathbf{k}}{(2\pi)^3}\tilde{C}_G(k)[\chi_L(\mathbf{k},\omega) + \chi_T(\mathbf{k},\omega)] \qquad (9)$$

where we have put $C_G(k)=f_G\tilde{C}_G(k)$ with $\frac{1}{8\pi^3}\times\int_{|\mathbf{k}|<k_D}d^3\mathbf{k}\tilde{C}_G(k)=1$ and $\gamma\propto f_G\propto\langle\Delta G^2\rangle$.

For the dynamic susceptibilities we have

$$\chi_{L,T}(\mathbf{k},\omega)=k^2G_{L,T}(\mathbf{k},\omega)=\frac{k^2}{-\omega^2+k^2[v^2_{L,T,0}-\Sigma_{L,T}(\omega)]}. \tag{10}$$

Here $G_{L,T}(\mathbf{k},\omega)=G'_{L,T}(\mathbf{k},\omega)+iG''_{L,T}(\mathbf{k},\omega)$ are the longitudinal and transverse Green's functions [33], $v_{L,T,0}$ are the unrenormalized sound velocities (the ones in the absence of disorder), and $\Sigma_T(\omega)=\Sigma(\omega)$, $\Sigma_L(\omega)=2\Sigma(\omega)$.

The DOS is obtained as usual [33] from the Green's functions:

$$g(\omega)=\frac{2\omega}{\pi}\frac{V}{3N}\int_{|\mathbf{k}|<k_D}\frac{d^3\mathbf{k}}{(2\pi)^3}[G''_L(k,\omega)+2G''_T(k,\omega)], \tag{11}$$

$\gamma\propto\langle\Delta G^2\rangle/\langle G\rangle^2$ is the parameter, which characterizes the state of disorder of the material. If γ exceeds a critical value γ_c the system becomes unstable; i.e., the level density $g(\omega^2)=g(\omega)/2\omega$ extends to values of $\omega^2<0$. As γ approaches γ_c the BP becomes more pronounced and shifts to lower frequencies [23–26].

Our other input parameters are the Debye wave number $k_D=\sqrt[3]{6\pi^2N/V}\approx 1/a$ (N is the number of atoms, V is the sample volume, and a is an interatomic spacing) the longitudinal and transverse sound velocities $v_{L,T}=\sqrt{v^2_{L,T,0}-\Sigma'_{L,T}(\omega=0)}$, and the correlation lengths $\xi_{1,2}$, ξ_G, which enter into the correlation functions $C_{1,2}(r)$, $C_G(r)$. In the present study we set $\xi_1=\xi_2=\xi_G\equiv\xi$ and assume Gaussian correlation functions of the form (8). This leaves only two adjustable parameters: γ and ξ [34]. For the polarized intensity and the depolarization ratio the ratio f_1/f_2 of the mean-squared Pockels constant fluctuations is needed as well.

In order to show that our theory works we study the example of glassy As_2S_3. In Fig. 1 it is demonstrated that the measured DOS and specific heat [7,35] can be accounted for by our SCBA theory. The correlation length, which fits the data, turns out to be just two interatomic spacings [34]. The specific heat has been calculated in the usual way from the DOS, weighted with the temperature derivative of $n(\omega)$ [25]. In Fig. 2 we use the same parameters (see figure captions) for calculating the VH intensity [Fig. 2(a)] and the depolarization ratio [Fig. 2(c)]. We see that the agreement with the measured data is satisfactory. We note that the position of the BP in $g(\omega)/\omega^2$ ($\sim$17 cm^{-1}) does *not* coincide at all with the maximum of the Raman intensity ($\sim$24 cm^{-1}). We conclude that the Raman maximum is *not* related with the BP of the reduced DOS. In order to explore deviations from the MB/Debye theory we have divided the data and the theoretical curves of Fig. 2(a) by ω^2. As to be expected, the MB curve decreases monotonically. In the data and in our theoretical curves an excess is visible which has its maximum near the position of the BP of $g(\omega)/\omega^2$. However, it is clear from comparing Figs. 1 and 2 that the Raman intensity is not related to the DOS in a simple way.

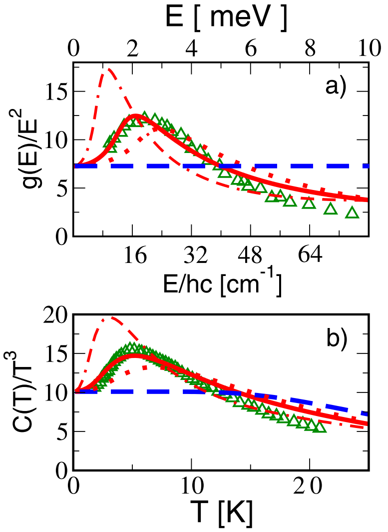

FIG. 1 (color online). (a) Measured reduced DOS [7] and reduced specific heat [35] of glassy As_2S_3 compared with theory: $v_T=1.4$ km/s, $v_L=1.8v_T$ [36], $k_D=1.32\times10^{10}$ m^{-1} from density data [37], $\gamma=0.835\gamma_c=0.352v_T^4$, $\xi_G=1/k_D$ (dots), $\xi_G=2/k_D$ (full line), $\xi_G=4/k_D$ (dash-dash-dotted line). The dashed lines correspond to the Debye model ($\gamma=0$).

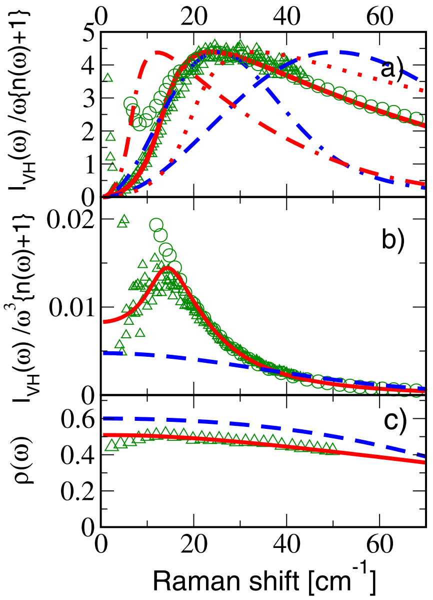

FIG. 2 (color online). (a) Measured reduced intensity $I_{VH}(\omega)/\omega[n(\omega)+1]$ of glassy As_2S_3 △: [36], ○: [7] compared with our theory using the same parameters and line codes as in Fig. 1. The dashed and dash-dotted lines are the Martin-Brenig theory (i.e., $\gamma=0$) with $\xi=2/k_D$ (dashed line) and $\xi=4/k_D$ (dash-dotted line); (b) same data divided by ω^2; only theory curves with $\xi=2/k_D$ are displayed. (c) Measured depolarization ratio [36] with theoretical curves calculated with $f_1/f_2=0.65$ and $\xi=2/k_D$.

In conclusion we have achieved to formulate a theory for low-frequency Raman scattering in disordered solids based on the model assumption of spatially fluctuating Pockels constants. We have shown that it is possible to describe the different vibrational spectral properties of a disordered solid within a unified theoretical framework. For the case of glassy As_2S_3 we demonstrated that the vibrational anomalies of specific heat and DOS can be reconciled with the Raman spectrum to obtain good agreement between theory and experiment.

W. S. is grateful for hospitality at University of Mainz. Helpful discussions with R. Schilling, A. Schulte, and J. Wiedersich are gratefully acknowledged.

---

*Now at Institut für Physik, Universität Mainz, D-55099 Mainz, Germany.

[1] B. J. Berne and R. Pecora, *Dynamic Light Scattering* (Wiley, New York, 1976).

[2] P. Y. Yu and M. Cardona, *Fundamentals of Semiconductors* (Springer, Berlin, 2001).

[3] At high frequencies (much higher than 100 cm^{-1}) the observed optical modes are essentially local bond modes which are still present in the amorphous state.

[4] J. Jäckle, in *Amorphous Solids: Low-Temperature Properties*, edited by W. A. Phillips (Springer, Heidelberg, 1981), p. 135.

[5] M. Montagna *et al.*, Phys. Rev. Lett. **65**, 1136 (1990); V. Mazzacurati *et al.*, Phys. Rev. B **45**, 2126 (1992); G. Viliani *et al.*, Phys. Rev. B **52**, 3346 (1995).

[6] J. S. Lannin, Phys. Rev. B **15**, 3863 (1977); E. Duval *et al.*, J. Phys. Condens. Matter **2**, 10 227 (1990); P. Benassi *et al.*, Phys. Rev. B **44**, 11 734 (1991); M. G. Zemlyanov *et al.*, Sov. Phys. JETP **74**, 151 (1992); T. Archibat *et al.*, J. Chem. Phys. **99**, 2046 (1993); A. P. Sokolov *et al.*, Phys. Rev. B **48**, 7692 (1993); E. Duval *et al.*, Phys. Rev. B **48**, 16 785 (1993); T. Ohsaka *et al.*, Phys. Rev. B **50**, 9569 (1994); N. V. Surovtsev *et al.*, Phys. Rev. Lett. **82**, 4476 (1999); V. N. Novikov and N. V. Surovtsev, Phys. Rev. B **59**, 38 (1999); L. Saviot *et al.*, Phys. Rev. B **60**, 18 (1999); E. Duval *et al.*, Philos. Mag. B **79**, 2051 (1999); B. Hehlen *et al.*, Phys. Rev. Lett. **84**, 5355 (2000); N. V. Surovtsev, Phys. Rev. E **64**, 061102 (2001); M. Ivanda *et al.*, Solid State Commun. **117**, 423 (2001); N. V. Surovtsev and A. P. Sokolov, Phys. Rev. B **66**, 054205 (2002); N. V. Surovtsev *et al.*, Phys. Rev. B **67**, 024203 (2003); N. V. Surovtsev, J. Phys. Condens. Matter **16**, 3035 (2004); Phys. Status Solidi (c) **1**, 2867 (2004); E. Courtens *et al.*, J. Phys. Condens. Matter **15**, S1279 (2003); S. N. Yannopoulos and K. S. Andrikopoulos, J. Chem. Phys. **121**, 4747 (2004); A. Schulte *et al.*, Vibrational Spectroscopy (to be published).

[7] V. K. Malinovsky *et al.*, Europhys. Lett. **11**, 43 (1990).

[8] N. J. Tao, G. Li, X. Chen, W. M. Du, and H. Z. Cummins, Phys. Rev. A **44**, 6665 (1991).

[9] S. N. Yannopoulos and G. N. Papatheodorou, Phys. Rev. B **62**, 3728 (2000).S. N. Yannopoulos and D. Th. Kastrissios, Phys. Rev. E **65**, 021510 (2002).

[10] J. P. Johari, J. Non-Cryst. Solids **307–310**, 114 (2002).

[11] S. N. Taraskin *et al.*, Phys. Rev. Lett. **97**, 055504 (2006).

[12] R. Stegeman *et al.*, Opt. Lett. **28**, 1126 (2003); Opt. Express **13**, 1144 (2005).

[13] R. Shuker and R. Gammon, Phys. Rev. Lett. **25**, 222 (1970).

[14] A. J. Martin and W. Brenig, Phys. Status Solidi (b) **64**, 163 (1974).

[15] V. L. Gurevich *et al.*, Phys. Rev. B **48**, 16 318 (1993).

[16] V. G. Karpov, M. I. Klinger, and F. N. Ignatev, Sov. Phys. JETP **57**, 439 (1983); U. Buchenau *et al.*, Phys. Rev. B **43**, 5039 (1991); **46**, 2798 (1992); U. Buchenau *et al.*, Phys. Rev. B **46**, 2798 (1992).

[17] See, e.g., [18–20] for an extended bibliography.

[18] J. Horbach *et al.*, Eur. Phys. J. B **19**, 531 (2001).

[19] T. Nakayama, Rep. Prog. Phys. **65**, 1195 (2002).

[20] V. L. Gurevich *et al.*, Phys. Rev. B **67**, 094203 (2003).

[21] R. O. Pohl, in Amorphous Solids: Low-Temperature Properties (Ref. [4]), p. 27.

[22] J. J. Freeman and A. C. Anderson, Phys. Rev. B **34**, 5684 (1986).

[23] W. Schirmacher, Europhys. Lett. **73**, 892 (2006).

[24] W. Schirmacher, M. Wagener, in *Dynamics of Disordered Materials*, edited by D. Richter, A. J. Dianoux, W. Petry, and J. Teixeira (Springer, Heidelberg, 1989), p. 231; W. Schirmacher and M. Wagener, Philos. Mag. B **65**, 607 (1992); R. Kühn and U. Horstmann, Phys. Rev. Lett. **78**, 4067 (1997); W. Schirmacher *et al.*, Phys. Rev. Lett. **81**, 136 (1998); S. N. Taraskin *et al.*, Phys. Rev. Lett. **86**, 1255 (2001); W. Götze and M. R. Mayr, Phys. Rev. E **61**, 587 (2000); J. W. Kantelhardt *et al.*, Phys. Rev. B **63**, 064302 (2001); T. S. Grigera *et al.*, Nature (London) **422**, 289 (2003); S.-H. Chong, Phys. Rev. E **74**, 031205 (2006); V. Gurarie and A. Altland, Phys. Rev. Lett. **94**, 245502 (2005).

[25] E. Maurer and W. Schirmacher, J. Low Temp. Phys. **137**, 453 (2004). In this article the fluctuating-elastic-force-constant model is compared with a local oscillator model.

[26] W. Schirmacher, G. Ruocco, and T. Scopigno, Phys. Rev. Lett. **98**, 025501 (2007).

[27] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon, New York, 1960), p. 391.

[28] B. Schmid, Diplom thesis, Technische Universität München, 2007 (unpublished, in English).

[29] L. Schäfer and F. Wegner, Z. Phys. B **38**, 113 (1980); A. J. McKane and M. Stone, Ann. Phys. (N.Y.) **131**, 36 (1981).

[30] S. John *et al.*, Phys. Rev. B **27**, 5592 (1983); **28**, 6358 (1983).

[31] W. Schirmacher *et al.*, Phys. Status Solidi (c) **5**, 862 (2008).

[32] A small positive imaginary part must be added to ω.

[33] E. N. Economou, *Green's Functions in Quantum Physics* (Springer, Heidelberg, 1979).

[34] It would be interesting to determine the fluctuation quantities γ and ξ for model solids by computer simulations. See, e.g., S. Sengupta *et al.*, Phys. Rev. E **61**, 1072 (2000).

[35] N. Ahmad, K. W. Hutt, and W. A. Phillips, J. Phys. C **19**, 3765 (1986).

[36] R. J. Nemanich, Phys. Rev. B **16**, 1655 (1977).

[37] D. Gerlich *et al.*, Phys. Rev. B **20**, 2529 (1979).

[38] M. Montagna *et al.*, J. Raman Spectrosc. **27**, 707 (1996).