

Frequency behavior of Raman coupling coefficient in glasses

N. V. Surovtsev¹ and A. P. Sokolov²

¹*Institute of Automation and Electrometry, Russian Academy of Sciences, pr. Ak. Koptuyga 1, Novosibirsk, 630090, Russia*

²*Department of Polymer Science, University of Akron, Akron, Ohio 44325-3909*

(Received 8 December 2001; revised manuscript received 12 April 2002; published 29 August 2002)

The low-frequency Raman coupling coefficient $C(\nu)$ of 11 different glasses is evaluated. It is shown that the coupling coefficient demonstrates a universal linear frequency behavior $C(\nu) \propto (\nu/\nu_{BP} + B)$ near the boson peak maximum ν_{BP} . The frequency dependence of $C(\nu)$ allows us to separate the glasses studied into two groups: the first group has a frequency independent contribution $B \sim 0.5$, while the second one has $B \sim 0$. It was found that $C(\nu)$ demonstrates a superlinear behavior at very low frequencies. This observation suggests a vanishing of the coupling coefficient when frequency tends to zero.

DOI: 10.1103/PhysRevB.66.054205

PACS number(s): 63.50.+x, 64.70.Pf, 63.20.Pw

I. INTRODUCTION

One of the most interesting topics in solid-state physics is the nature of the low-frequency (0.1–3 THz) collective vibrations in glasses. While these frequencies are in the range of acoustic excitations, there is experimental evidence that the vibrations are not pure acoustic plane waves and their density of vibrational states $g(\nu)$ does not follow the Debye behavior ($\propto \nu^2$, where ν is the frequency). A maximum in $g(\nu)/\nu^2$ that appears at some frequency ν_{BP} is usually called the boson peak. Vibrations around the boson peak can be studied by several experimental techniques: low-temperature specific heat and thermal conductivity,¹ inelastic neutron² and x-ray^{3,4} scattering, infrared absorption,⁵ and Raman scattering.⁶ In the case of the low-frequency Raman spectroscopy, the density of vibrational states appears in the light scattering spectrum via the so-called light-vibration coupling coefficient, $C(\nu)$ ⁷

$$I(\nu) = C(\nu)g(\nu) \frac{n+1}{\nu}, \quad (1)$$

where $I(\nu)$ is the Raman intensity for the Stokes side of the spectrum and n is the Bose factor.

Knowledge of $C(\nu)$ and an understanding of its frequency dependence have significant importance for the topic of low-frequency vibrations. First of all, knowledge of $C(\nu)$ provides a relatively simple method to extract the vibrational density of states from a Raman experiment. Second, the light-vibration coupling coefficient contains information on the vibrational wave function⁷ and, therefore, can be used as a test of different models.

Two classical models suggested for the description of $C(\nu)$ lead to different predictions: (i) Shucker and Gammon⁷ assumed that vibrations are localized on a distance much shorter than the light wavelength and predicted $C(\nu) = \text{const}$, while (ii) Martin and Brenig⁸ have demonstrated that a polarizability disorder mechanism applied to slightly damped acoustic waves leads to $C(\nu) \sim \nu^2$ behavior at low frequencies and a peak at higher frequencies, related to a correlation length of the polarizability fluctuations. It was shown that quasi-plane acoustic waves with finite mean free path l will also contribute to the low-frequency Raman spec-

trum with $C(\nu) \sim \nu^2$, when $l^{-1} \propto \nu^4$ (Refs. 6 and 9) or with $C(\nu) = \text{const}$, when $l^{-1} \propto \nu^2$ Refs. 9 and 10.

There are a few challenges for experimental evaluation of the true vibration coupling coefficient: Very-low-temperature data for both Raman spectra and $g(\nu)$ should be used in order to avoid a quasielastic contribution (fast relaxation) (Refs. 6 and 11); it is not obvious whether all vibrations at one frequency contribute to the Raman spectra with the same $C(\nu)$, or there are different kinds of vibrations and each contributes with its own $C(\nu)$. A comparison of the low-temperature low-frequency Raman spectra of glasses with the total $g(\nu)$ obtained from low-temperature specific heat or inelastic neutron data has demonstrated that the coupling coefficient appears to vary nearly linearly with frequency.^{12–16}

However, this comparison did not consider the possibility that two different kinds of vibrational excitations could coexist around the boson peak. Although most of the authors at present accept the idea that the vibrations around the boson peak are strongly hybridized and cannot be easily separated, the question is not yet completely settled. This question became especially important in the light of the results of hyper-Raman scattering experiments.¹⁷ The existence of differences in the behavior of THz spectra in Raman and hyper-Raman scattering experiments was interpreted as evidence of the coexistence of two types of vibrational excitations. Also, there are theoretical approaches describing the THz dynamics of glasses as the coexistence of two different types of vibrations in this spectral range (for example, Refs. 18 and 19). In this case, the Raman coupling coefficient can lose its good physical meaning.²⁰ One of the strong arguments in favor of the existence of a single type of vibrational excitation could be the universal behavior of $C(\nu)$ for glasses with various structures. This universality suggests that the two hypothetical types of vibrations are interrelated.

A detailed analysis performed for silica glass has shown that $C(\nu)$ varies linearly with frequency,

$$C(\nu) = A(\nu/\nu_{BP} + B), \quad (2)$$

in the range 10–50 cm^{-1} .²¹ This result was interpreted in Ref. 21 as evidence that the coupling coefficient extrapolates

TABLE I. References used for $C(\nu)$ calculation in the present work [for Raman data, neutron scattering data, specific heat data $C_p(T)$], previously published $C(\nu)$, and the boson peak position, defined as the position of maximum of g/ν^2 , for different glasses.

	Glass	Raman	Neutron	$C_p(T)$	$C(\nu)$	BP position [cm^{-1}]
1	SiO_2	25,26	21,27	28	12,21	33.5
2	B_2O_3	29		30,31	18	18
3	$(\text{Ag}_2\text{O})_{0.14}(\text{B}_2\text{O}_3)_{0.86}$	32		32		22.5
4	Se	13	33	34,35		12
5	As_2S_3	13	36	14		16.5
6	CKN	37	38	39		20.5
7	GeSe_2	40	41	41		10
8	GeO_2	42		42		27
9	PC				10	11
10	PS	43		31,44	12	11.5
11	PMMA	45		31	46	12.5

to a nonvanishing value in the limit $\nu \rightarrow 0$. However, it was shown in Ref. 22 that the coupling coefficient demonstrates a superlinear behavior just below 10 cm^{-1} , i.e., the observed linear behavior cannot be extrapolated to zero frequency. It would be very important to know whether this behavior is general also for other glasses.

The present contribution analyzes the frequency behavior of the coupling coefficient in a broad set of different glasses, strong and fragile, covalently and ionically bonded, low molecular weight, and polymeric. It is shown that all glasses demonstrate the linear behavior of $C(\nu)$ [Eq. (2)] around the boson peak frequency. One of the most striking results is that there are two groups of glasses. One has a frequency-independent contribution B with a universal value ~ 0.5 , while the second group of glasses has $B \approx 0$. An interpretation of the results is proposed and a correlation with low-temperature thermal conductivity is found.

II. COUPLING COEFFICIENT EVALUATION

The density of vibrational states must be known in order to extract the Raman coupling coefficient [see Eq. (1)]. It has been shown^{6,11,23} that relaxationlike processes give a significant contribution to the Raman spectra and $g(\nu)$ at frequencies below the boson peak even at temperatures as low as 50 K. Thus, experimental data obtained at T below 50 K should be used for extracting vibrational $g(\nu)$. Two experimental techniques provide information on $g(\nu)$: inelastic neutron scattering² and measurements of low-temperature specific heat.¹ The latter has a few advantages: (i) the number of glasses for which specific heat data are available is much larger than the number of glasses for which inelastic neutron scattering data are available; (ii) the density of states calculated from low-temperature specific heat data corresponds to a very low temperature, where usually no neutron data are available. While in the past only a phenomenological analysis was available for extraction of the coupling coefficient from comparison of the specific heat and Raman data (for example, Refs. 13 and 24), recently it was shown that the integral equation for the specific heat temperature depen-

dence can be solved numerically and therefore the density of vibrational states may now be obtained from heat capacity measurements.²²

By using published data for the low-frequency Raman spectra, inelastic neutron scattering data, and the low-temperature specific heat we have collected the coupling coefficient for 11 glasses of different types: strong, covalent glasses ($\text{SiO}_2, \text{B}_2\text{O}_3, \text{As}_2\text{S}_3, \text{GeO}_2, \text{GeSe}_2$), binary covalent glass $(\text{Ag}_2\text{O})_{0.14}(\text{B}_2\text{O}_3)_{0.86}$, covalent glass of medium fragility (Se), fragile, ionically bonded glass (CKN), and polymer glasses: polystyrene (PS), polycarbonate (PC), and polymethylmethacrylate (PMMA). The references used for these glasses are shown in Table I. Table I also presents values of ν_{BP} for various glasses defined as the position of the maximum in the curve $g(\nu)/\nu^2$.

More details of the coupling coefficient calculation, figures for the frequency dependence of the coupling coefficient, comparisons between different data, or ways of calculation are presented in the supplied material.⁴⁷ Figure 1 shows the most striking result that two glasses with extreme fragilities— SiO_2 and CKN—show a very similar frequency behavior in the low-frequency part of the Raman spectrum. It can be described by the linear behavior near the frequency of

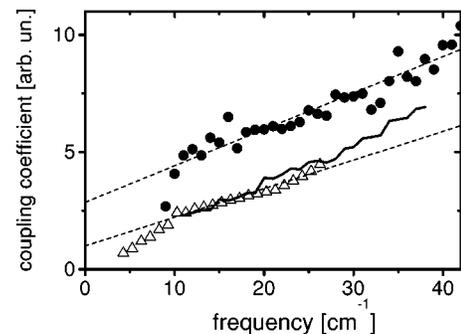


FIG. 1. Frequency dependence of the coupling coefficient $C(\nu)$ for SiO_2 (circles) and CKN glasses (triangles correspond to density of states evaluated from specific heat data; lines are for density of states from inelastic neutron scattering). Dashed lines are descriptions by linear dependence.

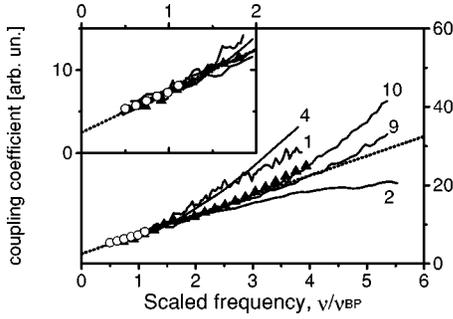


FIG. 2. Frequency dependence of the coupling coefficient $C(\nu)$ for glasses: SiO_2 , B_2O_3 , Se, CKN, $(\text{Ag}_2\text{O})_{0.14}(\text{B}_2\text{O}_3)_{0.86}$, PS, and PC, vs scaled frequency ν/ν_{BP} . Only region above $0.5\nu_{BP}$ is presented. Numbers of lines correspond to the numbers in Table I. Triangles are $(\text{Ag}_2\text{O})_{0.14}(\text{B}_2\text{O}_3)_{0.86}$; circles are CKN data. The dashed line is a fit $C(\nu) \propto \nu/\nu_{BP} + 0.5$. The inset shows the low-frequency part of $C(\nu)$ in detail.

the boson peak maximum. This observation is important because the density of vibrational states at the boson peak in SiO_2 exceeds the Debye level by ~ 4 times [i.e., $g(\nu)$ is dominated by the excess vibrations] while in CKN the excess vibrations are only ~ 0.4 of the Debye level. Nevertheless, $C(\nu)$ appears to be very similar.

III. GENERAL FEATURES OF $C(\nu)$

The results for the coupling coefficient shown in the figures of Ref. 47 indicate that the frequency behavior of the coupling coefficient can be considered in three frequency ranges: significantly below the frequency of the boson peak maximum, ν_{BP} ; around ν_{BP} ; and significantly above ν_{BP} . We compare data for various glasses with the frequency axis scaled to ν_{BP} .

A. Linear dependence of $C(\nu)$ near ν_{BP}

A linear behavior of $C(\nu)$ for frequencies near that corresponding to the boson peak maximum can be seen for all the glasses. This linear behavior can be described by Eq. (2). The constant B characterizes the relative contribution of two additive terms in Eq. (2). Figure 2 presents a plot of $C(\nu)$ for seven glasses [SiO_2 , B_2O_3 , Se, CKN, $(\text{Ag}_2\text{O})_{0.14}(\text{B}_2\text{O}_3)_{0.86}$, PS, PC] plotted against scaled frequency [amplitudes of $C(\nu)$ were normalized near $\nu/\nu_{BP} = 1$]. For clarity, only data above $0.5\nu_{BP}$ are presented in this figure. Clear differences in $C(\nu)$ of the different glasses are observed at high frequencies. However, $C(\nu)$ tends to a master curve (universal frequency dependence) at frequencies below $\sim 1.5\nu_{BP}$. The universal behavior shown by the dashed line presents the dependence

$$C(\nu) \propto \nu/\nu_{BP} + 0.5. \quad (3)$$

The linear frequency dependence describes well the behavior of $C(\nu)$ found experimentally starting from the frequency $\sim 0.5\nu_{BP}$. The high-frequency limit of this behavior varies from $1.5\nu_{BP}$ for SiO_2 and Se up to about $4\nu_{BP}$ for the PC glass.

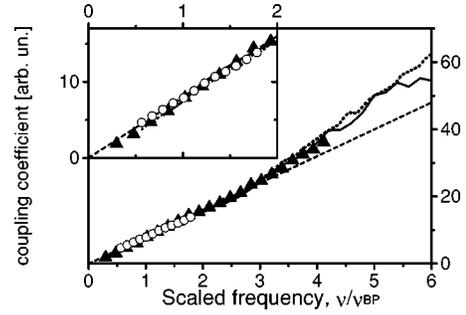


FIG. 3. Frequency dependence of the coupling coefficient $C(\nu)$ for glasses: PMMA (dotted line), As_2S_3 (triangles), GeSe_2 (solid line), and GeO_2 (circles) vs scaled frequency ν/ν_{BP} . The dashed line is a fit $C(\nu) \propto \nu$. The inset shows the low-frequency part of $C(\nu)$ in detail.

However, there exists another group of glasses that does not follow the frequency behavior highlighted in Fig. 2. The results for the other four glasses (PMMA, As_2S_3 , GeSe_2 , GeO_2) are presented in Fig. 3. Here $C(\nu)$ for these glasses can be well described by a simple linear dependence with the constant B in Eq. (2) having a value of zero.

Thus, all the glasses analyzed here are separated into two groups: those with $C(\nu) \propto \nu/\nu_{BP} + 0.5$ near the boson peak maximum (Fig. 2) and another group with $C(\nu) \propto \nu$ (Fig. 3). In the following we will refer to these two groups with the designation of “type-I” and “type-II,” respectively.

B. Low-frequency behavior of $C(\nu)$ ($\nu < 0.5\nu_{BP}$)

At least four glasses (SiO_2 , Se, PS, CKN) demonstrate a superlinear frequency dependence in this spectral range. The low-frequency portions of $C(\nu)$ for these glasses are presented in Fig. 4 on a log-log scale together with the function $C(\nu) \propto \nu/\nu_{BP} + 0.5$. The coupling coefficient varies superlinearly below some frequency $\sim 0.5\nu_{BP}$, deviating strongly from the extrapolation of linear behavior (Fig. 4). The crossover frequency of a transition to superlinear behavior appears to be $\sim 0.3\nu_{BP}$ for SiO_2 and $\sim 0.5\nu_{BP}$ for Se, PS, and CKN.

It is remarkable that these systems have significantly different microstructure and fragility. This suggests that the su-

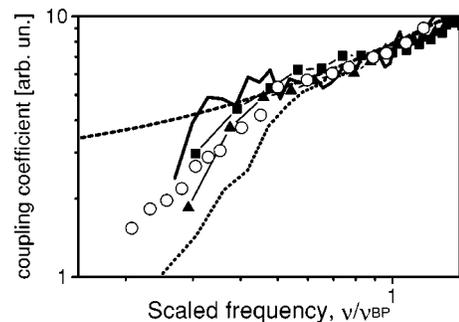


FIG. 4. The low-frequency part of the coupling coefficient $C(\nu)$ for glasses: SiO_2 (solid line), Se (triangles), PS (squares), and CKN (circles) in logarithmic scale. The dashed line is $C(\nu) \propto \nu/\nu_{BP} + 0.5$. The dotted line is $C(\nu)$ for the corrected Raman spectrum of B_2O_3 glass as explained in the text.

perlinear frequency behavior for $\nu < (0.3-0.5)\nu_{BP}$ may be general for various glasses. The fact that we did not observe the superlinear frequency behavior of $C(\nu)$ in other glasses can be explained by two reasons: either the experimental data are not extended to low enough frequencies or they are measured at temperatures that are not low enough and the presence of the fast relaxation at low frequencies masks the true vibrational behavior. The importance of the relaxation contribution even at temperature as low as $T=15$ K can be demonstrated in the case of the B_2O_3 glass. Indeed, from Fig. 1 of Ref. 29 it is evident that the fast relaxation is not negligible at $T=15$ K and dominates for $\nu < 3$ cm^{-1} . Since the spectral shape of the fast relaxation spectrum in B_2O_3 does not depend on temperature,²⁹ we can subtract it from the Raman spectrum at $T=15$ K using the spectrum of the fast relaxation determined in Ref. 29. The Raman spectrum of B_2O_3 glass corrected in this way (by adjusting amplitude of the relaxational spectrum at the lowest points of the spectrum in Fig. 1 of Ref. 29) gives the coupling coefficient shown by the dotted line in Fig. 4. This revised coupling coefficient depicts the superlinear behavior at $\nu < 0.5\nu_{BP}$.

C. High-frequency behavior of $C(\nu)$ ($\nu > 2\nu_{BP}$)

Figures 2 and 3 show no universal behavior of the coupling coefficient in this frequency range. It varies from sub-linear to strongly superlinear behavior for different glasses.

IV. DISCUSSION

The observation of the superlinear behavior of the coupling coefficient below some frequency $\nu < (0.3-0.5)\nu_{BP}$ is very important. It has been shown that $C(\nu)$ for acousticlike vibrations should increase $\sim \nu^2$. This prediction was obtained in the framework of different model approximations (see, for example, Refs. 6,8 and 9).

Based on their experimental observations, the authors of Ref. 21 suggested that the linear behavior of $C(\nu)$ can be extrapolated to the limit $\nu \rightarrow 0$ and $C(\nu=0)$ has a nonvanishing value. The results of the present work show that this extrapolation is not correct and the character of the frequency dependence changes at lower ν , corresponding to the expectation that $C(\nu) \rightarrow 0$ when $\nu \rightarrow 0$. However, the existing experimental data do not allow one to establish the exact frequency dependence, and this topic still requires further investigation.

At higher frequencies, $C(\nu)$ demonstrates the universal linear behavior for type-I glasses (Fig. 2). The glasses in this class vary significantly in structure, fragility, and ratio of the excess vibrations to the Debye level. There are many models that assume two different kinds of vibrations coexisting at frequencies around the boson peak: propagating and localized or quasilocal. For example, in the framework of the soft potential model⁴⁸ it is assumed that propagating waves have a Debye-like density of states and do not contribute to the Raman spectra, while excess vibrations are localized and have $C(\nu) = \text{const}$. The ratio of the excess vibrational density of states to the Debye level around the boson peak is ~ 4 in SiO_2 and ~ 0.4 in KFN (Ref. 39); i.e., it differs up to 10

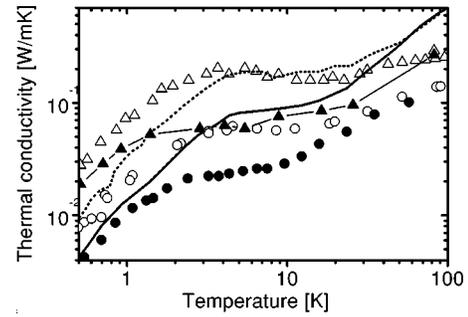


FIG. 5. Thermal conductivity of SiO_2 (solid line), GeO_2 (dotted line), PMMA (open circles), PS (solid circles), As_2S_3 (open triangles), and Se (solid triangles). Data are taken from Refs. 49–51.

times. In that respect, the observed universality of $C(\nu)$, obtained using the total density of vibrational states (Figs. 1–3), supports an alternative idea that all vibrations around the boson peak are hybridized and cannot be separated into propagating and localized.

In order to explain the observed universality of $C(\nu)$, significant theoretical work should be done. A simple model implying that the boson peak vibrations combine properties of both localized and extended excitations was proposed in Ref. 47. According to this model, the relative contributions to $C(\nu)$ of a frequency-independent term and a term proportional to frequency reflect the relative weights of the localized and extended parts of the boson peak vibration. The result of Fig. 2 means that at the boson peak maximum the ratio of the localized and extended parts is the same for these glasses in the framework of Ref. 47.

However, the frequency-independent contribution to $C(\nu)$ for some of glasses is negligibly small (Fig. 3). We do not have a clear explanation for the observed difference and it remains a challenge for future investigations. At present we only show another hint that the peculiarity of type-II glasses may be related to weaker localization of the boson peak vibrations (at least this is expected from the model⁴⁷). This difference between two types of glasses has to show up in vibration transport properties. Figure 5 presents the thermal conductivity of SiO_2 , PS, Se, GeO_2 , PMMA, and As_2S_3 glasses (data from Refs. 49–51). The first three glasses are type-I and the next three are type-II. It is convenient to compare the pairs of glasses in which the two members of the pair have closely similar chemical nature but belong to different classes, for example, SiO_2 and GeO_2 , PS and PMMA, and As_2S_3 and Se. It appears (Fig. 5) that glasses of different type (but of a similar chemical nature) have comparable thermal conductivity at higher T but type-II glasses have higher thermal conductivity at the plateau. It is known that the plateau region in thermal conductivity corresponds to conductivity by vibrations around the boson peak. Thus, this comparison reveals weaker localization of the boson peak vibrations in type-II glasses and supports the above speculations. However, the question is far from settled and further investigations are needed in order to provide a microscopic explanation of the difference between the two types of glasses.

There are no universalities in the frequency dependence

of $C(\nu)$ for $\nu > 2\nu_{BP}$. The high-frequency vibrations depend strongly on a particular atomic organization of a glass, its microstructure. A relation to peculiar microstructure may be the reason for different behaviors of $C(\nu)$ in this frequency range.

V. CONCLUSION

The Raman coupling coefficient $C(\nu)$ is analyzed for a large number of glasses strongly different in their chemical structure and fragility. It is demonstrated that $C(\nu)$ has a universal linear frequency dependence near the boson peak maximum: $C(\nu) \propto \nu/\nu_{BP} + B$, with $B \sim 0.5$ for one group of glasses and $B \sim 0$ for the second group. The observed universality suggests that the vibrations around the boson peak have some universal properties for glasses with different structure. It is found that the difference in the behavior of

$C(\nu)$ in the two groups of glasses correlates with the observation of different behavior of thermal conductivity in these two types of glasses. It is also shown that $C(\nu)$ has a super-linear behavior at frequencies below $\sim (0.3-0.5)\nu_{BP}$. A sharp rise in mean free path of the vibrations with a decrease in ν may be the reason for this fast decrease in $C(\nu)$. No universality is observed at higher frequencies (above $\sim 2\nu_{BP}$), suggesting that the particular atomic organization of glasses is important in this spectral range.

ACKNOWLEDGMENTS

The help of S. Adichtchev in the literature search is appreciated. This work was supported by RFFI Grant Nos. 01-05-65066 and 02-02-16112. A.P.S. acknowledges financial support from the NSF (Grant No. DMR-0080035) and NATO (Grant No. PST.CLG.976150).

-
- ¹*Amorphous Solids: Low-Temperature Properties*, edited by W. A. Phillips (Springer, Berlin, 1981).
- ²U. Buchenau, N. Nücker, and A.J. Dianoux, Phys. Rev. Lett. **53**, 2316 (1984).
- ³P. Benassi, M. Krisch, C. Masciovecchio, V. Mazzacurati, G. Monaco, G. Ruocco, F. Sette, and R. Verbeni, Phys. Rev. Lett. **77**, 3835 (1996).
- ⁴M. Foret, E. Courtens, R. Vacher, and J.B. Suck, Phys. Rev. Lett. **77**, 3831 (1996).
- ⁵U. Strom and P.C. Taylor, Phys. Rev. B **16**, 5512 (1977).
- ⁶J. Jäckle, in *Amorphous Solids: Low-Temperature Properties*, edited by W. A. Phillips (Springer, Berlin, 1981).
- ⁷R. Shuker and R.W. Gammon, Phys. Rev. Lett. **25**, 222 (1970).
- ⁸A.J. Martin and W. Brenig, Phys. Status Solidi B **64**, 163 (1974).
- ⁹E. Duval, L. Saviot, N. Surovtsev, J. Wiedersich, and A.J. Dianoux, Philos. Mag. B **79**, 2051 (1999).
- ¹⁰L. Saviot, E. Duval, N. Surovtsev, J.F. Jal, and A.J. Dianoux, Phys. Rev. B **60**, 18 (1999).
- ¹¹G. Winterling, Phys. Rev. B **12**, 2432 (1975).
- ¹²A.P. Sokolov, U. Buchenau, W. Steffen, B. Frick, and A. Wischnewski, Phys. Rev. B **52**, R9815 (1995).
- ¹³A.P. Sokolov, A. Kisliuk, D. Quitmann, and E. Duval, Phys. Rev. B **48**, 7692 (1993).
- ¹⁴N. Ahmad, K.W. Hutt, and W.A. Phillips, J. Phys. C **19**, 3765 (1986).
- ¹⁵N. Ahmad and Matiulah, Solid State Commun. **76**, 433 (1990).
- ¹⁶A. Fontana, F. Rocca, M.P. Fontana, B. Rosi, and A.J. Dianoux, Phys. Rev. B **41**, 3778 (1990).
- ¹⁷B. Hehlen, E. Courtens, R. Vacher, A. Yamanaka, M. Kataoka, and K. Inoue, Phys. Rev. Lett. **84**, 5355 (2000).
- ¹⁸D. Engberg, A. Wischnewski, U. Buchenau, L. Börjesson, A.J. Dianoux, A.P. Sokolov, and L.M. Torell, Phys. Rev. B **58**, 9087 (1998).
- ¹⁹V.G. Karpov, M.I. Klinger, and F.N. Ignatiev, Sov. Phys. JETP **57**, 439 (1983).
- ²⁰E. Courtens, M. Foret, B. Hehlen, and R. Vacher, Solid State Commun. **117**, 187 (2001).
- ²¹A. Fontana, R. Dell'Anna, M. Montagna, F. Rossi, G. Viliani, G. Ruocco, M. Sampoli, U. Buchenau, and A. Wischnewski, Europhys. Lett. **47**, 56 (1999).
- ²²N.V. Surovtsev, Phys. Rev. E **64**, 061102 (2001).
- ²³N.V. Surovtsev, T. Achibat, E. Duval, A. Mermet, and V.N. Novikov, J. Phys.: Condens. Matter **7**, 8077 (1995).
- ²⁴A. Fontana, F. Rossi, G. Carini, G. D'Angelo, G. Tripodo, and A. Bartolotta, Phys. Rev. Lett. **78**, 1078 (1997).
- ²⁵A.P. Sokolov, V.N. Novikov, and B. Strube, Europhys. Lett. **38**, 49 (1997).
- ²⁶N.V. Surovtsev, J. Wiedersich, V.N. Novikov, E. Rössler, and E. Duval, Phys. Rev. Lett. **82**, 4476 (1999).
- ²⁷U. Buchenau, M. Prager, N. Nücker, A.J. Dianoux, N. Ahmad, and W.A. Phillips, Phys. Rev. B **34**, 5665 (1986).
- ²⁸Y. Inamura, M. Arai, O. Yamamuro, A. Inaba, N. Kitamura, T. Otomo, T. Matsuo, S.M. Bennington, and A.C. Hannon, Physica B **263-264**, 299 (1999).
- ²⁹N.V. Surovtsev, J.A.H. Wiedersich, E. Duval, V.N. Novikov, E. Rössler, and A.P. Sokolov, J. Chem. Phys. **112**, 2319 (2000).
- ³⁰E. Pérez-Enciso, M.A. Ramos, and S. Vieira, Phys. Rev. B **56**, 32 (1997).
- ³¹G.K. White, S.J. Collocott, and J.S. Cook, Phys. Rev. B **29**, 4778 (1984).
- ³²G. Tripodo, G. D'Angelo, G. Carini, A. Bartolotta, A. Fontana, and F. Rossi, J. Phys.: Condens. Matter **11**, A229 (1999).
- ³³W.A. Phillips, U. Buchenau, N. Nücker, A.-J. Dianoux, and W. Petry, Phys. Rev. Lett. **63**, 2381 (1989).
- ³⁴J.C. Lasjaunias and R. Maynard, J. Non-Cryst. Solids **6**, 101 (1971).
- ³⁵R.C. Zeller and R.O. Pohl, Phys. Rev. B **6**, 2029 (1971).
- ³⁶V.K. Malinovsky, V.N. Novikov, and A.P. Sokolov, Phys. Lett. A **153**, 63 (1991).
- ³⁷N.V. Surovtsev, J.A.H. Wiedersich, V.N. Novikov, E. Rössler, and A.P. Sokolov, Phys. Rev. B **58**, 14 888 (1998).
- ³⁸M. Russina, F. Mezei, R. Lechner, S. Longeville, and B. Urban, Phys. Rev. Lett. **84**, 3630 (2000).
- ³⁹A.P. Sokolov, R. Calemczuk, B. Salce, A. Kisliuk, D. Quitmann, and E. Duval, Phys. Rev. Lett. **78**, 2405 (1997).
- ⁴⁰S. Sugai, Phys. Rev. B **35**, 1345 (1987).

- ⁴¹W.A. Kamitakahara, R.L. Cappelletti, P. Boolchand, B. Halfpap, F. Gompf, D.A. Neumann, and H. Mutka, *Phys. Rev. B* **44**, 94 (1991).
- ⁴²G. Carini, G. D'Angelo, G. Tripodo, A. Fontana, A. Leonardi, G.A. Saunders, and A. Brodin, *Phys. Rev. B* **52**, 9342 (1995).
- ⁴³V.N. Novikov, A.P. Sokolov, B. Strube, N.V. Surovtsev, E. Duval, and A. Mermet, *J. Chem. Phys.* **107**, 1057 (1997).
- ⁴⁴B.V. Lebedev, N.N. Smirnova, N.K. Kozyreva, A.I. Kirilin, and V.V. Korchak, *Dokl. Akad. Nauk SSSR* **270**, 129 (1983).
- ⁴⁵N.V. Surovtsev, A. Mermet, E. Duval, and V.N. Novikov, *J. Chem. Phys.* **104**, 6818 (1996).
- ⁴⁶A. Mermet, N.V. Surovtsev, E. Duval, J.F. Jal, J. Dupuy-Philon, and A.J. Dianoux, *Europhys. Lett.* **36**, 277 (1996).
- ⁴⁷N.V. Surovtsev and A.P. Sokolov, cond-mat/0206295 (unpublished).
- ⁴⁸D.A. Parshin, *Phys. Solid State* **36**, 991 (1994).
- ⁴⁹C.C. Yu and J.J. Freeman, *Phys. Rev. B* **36**, 7620 (1987).
- ⁵⁰J.E. Graebner, B. Golding, and L.C. Allen, *Phys. Rev. B* **34**, 5696 (1986).
- ⁵¹D.G. Cahill and R.O. Pohl, *Phys. Rev. B* **35**, 4067 (1987).