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Low-frequency Raman scattering in glasses

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Low-frequency (10–80 cm^{-1}) Raman spectra of glasses are considered theoretically. It is shown that taking into account the interaction of light both with phononlike and quasilocal vibrational states leads to the violation of the famous Shuker-Gammon formula for the scattering intensity. We find that this violation can explain the experimental results comparing Raman and inelastic-neutron-scattering spectra. Expressions allowing, in principle, the determination of the parameters of the interaction of light with both phononlike vibrations and quasilocal oscillations from experiments are derived.

I. INTRODUCTION

Low-frequency ($\omega \leq 80$ cm^{-1}) vibrational spectra of inelastic light scattering (Raman spectra) in glasses have been intensively investigated both experimentally and theoretically during the last two decades. One can find a review of early investigations in Ref. 1, and the recent results were reviewed in Ref. 2. The most noticeable feature of Raman spectra in glasses as compared with crystals is a low-frequency broad peak, the maximum of which falls in the frequency range $\omega \sim 20$–60 cm^{-1}. In crystals only acoustic phonons exist in this region, which manifest themselves in the first-order Raman spectra at frequencies $\omega \leq 1$ cm^{-1} as Mandelstam-Brillouin lines. Therefore it is evident that the most general cause for the observed spectra is the disorder of the glass structure, which leads to violation of wave vector selection rules.

The temperature dependence of the spectra in the peak region[1] appears to be determined by a Bose distribution function:

$$n(\omega) = \left[\exp\left(\frac{\hbar\omega}{k_B T}\right) - 1\right]^{-1}.$$

This behavior is routine for first-order Raman spectra, and is caused by scattering from harmonic vibrational excitations, which obey Bose statistics. This fact gave the name "boson peak" to the discussed feature of the spectra.

In the range $\omega \leq 10$ cm^{-1} the frequency and temperature dependencies of the scattered intensity are considerably altered. The scattering in this region is known as excess or quasielastic scattering.[3] We will not be concerned with this region in the present paper.

The problem of the boson peak has assumed additional significance with recognition of the fact that the low-frequency dynamics of glasses display a number of universal anomalies (as compared to the Debye model).[4] In the frequency region considered in our paper, these anomalies manifest themselves in substantial excess of the vibrational density of states above the Debye value. This fact has been directly observed in inelastic-neutron-scattering experiments.[5] Such behavior of the density of states leads to a departure of the temperature dependence of the heat capacity C from the T^3 law: the function $C(T)/T^3$ has a broad maximum at $T \sim 5$–10 K.[4] The heat conductivity was also found to have a very unusual temperature dependence at the same temperatures (the famous "plateau"[6]).

To explain these observations it has been assumed that there exist some quasilocal vibrations in glasses at THz frequencies, which coexist with phonons. This assumption was supported by computer simulations.[7] The most developed theory of glass dynamics involving this assumption has been elaborated on the basis of the "soft potential model."[8] According to this model, local fluctuations of the glass structure can give rise to a strong softening of force parameters for some fraction of the atoms. As a result, double-well potentials and soft single-well potentials can appear in glasses. The double-well potentials are responsible for the dynamics of glasses at very low frequencies, which are not considered in this paper, while vibrations of atoms in the soft single-well potentials have the above-mentioned quasilocal nature. A detail presentation of the soft potential model can be found in Refs. 9–11.

An early explanation of the boson peak was suggested by Martin and Brenig.[12] Within the Martin-Brenig model the boson peak was related to the light scattering from long-wave acoustic phonons. This scattering is admissible in the whole wave number region due to the lack of translational symmetry in glasses. In this model phonons were postulated as propagating in homogeneous continuum media and the disorder of the glass structure was suggested to influence only the light-phonon coupling parameters. In Ref. 12 the scattering intensity was found to be

$$I(\omega) \sim \omega[n(\omega) + 1]g_D(\omega)S\left(\frac{\omega}{v}\right), \tag{1}$$

where $g_D(\omega)$ is the Debye vibrational state density, $S(\omega/v)$ is the Fourier transform of the correlation function of the spatial fluctuations of the elasto-optic parameters, and v is a sound velocity. The expression (1) de-

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scribes the ω^2 dependence of the reduced intensity defined as

$$I_R(\omega) = \frac{I(\omega)}{\omega[n(\omega)+1]} \tag{2}$$

at $\omega \ll \omega_{\max}$, observed in several experiments[13] [$\omega_{\max}$ here is the frequency of the maximum of $I_R(\omega)$]. Equation (1) also describes adequately the frequency dependence of $I_R(\omega)$ in a wide frequency interval around $\omega_{\max}$, if $S(\omega/v)$ is chosen in the form of the Gauss function. Owing to this fact the Martin-Brenig model was widely used to treat experimental data (see Ref. 1 and Ref. 2). Summing up, one can say that the shape of the intensity curve $I_R(\omega)$ is determined in this model by the form of the function $S(\omega/v)$, and the position of the maximum $\omega_{\max}$ is determined by the radius of correlation of the inhomogeneities of the elasto-optic parameters.

The second important theoretical result related to Raman scattering in glasses is due to Shuker and Gammon.[14] They argued that the reduced intensity of the first-order Raman scattering in glasses can be presented in the form

$$I_R(\omega) = \sum_b c_b(\omega)\frac{g_b(\omega)}{\omega^2}, \tag{3}$$

where $g_b(\omega)$ is the density of states of the bth vibrational mode and $c_b(\omega)$ is an effective coupling parameter of this mode with light. In deducing this expression it was, in fact, postulated that vibrational excitations in glasses are localized in some finite region. If one admits that only one mode makes a considerable contribution to scattering, then the sum in Eq. (3) contains only one term. In just such a form this result was used in further experimental[15–17] and theoretical[18] investigations. Based on the Shuker-Gammon formula (3) one can suggest that the boson peak arises due to the anomalous behavior of the density of states rather than the frequency dependence of the coupling parameter. Such an approach has been realized in a recent paper,[19] where in the framework of the soft potential model the intensity has been found in the form of Eq. (3) with $c(\omega)$ =const. It was suggested in Ref. 19 that phonons make no contribution to the scattering of light, which is, hence, caused by the interaction of light with quasilocal vibrations only.

The main question regarding the nature of the boson peak can be stated now as follows. The frequency dependence of which factor, the effective coupling parameter or the vibrational state density, mainly determines the behavior of the intensity $I(\omega)$? In order to provide the answer to this question, the Raman spectra of some glasses have been compared to inelastic neutron spectra.[15,16] On the basis of the measurements made, the value

$$c(\omega) = \frac{I^R(\omega)\omega^2}{g(\omega)} \tag{4}$$

has been calculated. It was found that $c(\omega)$ is a rather smooth monotonically increasing function in the range of the boson peak. According to Refs. 15 and 16 this function can be approximated by a simple linear dependence. It is evident from these results that the Martin-Brenig model is not adequate to describe the Raman spectra of glasses and the boson peak is traced to non-Debye behavior of the vibrational state density.

However, the nature of such frequency dependence of $c(\omega)$ has not been so far understood. The abovementioned calculations within the soft potential model[19] gave $c(\omega)$ =const. Several models based on the Shuker-Gammon formula (3) have been suggested to explain this dependence;[18] however, some very specific *ad hoc* assumptions concerning glass structure have been made in these papers. From our viewpoint, the frequency dependence of $c(\omega)$ defined by Eq. (4) should naturally result from the theory, taking into account the scattering of light from both kinds of vibrational excitations of glasses and the interaction between them. To develop such a theory in the framework of the standard soft potential model is the purpose of this paper.

In so doing we derive an expression for intensity $I^R(\omega)$, which in the general case cannot be reduced to the Shuker-Gammon expression (3). It follows from this result that the value $c(\omega)$ cannot have the direct physical meaning of an effective coupling parameter of light with any mode. We find that under some conditions its frequency dependence has a form close to a linear function. It will be shown that such behavior results from the phonon contribution to scattering.

II. CALCULATION OF THE VIBRATIONAL GREEN'S FUNCTION

It is convenient to start with stating the model in the framework of which the dynamics of glasses will be considered. The appropriate Hamiltonian within the harmonic approximation can be written down as

$$H = \sum_{\mathbf{r}} \frac{M\dot{\mathbf{u}}^2(\mathbf{r})}{2} + \sum_{\mathbf{r}_1,\mathbf{r}_2} D_{ij}(\mathbf{r}_1-\mathbf{r}_2)u_i(\mathbf{r}_1)u_j(\mathbf{r}_2) + \sum_{\mathbf{r}_1,\mathbf{r}_2} U_{ij}(\mathbf{r}_1,\mathbf{r}_2)u_i(\mathbf{r}_1)u_j(\mathbf{r}_2), \tag{5}$$

where the first summand is the kinetic energy of the atoms, the second one describes the part of the interaction between atoms corresponding to an ideal lattice, and the last summand takes into account local fluctuations of the force parameters. These fluctuations can be divided into two parts: small smooth inhomogeneities and strong deviations from ideal values, which are rare in occurrence. The first type gives rise to nonresonant Rayleigh scattering, while the second is responsible for the appearance of resonant quasilocal states. According to the soft potential model, just the latter kind of glass inhomogeneities gives the main contribution to the properties of glasses under consideration. Therefore we consider the last summand in (5) as describing only this part of the inhomogeneities. These strong fluctuations are localized around a small number of sites (we designate these sites by the letter l) and hence they can be described by means of some local pseudopotentials. In so doing the fluctuation part of the Hamiltonian (5) can be replaced by the sum of local pseudopotentials $U_{ij}(\boldsymbol{l},\varepsilon)$, where $\varepsilon = \omega^2$:

$$U_{ij}(\mathbf{r}_1,\mathbf{r}_2) = \sum_{l} U_{ij}(\boldsymbol{l},\varepsilon)\delta_{l,\mathbf{r}_1}\delta_{l,\mathbf{r}_2}, \tag{6}$$

where $\boldsymbol{l}$ is associated with the defect sites. This approach to describing glass dynamics was suggested in Ref. 20. It can be shown that for defects corresponding to the soft potential model such a reduction can be carried out explicitly to give the pseudopotential of the form[21]

$$U(l,\varepsilon) = \frac{\varepsilon\varepsilon_l}{\varepsilon - \varepsilon_l},$$

where ε_l is the square of the frequency of the atom's vibrations in the soft potential without taking into account its interaction with other atoms. However, for further consideration we will not need any particular form of the frequency dependence of the pseudopotential.

The Green's function $G_{ij}(\mathbf{r}_1,\mathbf{r}_2)$ corresponding to the Hamiltonian (5), taking into account the relation (6), can be expressed in terms of a scattering matrix $T_{ij}(\boldsymbol{l}_1,\boldsymbol{l}_2)$ in the standard way:

$$G_{ij}(\mathbf{r}_1,\mathbf{r}_2) = G^{(0)}_{ij}(\mathbf{r}_1 - \mathbf{r}_2) + \sum_{l_1,l_2} G^{(0)}_{ik}(\mathbf{r}_1 - \boldsymbol{l}_1) \times T_{km}(\boldsymbol{l}_1,\boldsymbol{l}_2)G^{(0)}_{mj}(\boldsymbol{l}_2 - \mathbf{r}_2), \tag{7}$$

where $G^{(0)}_{ij}(\mathbf{r}_1 - \mathbf{r}_2)$ is the initial Green's function of an ideal lattice and we sum over sites occupied by the defects. The T matrix is defined by the equation

$$T_{ij}(\boldsymbol{l}_1,\boldsymbol{l}_2) = U_{ij}(\boldsymbol{l}_1)\delta_{l_1,l_2} + \sum_{l} U_{ik}(\boldsymbol{l}_1)G^{(0)}_{km}(\boldsymbol{l}_1 - \boldsymbol{l})T_{mj}(\boldsymbol{l},\boldsymbol{l}_2). \tag{8}$$

According to the soft potential model strong softening of the force parameters at a given point is most likely only for displacements in some single direction.[8,9] In other words, this means that the local fluctuations of the force parameters that are responsible for quasilocal vibrations are rather anisotropic. The isotropic part of these fluctuations within this model of glass dynamics is considered to be quite small, as has been explained before, and may be neglected for the problems under consideration. Taking into account this remark, let us write down the site pseudopotential $U_{ij}(\boldsymbol{l},\varepsilon)$ in the form

$$U_{ij}(\boldsymbol{l},\varepsilon) = U(\boldsymbol{l},\varepsilon)v_i(\boldsymbol{l})v_j(\boldsymbol{l}), \tag{9}$$

where the unit vector $\mathbf{v}$ prescribes nonzero components of the pseudopotential at the given site $\boldsymbol{l}$. In essence, the vector $\mathbf{v}$ sets the direction of the eigenvector of the soft quasilocal vibration at the site $\boldsymbol{l}$. Presenting the pseudopotential in the form (9) distinguishes our consideration from that of Ref. 20, where $U_{ij}(\boldsymbol{l})$ was regarded as an isotropic tensor. This difference is of no importance for the calculations of the density of states, but is very essential if Raman scattering is considered. By this means statistical properties of the pseudopotential in our consideration are determined by the fluctuations of two parameters: the value of the pseudopotential $U(\boldsymbol{l})$ and the directions of the local axes of the anisotropy $\mathbf{v}$. These fluctuations are regarded as independent of each other. This assumption looks rather natural since the correlator of the form $\langle Uv_i\rangle$ should vanish due to the macroscopic isotropy of glasses.

One can considerably simplify the treatment of Eq. (8) by neglecting the difference between transversal and longitudinal initial phonons (the so called scalar model). In such a case the initial Green's function has the form $G^0(\mathbf{r}_1 - \mathbf{r}_2)\delta_{ij}$. This assumption does not allow us to describe in detail the behavior of the depolarization ratio of the scattering light, but we believe that it does not influence noticeably the frequency dependence of the scattering intensity.

Separating out of the sum in Eq. (8) the term with $\boldsymbol{l} = \boldsymbol{l}_1$ one can rewrite this equation in the form

$$T_{ij}(\boldsymbol{l}_1,\boldsymbol{l}_2) = S_{ik}(\boldsymbol{l}_1)U_{kj}(\boldsymbol{l}_1)\delta_{l_1,l_2} + \sum_{l\neq l_1} S_{im}(\boldsymbol{l}_1)U_{mk}(\boldsymbol{l}_1)G^0(\boldsymbol{l}_1 - \boldsymbol{l})T_{kj}(\boldsymbol{l},\boldsymbol{l}_2), \tag{10}$$

where the matrix $S_{ik}(\boldsymbol{l})$ is defined as

$$S_{ik}(\boldsymbol{l}) = [\delta_{ik} - P(\varepsilon)U_{ik}(\boldsymbol{l})]^{-1} \tag{11}$$

and $P(\varepsilon) = G^0(0)$. Taking into consideration the definition of the pseudopotential (9), one can calculate the product of the matrices $S_{ik}U_{kj}$ entering into Eq. (10) to obtain

$$S_{ik}(\boldsymbol{l})U_{kj}(\boldsymbol{l}) = t_l\rho_{ij}(\boldsymbol{l}), \tag{12}$$

where $\rho_{ij} = v_iv_j$ and

$$t_l = \frac{U_l}{1 - U_l}P(\varepsilon). \tag{13}$$

The value $t_l\rho_{ij}(\boldsymbol{l})$ determines the scattering matrix, if one neglects the interaction between defects, described by the second term in Eq. (10). The pole of t_l gives the frequency and the linewidth of a single quasilocal vibration.

We seek a solution of Eq. (10) in the form

$$T_{ij}(\boldsymbol{l}_1,\boldsymbol{l}_2) = \tilde{T}(\boldsymbol{l}_1,\boldsymbol{l}_2)t_{l_1}\rho_{ij}(\boldsymbol{l}_1). \tag{14}$$

With Eq. (12) one can find the equation for $\tilde{T}$ to look as follows:

$$\tilde{T}(\boldsymbol{l}_1,\boldsymbol{l}_2)\rho_{ij}(\boldsymbol{l}_1) = \delta_{l_1,l_2}\rho_{ij}(\boldsymbol{l}_1) + \rho_{ik}(\boldsymbol{l}_1)\sum_{l}\rho_{kj}(\boldsymbol{l})t_l \times G^0(\boldsymbol{l}_1 - \boldsymbol{l})\tilde{T}(\boldsymbol{l},\boldsymbol{l}_2). \tag{15}$$

It has been argued in Ref. 20 that the value $\rho_{ij}(\boldsymbol{l})t_l$ appearing in the sum over $\boldsymbol{l}$ in Eq. (15) may be regarded under certain conditions as a self-averaging one. In such a case one can replace it by the averaged values

$$\langle\rho_{kj}(\boldsymbol{l})t_l\rangle = \frac{1}{3}\delta_{kj}\langle t\rangle .$$

The average value of ρ_{ij} is found by taking into account that any direction of the vector $\mathbf{v}$ has equal probability. The average value $\langle t\rangle$ is calculated with a distribution function $f(U)$:

$$\langle t\rangle = \int \frac{Uf(U)}{1-UP(\varepsilon)}dU. \tag{16}$$

Then the equation for $\tilde{T}(\boldsymbol{l}_1,\boldsymbol{l}_2)$ can be solved to give

$$\tilde{T}(\boldsymbol{l}_1,\boldsymbol{l}_2) = \frac{1}{N}\sum_{\mathbf{k}} \frac{e^{i\mathbf{k}(\boldsymbol{l}_1-\boldsymbol{l}_2)}}{1-\frac{1}{3}\langle t\rangle\,\hat{G}^0(\mathbf{k})}, \tag{17}$$

where N is the number of sites, and $\hat{G}^0(\mathbf{k})$ is the Fourier transform of the initial Green's function. We obtain the final expression for the Green's function $G(\mathbf{r}_1,\mathbf{r}_2)$ as

$$\begin{aligned} G_{ij}(\mathbf{r}_1,\mathbf{r}_2) = {} & G^0(\mathbf{r}_1-\mathbf{r}_2)\delta_{ij} \\ & +\sum_{\boldsymbol{l}_1,\boldsymbol{l}_2} t_{\boldsymbol{l}_1}\rho_{ij}(\boldsymbol{l}_1)G^0(\mathbf{r}_1-\boldsymbol{l}_1) \\ & \times\tilde{T}(\boldsymbol{l}_1-\boldsymbol{l}_2)G^0(\boldsymbol{l}_2-\mathbf{r}_2). \end{aligned} \tag{18}$$

The vibrational state density corresponding to this Green's function can be written down in the form

$$g(\varepsilon) = \frac{1}{\pi}\left[\mathrm{Im}P(\varepsilon-i\varepsilon\xi) - \frac{\pi}{3}\varepsilon_D n_0(\varepsilon)\mathrm{Re}P(\varepsilon-i\varepsilon\xi)\right], \tag{19}$$

which coincides with the corresponding expression of Ref. 20. ReP and ImP symbolize taking the real and imaginary parts of P. In Eq. (19) we have used the following designations:

$$\begin{aligned} P(\varepsilon-i\varepsilon\xi) &= \frac{1}{N}\sum_{\mathbf{k}}\frac{1}{\varepsilon-\varepsilon_{\mathbf{k}}-i\varepsilon\xi}, \\ \xi &= \frac{n_0(\varepsilon)\Gamma(\varepsilon)}{\varepsilon g_D(\varepsilon)}. \end{aligned} \tag{20}$$

The values $n_0(\varepsilon)$ and $\Gamma(\varepsilon)$ are the initial state density of the noninteracting quasilocal vibrations and their linewidth, respectively, $\varepsilon_{\mathbf{k}}$ is the initial phonon dispersion law $\varepsilon_{\mathbf{k}} = v^2k^2$, and ε_D is the square of the Debye frequency. In deducing expressions (19) and (20) we used the method of calculating the averages over the magnitude of the pseudopotential suggested in Ref. 20. As was shown in the same Ref. 20 the averaged t-matrix approximation used to derive Eqs. (18)–(20) is good if

$$\frac{\varepsilon}{\varepsilon_D}\xi \ll 1. \tag{21}$$

Since $\varepsilon \ll \varepsilon_D$ in the actual frequency range for this problem, the condition (21) allows us to consider not only the case when $\xi \ll 1$, but the case when $\xi \geq 1$ as well.

The values $n_0(\varepsilon)$ and $\Gamma(\varepsilon)$ in this approach are regarded as knowns. They should be determined separately from the consideration of the noninteracting quasilocal vibrations. For the width of a single quasilocal state one can use a well-known expression (see, for example, Ref. 21)

$$\Gamma(\varepsilon) \approx \pi\varepsilon^2 g_D(\varepsilon).$$

In so doing the parameter ξ takes the form

$$\xi = \pi\varepsilon n_0(\varepsilon). \tag{22}$$

Calculating the initial state density $n_0(\varepsilon)$ in the framework of the soft potential model leads to the expression as follows:[22]

$$n_0(\varepsilon) = \frac{N_s}{8}\frac{\varepsilon^{3/2}}{w}\int_0^1 \exp\left[-\left(\frac{x}{8\delta}\right)^2\frac{\varepsilon^3}{w^6}\right]dx \tag{23}$$

(the derivation of these expression is given in Appendix A). Here N_s is the number of defects per atom and w is the characteristic energy of the movement in the soft potential, associated with crossover from two-level systems to the almost harmonic soft vibrations. The integral in Eq. (22) takes into account, according to Ref. 22, the cutoff of the distribution of the soft potential parameters. The value $\delta \sim (k_BT_g/\hbar w)^{3/4}$,[23] where k_B is the Boltzmann constant and T_g is the glass transition temperature, sets the scale of this cutoff.

The density of states (19), calculated taking into account Eqs. (22) and (23) reproduces rather well the main peculiarities of the vibrational state density known from neutron experiments[5] and numerical calculations:[22] the position of the maximum ω_m, the ω^4 law at small frequencies, and the linear dependence at $\omega > \omega_m$.

III. CALCULATION OF SCATTERING INTENSITY

In this section we make use of the Green's function calculated above to derive the frequency dependence of the scattering intensity $I(\omega)$. We start, following Ref. 24, from the general expression for the scattering intensity,

$$I(\omega) \sim \sum_{\alpha\beta\gamma\delta} m_\alpha n_\beta m_\gamma n_\delta \langle P_{\alpha\beta}(t)P_{\gamma\delta}(0)\rangle_\omega, \tag{24}$$

where unit vectors $\mathbf{n}$ and $\mathbf{m}$ describe the polarization of the incident and scattering light, ω is the change in the light frequency due to scattering, and

$$K_{\alpha\beta\gamma\delta} = \langle P_{\alpha\beta}(t)P_{\gamma\delta}(0)\rangle_\omega = \int dt e^{i\omega t}\langle P_{\alpha\beta}(t)P_{\gamma\delta}(0)\rangle \tag{25}$$

is the Fourier transform of the correlation function of the electronic polarizability tensor $P_{\alpha\beta}$. In Eq. (24) we omit factors which are independent of ω. The angle brackets in (24) indicate both thermodynamics and disorder averaging. The polarizability tensor $P_{\alpha\beta}$ depends on atom displacements $\mathbf{u}(\mathbf{r})$ and can be written as a power series in $\mathbf{u}$:

$$P_{\alpha\beta}(t) = P^0_{\alpha\beta} + \sum_{\mathbf{r}}\frac{\partial P_{\alpha\beta}}{\partial u_i}u_i(\mathbf{r}) + \cdots. \tag{26}$$

The first term of this series describes Rayleigh scattering; the second is responsible for the first order Raman scattering. The values

$$L_{\alpha\beta,i} = \left.\frac{\partial P_{\alpha\beta}}{\partial u_i}\right|_{u_i=0} \tag{27}$$

are coupling parameters between the light and the displacements of an atom located at site $\mathbf{r}$. It should be noticed that these parameters taken at ideal sites and at defect sites are rather different in their properties. The first of them, which we designate as $L^{\mathrm{id}}_{\alpha\beta,i}$, are caused by those small fluctuations of glass structure which have been neglected while the Green's function was calculated and hence they do not correlate with the parameters of the pseudopotentials. The coupling parameters at defect sites ($L^{def}_{\alpha\beta,i}$) are determined by the strong distortions of glass structure which cause the quasilocal vibrations. Since fluctuations of $L^{\mathrm{id}}_{\alpha\beta,i}$ and $L^{def}_{\alpha\beta,i}$ are determined by different kinds of glass inhomogeneities it is natural to suggest that they do not correlate with each other. It should be noted also that the coupling parameter $L_{\alpha\beta,i}$ satisfies a rather general condition[25]

$$\sum_{\mathbf{r}} L_{\alpha\beta,i}(\mathbf{r}) = 0, \tag{28}$$

where the sum over all kinds of sites is included. This condition provides for the modulation of the polarization to be equal to zero if the system is moved as a whole.

Since the disorder in the system is regarded as frozen one can carry out thermodynamics and disorder averaging independently. Resolving the displacements $\mathbf{u}(\mathbf{r},t)$ into normal coordinates and finding by the standard way the thermodynamic average, we obtain the Stokes part of the correlator (25) as follows:

$$K_{\alpha\beta\gamma\delta} \sim [1+n(\omega)] \sum_{\mathbf{r}_1,\mathbf{r}_2} \Big\langle L_{\alpha\beta,i}(\mathbf{r}_1) L_{\gamma\delta,j}(\mathbf{r}_2) \times \sum_s e^s_i(\mathbf{r}_1) e^s_j(\mathbf{r}_2) \delta(\omega^2-\omega_s^2) \Big\rangle_{\mathrm{dis}}. \tag{29}$$

Here $n(\omega)$ is the Bose distribution function, $\mathbf{e}^s(\mathbf{r})$ is an eigenvector of the sth normal mode, and ω_s is its associated eigenfrequency. The angle brackets $\langle\cdots\rangle_{\mathrm{dis}}$ designate now averaging over all kinds of disorder present in the system. The summation in (29) is over all eigenmodes and all sites.

The sum over s in (29) can be expressed in terms of the Green's function according to the relation

$$\sum_s e^s_i(\mathbf{r}_1) e^s_j(\mathbf{r}_2) \delta(\omega^2-\omega_s^2) = \mathrm{Im}\left[\delta_{ik} - \frac{dU_{ik}(\mathbf{r}_1,\varepsilon)}{d\varepsilon}\right] G_{kj}(\mathbf{r}_1,\mathbf{r}_2), \tag{30}$$

which is established in Appendix B. The term $dU_{ik}/d\varepsilon$ takes into account the pseudopotential nature of the value $U_{ik}(\mathbf{r},\varepsilon)$. With this relation one can write the correlator (29) as

$$\frac{K_{\alpha\beta\gamma\delta}}{1+n(\omega)} \sim K^{(1)}_{\alpha\beta\gamma\delta} + K^{(2)}_{\alpha\beta\gamma\delta}, \tag{31}$$

$$K^{(1)}_{\alpha\beta\gamma\delta} = \sum_{\mathbf{r}_1,\mathbf{r}_2} \langle L_{\alpha\beta,i}(\mathbf{r}_1) L_{\gamma\delta,j}(\mathbf{r}_2) G_{ij}(\mathbf{r}_1,\mathbf{r}_2)\rangle_{\mathrm{dis}}, \tag{32}$$

$$K^{(2)}_{\alpha\beta\gamma\delta} = -\sum_{\mathbf{r}_1,\mathbf{r}_2} \Big\langle L_{\alpha\beta,i}(\mathbf{r}_1) L_{\gamma\delta,j}(\mathbf{r}_2) \times \frac{dU_{ik}(\mathbf{r}_1)}{d\varepsilon} G_{kj}(\mathbf{r}_1,\mathbf{r}_2) \Big\rangle_{\mathrm{dis}}. \tag{33}$$

The calculation of the averages in (32) and (33) implies a triple averaging: over the magnitude of the pseudopotential $U_l(\varepsilon)$, over directions of the vectors $\mathbf{v}$, and over distribution of the coupling parameters $L_{\alpha\beta,i}(\mathbf{r})$. Substituting Eq. (18) for the Green's function into (32), there appear correlators of the form

$$\langle L_{\alpha\beta,i}(\mathbf{r}_1) L_{\gamma\delta,j}(\mathbf{r}_2) t_l \rho_{ij}(\boldsymbol{l})\rangle_{\mathrm{dis}}.$$

Making use of the above-discussed statistical properties of the coupling parameters one can conclude that this expression is different from zero only if both r_1 and r_2 are ideal or defect sites, and it can be written down as

$$\begin{aligned}\langle L_{\alpha\beta,i}(\mathbf{r}_1) L_{\gamma\delta,j}(\mathbf{r}_2) \rho_{ij}(\boldsymbol{l}) t_l\rangle_{\mathrm{dis}} &= \delta_{\mathbf{r}_1\mathbf{r}_2}\delta_{\mathbf{r}_1 l} \big\langle L^{\mathrm{def}}_{\alpha\beta,i}(\boldsymbol{l}) L^{\mathrm{def}}_{\gamma\delta,j}(\boldsymbol{l}) \rho_{ij}(\boldsymbol{l}) t_l \big\rangle_{\mathrm{dis}} \\ &\quad + \frac{1}{3}(1-\delta_{\mathbf{r}_1 l})(1-\delta_{\mathbf{r}_2 l}) \\ &\quad \times \big\langle L^{\mathrm{id}}_{\alpha\beta,i}(\mathbf{r}_1) L^{\mathrm{id}}_{\gamma\delta,j}(\mathbf{r}_2)\big\rangle_{\mathrm{dis}} \langle t_l\rangle. \end{aligned} \tag{34}$$

The second summand in the right side of this expression describes the interaction of light with the atoms located in the sites corresponding to the ideal lattice, while the first one is related to the interaction with atoms oscillating in the soft potentials. In this term we factor out the term $\langle t_l\rangle$ from the angle brackets, taking into account the averaged t-matrix approximation, used in calculations of the Green's function. The expressions $L^{\mathrm{def}}_{\alpha\beta,i}$ involved in this term can be written as $\partial P_{\alpha\beta}/\partial u_v$, where u_v is the atom displacement along $\mathbf{v}$ and hence describes the modulation of the polarization due to quasilocal vibrations. Let us designate

$$\big\langle L^{\mathrm{def}}_{\alpha\beta,i}(\boldsymbol{l}) L^{\mathrm{def}}_{\gamma\delta,j}(\boldsymbol{l}) \rho_{ij}(\boldsymbol{l})\big\rangle_{\mathrm{dis}} \equiv \frac{1}{3} D_{\alpha\beta,\gamma\delta} \tag{35}$$

and consider the tensor $D_{\alpha\beta,\gamma\delta}$ as an isotropic fourth-rank tensor, components of which are phenomenological parameters and have to be determined from experiment. We would like to note that this tensor corresponds to the tensor $\overline{\alpha_{ik}\alpha_{lm}}$ from Ref. 19. The correlator $\langle L^{\mathrm{id}}_{\alpha\beta,i}(\mathbf{r}_1) L^{\mathrm{id}}_{\gamma\delta,j}(\mathbf{r}_2)\rangle$ is presented in the form

$$\big\langle L^{\mathrm{id}}_{\alpha\beta,i}(\mathbf{r}_1) L^{\mathrm{id}}_{\gamma\delta,j}(\mathbf{r}_2)\big\rangle_{\mathrm{dis}} = B_{\alpha\beta,\gamma\delta} F(\mathbf{r}_1-\mathbf{r}_2), \tag{36}$$

where $B_{\alpha\beta,\gamma\delta}$ also defines the set of phenomenological parameters for light coupling with nondefect atoms. The correlation function $F(\mathbf{r}_1-\mathbf{r}_2)$ describes the correlation properties of the fluctuations of values $L^{\mathrm{id}}_{\alpha\beta,i}$. This evidently obeys a condition similar to Eq. (28):

$$\sum_{\mathbf{r}} F(\mathbf{r}) = 0. \tag{37}$$

Calculating $K^{(2)}_{\alpha\beta\gamma\delta}$ in the same way we obtain the eventual expression for the reduced intensity:

$$I^R \sim \frac{1}{\omega}\left[B\,\mathrm{Im}\frac{1}{N}\sum_{\mathbf{k}} \frac{S(k)}{\varepsilon-\varepsilon_k-i\varepsilon\xi} - D\frac{\pi}{3}\varepsilon_D n_0(\varepsilon)\,\mathrm{Re}P(\varepsilon-i\varepsilon\xi)\right], \tag{38}$$

where B and D stand for convolutions of the form $B_{\alpha\beta\gamma\delta}m_\alpha n_\beta m_\gamma n_\delta$ and $D_{\alpha\beta\gamma\delta}m_\alpha n_\beta m_\gamma n_\delta$, respectively. The function $S(k)$ is the Fourier transform of the correlation function (36) and satisfies the condition $S(0) = 0$, which follows from Eq. (37). If the correlation function decreases rather fast with increasing r, one can obtain that $S(k) \sim k^2$ at $k \ll r_c^{-1}$, where r_c is a characteristic size (correlation radius) of the inhomogeneities of $L^{\mathrm{id}}_{\alpha\beta,i}$. Taking into account this remark one can see that the first summand in Eq. (38) leads to the Martin-Brenig result (1) if $\xi = 0$, while the second one in the same case is in correspondence with the result of Ref. 19, if $n_0(\varepsilon)$ is regarded as the density of states of the quasilocal vibrations including the quasistatic interaction between them.

The general conclusion following from Eq. (38) is that this expression is not reduced to the Shuker-Gammon form (3). Such a reduction is possible only in two cases: if $\xi = 0$ or if $S(k)$=const and $B = D$. In the first case one has a sum of two terms of the form (3), one of which corresponds to phonons and the second is related to the quasilocal vibrations which are independent of them. In the second case we obtain a single term with $c(\omega)$ =const. It follows from this result that the function $c(\omega)$ defined by Eq. (4) does not have the direct sense of the effective coupling parameter of light with any vibrations.

A further analysis calls for a specification of the function $S(k)$. Keeping in mind the property $S(k) \sim k^2$ at small k, one can consider functions of two kinds, which are distinguished in the behavior at $k \gg r_c^{-1}$. The first of them (curve 1 in Fig. 1) goes down at large k and is in qualitative correspondence with the function of the Martin-Brenig model; the second tends to 1 at $k \gg r_c^{-1}$. Such function was used by Schlomann[26] for describing far infrared experiments in glasses. At $\omega \ll \omega_c = v r_c^{-1}$ both these functions are similar and from them one can obtain the approximation for I^R in this frequency range:

$$I^R \sim \frac{\omega^2}{\omega_c^2 \omega_D}\left[B + D\frac{N_s}{24}\left(\frac{\omega_D}{w}\right)^3 \frac{\omega_c^2}{w^2}\right], \qquad (39)$$

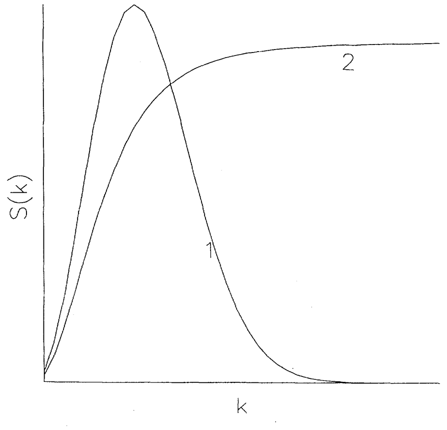

FIG. 1. Two kinds of spectral density $S(k)$ used in order to describe the correlation properties of the inhomogeneities of the elasto-optic parameters $L_{ij,k}$. Curve 1 presents the Martin-Brenig function and curve 2 the Schlomann function.

where expression (23) for $n_0(\varepsilon)$ has been used. In so doing we suggest that in the considered frequency range the integral in (23) can be omitted. It is seen that in such a case both summands in (38) lead to identical frequency dependencies ω^2 of I^R. The relationship of these contributions to each other is determined mainly by the ratio ω_c/w. If we want expression (39) to actually describe the scattering intensity in the frequency region of the boson peak, the correlation frequency ω_c must be larger than 20 cm^{-1} at least. Using the parameters N_s, ω_D, and w for a-SiO$_2$,[22] one can find that in this case the coefficient before D in (39) is larger than approximately 4. We will see later that it is reasonable to consider the magnitude of D to be less than B and hence we cannot be certain which term in Eq. (33) gives the more significant contribution. Additional experimental investigations of spectra are required to tackle this problem.

At frequencies $\omega > \omega_c$ the behavior of the intensity depends essentially on the form of the function $S(k)$. Inserting in (33) the function displayed by curve 1 in Fig.1, one finds the function $c(\omega)$ [Eq. (4)] to have a maximum, in contrast to the experimental results.[15–17] Therefore it seems more reasonable to use Schlomann's function (curve 2 in Fig. 2) to model the correlation properties of the elasto-optic parameters $L_{\alpha\beta,\gamma}$. In this case one can put $S(k) = 1$ if the frequencies $\omega > \omega_c$ are considered. Then Eq. (38) reads

$$I^R \sim \frac{1}{\omega}\left[B\,\mathrm{Im}P(\varepsilon - i\varepsilon\xi) - D\frac{\pi}{3}\varepsilon_D n_0(\varepsilon)\mathrm{Re}P(\varepsilon - i\varepsilon\xi)\right]. \qquad (40)$$

If ω_c is not too large so that at $\omega > \omega_c$ ξ is still considerably less than 1, an interesting situation, when $I^R(\omega)$ contains a term independent of frequency, becomes possible:

$$I^R \sim B + \frac{1}{2}DN_s\left(\frac{\omega_D}{w}\right)^3\left(\frac{\omega}{w}\right)^2.$$

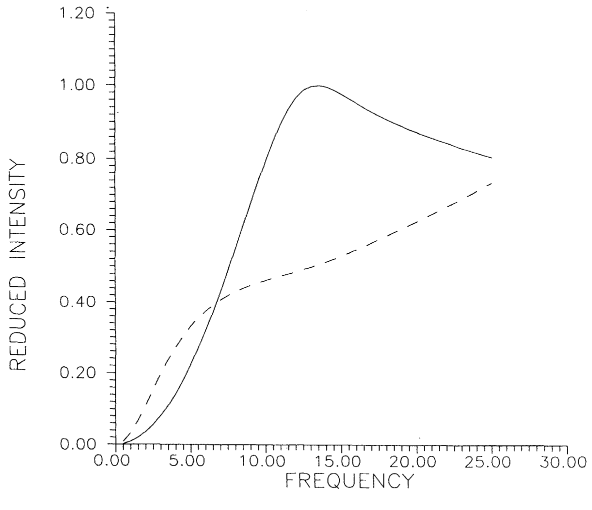

FIG. 2. The reduced intensity $I^R(\omega)$ (solid line) and the function $c(\omega)$ (dashed line). The frequency is scaled by the parameter w.

This term is expected to be usually masked by the excess scattering. However, since its contribution does not depend on temperature, it could be of interest to attempt to separate it out by means of investigation of the Raman spectra at very low temperatures.

At frequencies high enough to admit $\xi \gg 1$, Eq. (40) can be transformed to

$$I^R \sim B\omega + D\sqrt{\frac{2N_s\delta}{\pi}}\,\frac{\omega_D^3}{w}\,\frac{1}{\omega}. \tag{41}$$

In so doing we make use of the following expression for $\mathrm{Im}P(\varepsilon - i\varepsilon\xi)$:

$$\mathrm{Im}P(\varepsilon - i\varepsilon\xi) = \frac{3}{2}\pi\frac{\omega}{\omega_D^3}(1+\xi^2)^{1/4}\cos\varphi,$$

where $\varphi = \arctan\xi/2$, and take into account the cutoff due to the integral in Eq. (23) for $n_0(\varepsilon)$. The state density normalized by the square of the frequency, $g(\omega)/\omega^2$, can also be described by Eq. (41) if one puts $B = D = 1$. The linear term in the corresponding expression is always small in comparison to the second term in the frequency region under consideration. Hence the state density at these frequencies is described by the law $1/\omega$ associated with the contribution of the quasilocal vibrations.[19,22] However, the influence of the linear term on the scattering intensity can be more considerable if one admits that $B > D$. In this case this term could be responsible for the deviation of the frequency dependence of I^R from the law $1/\omega$. In order to make the latter remark clear, let us deduce the expression for the function $c(\omega)$ [(4)]:

$$c(\omega) \sim B + 3(B-D)\left(\frac{\omega}{\omega_D}\right)^3 \xi^{-1}(1+\xi^2)^{1/4}\cos\varphi. \tag{42}$$

If one assumes that in the region of the boson peak the cutoff of $n_0(\varepsilon)$ is already efficient to give

$$\xi \sim N_s\delta\left(\frac{\omega}{w}\right)^2,$$

then $c(\omega)$ actually displays close to linear behavior, consistent with the experiments if $B > D$. In Fig. 2 we present the functions $I^R(\omega)$ and $c(\omega)$ obtained by means of computer calculations of the expressions (38) and (19). In so doing we use the values P_s, δ, and w for SiO$_2$. One can see that $c(\omega)$ actually demonstrates quasilinear behavior in a wide region around the boson peak.

IV. CONCLUSION

Let us summarize the results obtained in this paper. We develop the phenomenological theory of the first-order Raman scattering in glasses. Unlike previous considerations[12,18,19] we do not assume that some particular vibration mode gives the major contribution to scattering and take into account within a unified approach the interaction of light with the vibrations of atoms in the defect soft potentials as well as with atoms oscillating in the regular rigid potentials corresponding to an ideal lattice. The dynamics of the atoms is described within the soft potential model.[8–11]

The important theoretical result of our consideration lies in the fact that the expression for scattering intensity is not generally reduced to the Shuker-Gammon form (3). From this it follows that the function $c(\omega)$ has not the direct meaning of the coupling parameter of light with any kind of vibrations. We show that in such a case the frequency dependence of $c(\omega)$ is determined by the relative contributions of various vibration modes to the scattering. The close to linear dependence of $c(\omega)$ on frequency observed in a number of experiments[15–17] follows from the standard soft potential model under the assumption that the coupling parameter of light with soft quasilocal vibrations is less than a similar parameter for the atoms placed in regular sites corresponding to an ideal lattice.

The expressions (39)–(42) obtained give a principal opportunity to determine these parameters (B and D) by means of special treatment of experimental data. For example, expression (42) for $c(\omega)$ does not contain any unknown parameters except B and D (the parameters of the soft potential model can be found from other experiments[11] and they are known for a number of glasses). Therefore by forcing the experimental data to fit Eq. (42) one can determine both values B and D. These data can be used for calculation of the reduced intensity according to Eqs. (39) and (40), which can be compared with corresponding experments. Such kind of investigations could serve as a test of the theory suggested.

It follows from our considerations that scattering by harmonic oscillations can under some conditions give a frequency- and temperature-independent contribution to the region of the excess scattering. This contribution can be considerable at very low temperatures.

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APPENDIX A

In this Appendix we deduce Eq. (23) for the initial state density of the quasilocal vibrations $n_0(\varepsilon)$. The soft potential can be written down in two forms:[10]

$$V(x) = W\left[\eta\left(\frac{x}{d}\right)^2 + t\left(\frac{x}{d}\right)^3 + \left(\frac{x}{d}\right)^4\right] \tag{A1}$$

or

$$V(x) = W\left[D_1\left(\frac{x}{d}\right) + D_2\left(\frac{x}{d}\right)^2 + \left(\frac{x}{d}\right)^4\right]. \tag{A2}$$

The difference in these forms is that in the first one the potential is expanded as a power series in the atom displacements around the equilibrium position of the mode located at a given point, while the second form corresponds to the expansion around some arbitrary point, which can be chosen the same for all soft modes [the third order term, omitted in (A.2), is not important for further consideration[10]]. Therefore to describe statistical properties of the potential one should set a distribution function for the parameters D_1 and D_2 rather than for η and t. For a given mode the sets of parameters D_1, D_2 and η, t are connected by the evident relations

$$D_1 = -\frac{1}{2}\eta t + \frac{1}{8}t^3,$$

$$D_2 = \eta - \frac{3}{8}t^2.$$

Due to this the distribution function of the parameters η, t is expressed in terms of the (D_1, D_2) distribution function:[10]

$$P(\eta,t) = \frac{1}{2}|\eta| P(D_1, D_2). \tag{A3}$$

According to Refs. 22 and 23 the function $P(D_1, D_2)$ is given as follows:

$$P(D_1, D_2) = N_s \exp\left[-\left(\frac{D_1}{\delta}\right)^2\right], \tag{A4}$$

where $\delta^{-2} = 0.169(W/k_BT_g)^{3/2}$.[23] Taking into account that the soft single-well potentials are realized if $\eta > t^2/4$, one obtains the state density as

$$n_0(\omega) = \int_0^\infty \int_0^{2\eta^{1/2}} d\eta dt P(\eta,t)\delta\left[\omega - \left(\frac{2W\eta}{Md^2}\right)^{1/2}\right].$$

Making use of (A3) and (A4) one derives the eventual expression for $n_0(\varepsilon) = n_0(\omega)/(2\varepsilon^{1/2})$:

$$n_0(\varepsilon) = \frac{N_s}{8}\frac{\varepsilon^{3/2}}{w^5}\int_0^1 \exp\left[-\left(\frac{x}{8\delta}\right)^2\frac{\varepsilon^3}{w^6}\right] dx,$$

where $w = W/\hbar$.

APPENDIX B

Formula (30) is established in this section. In so doing we use the method presented in Ref. 21 where the general expression for the state density of systems with pseudopotentials was deduced.

Let us consider a linear equation of the form

$$\hat{L}(\lambda)\phi - \lambda\phi = 0, \tag{B1}$$

where $\hat{L}(\lambda)$ is an arbitrary linear operator depending on the eigenvalue λ. The Green's operator (resolvent) of this equation is

$$\hat{G}(\lambda) = \left[\lambda - \hat{L}(\lambda)\right]^{-1}. \tag{B2}$$

Let ϕ_s be an eigenfunction and α_s an eigenvalue (for a given λ)

$$\hat{L}(\lambda)\phi_s = \alpha_s(\lambda)\phi_s. \tag{B3}$$

Then the eigenvalues λ_s are determined by the self-consistent equation

$$\lambda_s = \alpha_s(\lambda). \tag{B4}$$

Let us consider the resolvent as a function of λ:

$$\begin{aligned}\hat{G}(\lambda - i\gamma) &= \left[\lambda - \hat{L}(\lambda - i\gamma) - i\gamma\right]^{-1} \\ &= \left[1 - \frac{d\hat{L}}{d\lambda}\right]^{-1} \\ &\quad\times\left[\left[\lambda - \hat{L}(\lambda)\right]\left(1 - \frac{d\hat{L}}{d\lambda}\right)^{-1} - i\gamma\right]^{-1},\end{aligned} \tag{B5}$$

where $\gamma \to 0$. In order to obtain the Green's function $G(\mathbf{r} - \mathbf{r}')$ in coordinate representation one should act by the resolvent on the delta function $\delta(\mathbf{r} - \mathbf{r}')$. Making use of the completeness condition for the eigenfunction $\phi_s(\mathbf{r})$,

$$\delta(\mathbf{r} - \mathbf{r}') = \sum_s \phi_s(\mathbf{r})\phi_s(\mathbf{r}'),$$

one derives from (B5)

$$\begin{aligned}&\left(1 - \frac{d\hat{L}}{d\lambda}\right) G(\mathbf{r} - \mathbf{r}') \\ &\qquad = \sum_s \phi_s(\mathbf{r})\phi_s(\mathbf{r}')\left[\frac{\lambda - \alpha_s(\lambda)}{1 - \frac{d\alpha_s}{d\lambda}} - i\gamma\right]^{-1}.\end{aligned} \tag{B6}$$

Now we separate the imaginary part of the obtained expression in the case of $\gamma \to 0$, and making use of the identity

$$\delta(\lambda - \lambda_s) \equiv \left(1 - \frac{d\alpha_s}{d\lambda}\right)\delta[\lambda - \alpha(\lambda)]$$

we obtain eventually

$$\sum_s \phi_s(\mathbf{r})\phi_s(\mathbf{r}')\delta(\lambda - \lambda_s) = \text{Im}\left[1 - \frac{d\hat{L}(\lambda)}{d\lambda}\right] G_{\lambda - i\gamma}(\mathbf{r}, \mathbf{r}'). \tag{B7}$$

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