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Far-infrared attenuation in glasses

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The influence of quasilocal vibrations on the far-infrared spectra of glasses is investigated under the supposition that the quasilocal vibrations are directly inactive in the attenuation. It is shown that taking into account phonon-quasilocal-vibration coupling and both medium- and short-range structure of the inhomogeneities of a photon-phonon coupling parameter can give rise to an attenuation curve similar to one observed for a-SiO$_2$ and related materials.

I. INTRODUCTION

It is well known that dielectric (and semiconductor) disordered materials show the absorption of far-infrared radiation to be sufficiently greater than that in ideal crystals. The disorder-induced coupling of light with atom vibrations is generally accepted to be responsible for this.

In earlier experimental papers the absorption was found to satisfy either the law $\Gamma \sim \omega^2$,[1,2] or $\Gamma \sim \omega^4$,[3] in the frequency range $\omega = 20$ to 80 cm^{-1}. These results are described by the expression

$$\Gamma(\omega) = c(\omega) g_D(\omega) , \tag{1}$$

provided that the state density $g(\omega)$ is assumed to have Debye form and the efficient photon-phonon coupling parameter $c(\omega)$ is regarded either as a constant [the model of charge defects (MCD) (Refs. 4 and 5)] or as ω^2 [the model of induced dipoles (MID) (Ref. 6)].

The crossovers from the ω^4 law to the ω^2 law,[1,2,7] or to $\Gamma(\omega) \sim$ const,[7] which were observed in some materials, could be explained in terms of the same formula, taking into account the correlation properties of the spatial distribution of a parameter describing the interaction of light with phonons. In this case $c(\omega)$ becomes proportional to the spectral density $S(k)$ of the inhomogeneities of this parameter with the coefficient of proportionality being equal to 1 or to ω^2 depending on the model of the phonon-photon coupling.[5,8]

However, more precise experiments[9,10] have found the dependences $\Gamma(\omega)$ to be in fact more complicated in this frequency region. It appears, in particular, that the curve Γ/ω^2 has a maximum at frequency $\omega \sim 30$–40 cm^{-1}. This finding is in accord with other anomalous (with respect to the Debye model) properties of glasses. First of all one should mention so-called "boson peak" of Raman scattering and the observation of the excess vibrational state density by means of inelastic neutron scattering. All these facts, in addition, should be correlated with the departure of the temperature dependence of the specific heat from the T^3 law at temperatures ~ 10 K and the plateau on the temperature dependence of the heat conductivity at the same temperatures. A survey of these results can be found in Refs. 11–13. All these peculiarities are in the same frequency or the appropriate temperature regions and it is natural to suspect that they have a common origin.

At present, it is generally accepted that there exist some quasilocal vibrational modes in glasses at angular frequencies 10^{12} Hz which coexist with phonons and it is these modes that are responsible for all these phenomena. A first step to take into account the contribution of quasilocal vibrations (QV's) in themselves to the attenuation of light might be an expression of the form

$$\Gamma_{\mathrm{QV}}(\omega) = c_{\mathrm{QV}}(\omega) g_{\mathrm{QV}}(\omega) , \tag{2}$$

which looks very similar to Eq. (1). However, we should emphasize that Eqs. (1) and (2) have been derived with rather different assumptions and have dissimilar senses. To obtain Eq. (1) one should suppose a Debye model for the state density, while to derive Eq. (2) it is necessary to admit that the QV-light coupling parameter is a constant value. The $c(\omega)$ in this case is again either equal to a constant or proportional to ω^2 and $g_{\mathrm{QV}}(\omega)$ is the state density of QV's. These assumptions look rather reasonable since the QV-light coupling parameter is formed by the local structure of atom groups, which are responsible for QV's, and may have only small fluctuations around some average value. Unlike this, the phonon-light coupling parameter arises due to comparable-sized phonon wavelength spatial inhomogeneities of the glass structure and has, in the majority of cases, an average value equal to zero.[4,5,14,15]

The main question concerning the problem of far-infrared attenuation (and the "boson" peak as well) in glasses may be summarized as follows: Which expression [(1) or (2)] gives the most contributions to the attenuation? Since $g_D(\omega) \ll g_{\mathrm{QV}}(\omega)$, it seems more plausible to believe that the QV state density plays the main role. However, since the QV-light coupling parameter is mainly determined by the local structure of glasses, as has already been mentioned, the situation can appear when this parameter is very small or even equal to zero due to the peculiarities of the local structure. This situation might arise, for example, in a-SiO$_2$, where, in accordance with neutron data, QV's are assumed to be caused by the rotation of coupled tetrahedrons.[16] The symmetry of these modes does not allow them to be active in the far-infrared region, which was mentioned in Ref. 10. In this situation the state density of QV's could not give any contribution to this attenuation.

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Nevertheless, even in such a case Eq. (1) seems to be inappropriated to account for infrared attenuation in SiO_2 since it does not allow for the phonon-QV interaction, which is of great importance for the transport properties of glasses. Besides, one should note that the attenuation curve for SiO_2 is too smooth in the vicinity of the maximum to be described by Eq. (1) with a realistic model for the correlation properties of the phonon-light coupling parameter.

In this paper we discuss the form of the far-infrared attenuation curve in glasses taking into account the phonon-QV interaction and assuming that QV's are directly inactive in absorption. Besides, we take into consideration both medium- and short-range order of phonon-light coupling parameter.

We demonstrate that allowing for all these factors in combinations can give rise to attenuation curves rather similar to the experimental ones for SiO_2. We find also that three different shapes of the attenuation curve can appear depending upon the characteristic sizes of the medium- and short-range order of glasses and the concentration of QV's. We make some numerical estimations, which support the contention that the model suggested is quite realistic.

II. GENERAL EXPRESSION FOR ATTENUATION TAKING INTO ACCOUNT THE QUASILOCAL VIBRATIONS

In spite of the recent progress in solving the problem of QV's in glasses and a number of models suggested[17–20] there is still no complete picture of the dynamics properties of glasses, which could allow theoretical results to be in quantitative agreement with experimental data. Therefore in this paper we use a phenomenological approach, based on the ideas of Ref. 21.

In the case under consideration the only distinction of the expression for the attenuation compared to the Debye model is to allow the renormalization of the phonon Green's function $G(\epsilon,k)$ due to phonon-QV coupling, $\epsilon=\omega^2$. Assuming that the principal contribution to the attenuation is due to waves with only one polarization and neglecting the wave number of incident light, one can write down the common expression for both models mentioned in Introduction of the interaction between light and phonons:

$$\Gamma(\epsilon)\sim\sqrt{\epsilon}\int c(k)\,\mathrm{Im}\,G(\epsilon,k)\,d^3k\ , \tag{3}$$

where $c(k)=S(k)$ for the MCD and $c(k)=k^2S(k)$ for the MID. Deducing this expression we assumed that the phonon-light coupling parameter does not correlate with the characteristics of QV's, which seems quite natural. Equation (1) follows from Eq. (3) provided that

$$\mathrm{Im}G(\epsilon,k)\sim\delta(\epsilon-s^2k^2)\ ,$$

where s is the sound velocity. QV's result in the imaginary part of the Green's function becoming different from the δ function. In this case Eq. (3) cannot be written down as the product of the state and spectral densities.

To obtain the Green's function taking into account QV's we use the ideas of Ref. 21, which describes QV's introducing a site pseudopotential $U_e(\epsilon)$ and discusses them in terms of resonance scattering of phonons by this pseudopotential. The contribution of QV's to the state density is allowed for by adding the term $-\mathrm{Im}\{\mathrm{Sp}[dU/d\epsilon]G(\epsilon)\}$ to the standard expression $g(\epsilon)=\mathrm{Im}\{\mathrm{Sp}G(\epsilon)\}$ (for details see Ref. 21). The expression for the Green's function can be written in the usual way (see, for example, Ref. 22):

$$G_{kl}=G^0_{kl}+G^0_{km}T_{mn}G^0_{nl}\ , \tag{4}$$

where G^0_{kl} is the Green's function of the matter with no QV's, and T_{mn} is a scattering matrix, which can be written in the form of

$$T_{kl}=t_k\tilde{T}_{kl}\ ,$$

where

$$t_k=U_k/(1-U_kG^0_{kk}) \tag{5}$$

describes the scattering at one site and T obeys equation

$$T_{kl}=\delta_{kl}+G^0_{km}\tilde{t}_mT_{ml}(1-\delta_{km})\ .$$

The one-site pseudopotential $U_l(\epsilon)$ is considered to depend on the frequency; the form of this dependence is determined by the microscopic nature of QV's. This pseudopotential is treated as a random value defined by some distribution function $\rho(U)$. The parameters of the noninteracting QV's are determined by the poles of expression (5).

Since we consider quasilocal excitations, we can limit our analysis to the use of some one-site approximation. The best one is the coherent potential approximation. Using this approach we write down the Fourier transform of the Green's function under investigation in the form

$$G(k)=\frac{1}{\epsilon-s^2k^2-\Sigma(\epsilon)}\ , \tag{6}$$

where mass operator $\Sigma(\epsilon)$ should be determined by the condition that the averaged one-site t matrix calculated with the pseudopotential $U(\epsilon)-\Sigma(\epsilon)$ vanishes. The concrete form of Σ depends on the nature of QV's and the form of the distribution function $\rho(U)$, which are in fact unknown. So we regard Σ as a phenomenological parameter. The exact form of this value is not significant; it is only important that Σ be dependent on the frequency but be independent of wave number. This fact directly follows from the general assumption that the modes under consideration are local ones.

Later on we shall neglect the renormalization of the frequency due to the real part of Σ, suggesting that the main effects are due to the imaginary part, which we write down in the form $\mathrm{Im}\Sigma(\epsilon)=\epsilon\xi(\epsilon)$. This suggestion is supported by the experimental observation of the linear dispersion law of phonons up to 500 GHz frequencies.[23]

In spite of the lack of knowledge of the form of $\xi(\epsilon)$ one can give some qualitative arguments that this function monotonously increases with frequency in the region $\omega\ll\omega_D$, where ω is a Debye frequency. It is well known that at sufficiently small frequencies the pseudopotential $U(\epsilon)$ must have the form $U \sim U_0\epsilon$ to provide the existence of long wave phonons. Therefore the resonant frequency is proportional to U_0^{-1} and the amplitude of the resonant pseudopotentials decreases as the frequency increases. The decreasing of the amplitude causes an increase in the number of resonant pseudopotentials and it is the first reason why the state density of QV's increases with frequency. The second reason is the widening of separate QV's. It follows from the above arguments that the state of density n_{KK}^0 of the noninteracting QV's is an increasing function of frequency. Within the averaged t-matrix approximation the value $\xi \sim \epsilon n_{KK}^0$.[21] Hence ξ also increases with frequency. For example, the calculations, which were carried out[21] in the framework of this approximation on the basis of the model of soft potentials,[17] lead to the expression for $\xi \sim \epsilon^{5/2}$.

Since ξ determines a phonon mean free path, we can compare the results of these speculations with the appropriate experimental data. The dependence of the mean free path on frequency was obtained from the temperature behavior of the heat conductivity,[24] and up to angular frequencies 10^{13} Gz, it appears to be a monotonic function of frequency.

Thus, the main matter of our analysis should be the expression

$$\frac{\Gamma(\epsilon)}{\epsilon} = \mathrm{Im}\frac{1}{\sqrt{\epsilon}} \int \frac{c(k)}{\epsilon - s^2k^2 - i\epsilon\xi} d^3k \ , \tag{7}$$

where ξ is regarded as a steeply increasing function of frequency.

The behavior of Eq. (7) essentially depends on the form of function $c(k)$. This function will be considered to be proportional to k^2 at $k \ll r_c^{-1}$, where r_c is the correlation radius of the inhomogeneities of the phonon-photon coupling parameter. In the framework of the MID such a behavior follows from the relations $c(k) = k^2 S(k)$ with $S(0) = \text{const}$, which is in routine use in the description of the spatial inhomogeneities of disordered systems. In the context of the MCD this type of $c(k)$ was suggested by Schlomann[5] in order to allow anticorrelation effects in the spatial distribution of different kinds of charges.

There have been used two types of $c(k)$ to describe optical properties of glasses. In the Martin-Brenig model of Raman scattering in glasses[25] $c(k)$ was considered to tend to zero steeply at $k \gg r_c^{-1}$, while in Schlomann's paper[5] $c(k)$ was considered to be constant in this region of k. Neither of the two functions can describe the experimental attenuation curve in the model under consideration.

Let us consider first of all the Martin-Brenig function. The attenuation curve in this instance can be described by the following expressions:

$$\frac{\Gamma(\epsilon)}{\epsilon} = \begin{cases} c(\epsilon) + a_0\xi\epsilon^{1/2} \ , & \epsilon \ll \epsilon_c = s^2k_c^2 \ , \ \xi \ll 1 \ , \\ c(\epsilon) + a_\infty \xi\epsilon^{-3/2} \ , & \epsilon \gg \epsilon_c \ , \ \xi \ll 1 \ , \\ a_\infty \xi^{-1}\epsilon^{-3/2} \ , & \epsilon \gg \epsilon_c \ , \ \xi \gg 1 \ , \end{cases} \tag{8}$$

where we suppose that ϵ_c lies in the region of weak scattering. The first and second lines of this expression relate just to this frequency area and the first items describe the attenuation without any QV's. The second items include the contribution of QV's in the approximation of noninteracting QV's. The values a_0 and a_∞ are determined in concrete form by the function $c_0(k)$: $a_0 = \int dk\, c(k)/k^2$, $a_\infty = \int dk\, k^2 c(k)$. If ϵ_c belongs to the frequency region, wherein $\xi > 1$, then one should leave out the second line in Eq. (8). At first glance it would seem that QV's increase attenuation in the region $\epsilon > \epsilon_c$, $\xi \ll 1$ and can give rise to the rather smooth attenuation curve than the function $c(k)$ in itself, producing a better agreement with experimental data for SiO_2.[10] However, a more detailed examination of this curve shows that this effect can appear only at very small ξ, where it rather little influences the attenuation curve. The really marked effect of QV's in this case can appear only if $\xi \gg 1$. However, it is evident from the third line of Eq. (8) that in this case QV's lead to a very steep decreasing of the attenuation in contradiction with observations.

If one use Schlomann's function to calculate the attenuation curve by means of Eq. (7), it will be obtained that

$$\frac{\Gamma(\epsilon)}{\epsilon} = (1+\xi^2)^{1/4}\cos(1/2\arctan\xi) \tag{9}$$

at $\epsilon > \epsilon_c$. The attenuation curve in such a case has no maximum at all. Figure 1 gives the attenuation curves for both situations considered.

Both the Martin-Brening and Schlomann functions are the particular cases of the function

$$c_K(k) = (1 - z^{-3})^{-1}[zS_0(k/z) - z^{-3}S_0(k)] \ , \tag{10}$$

where $S_0(k)$ is a commonly used spectral density associated with, for example, exponential or Gaussian correlation functions. The function $c(k)$ introduced by Crivoglaz[26] is characterized by two correlation radii r_1 and r_2, $z = r_1/r_2 \geq 1$, and meets the condition $c(k) \sim k^2$ at

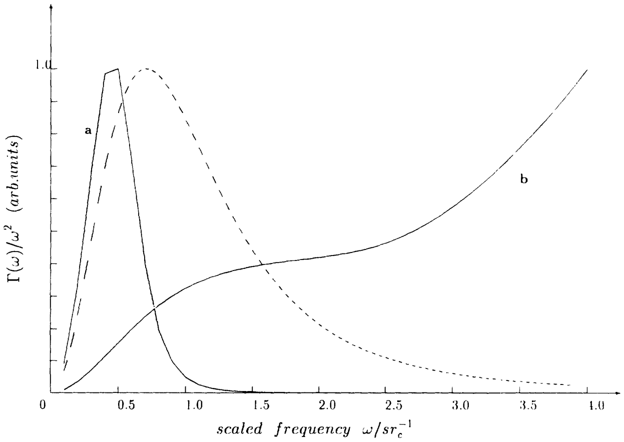

FIG. 1. The attenuation curve obtained with the use of the Martin-Brenig-like (a) and Shlomann (b) functions. The dashed line displays the attenuation curve for the Martin-Brenig-like function without taking into account QV's.

$k \ll r_1^{-1}$. The Martin-Brenig and Schlomann functions can be derived from (10) in the limits of $z \to 1$ and $z \to \infty$ accordingly. Considering this function we take into account both medium- (r_1) and short- (r_2) range order, which seems quite reasonable. Moreover, we will see that taking into account short-range order is of particular importance for the problem under consideration.

Let us consider the case of intermediate frequencies such as $sr_1^{-1} < \omega < sr_2^{-1}$ under conditions $z \gg 1$ and $\xi \ll 1$. By means of the above results we can find

$$\frac{\Gamma(\epsilon)}{\epsilon} = c_K(k) - \xi(a\epsilon^{-3/2} + z^{-1}a_0\sqrt{\epsilon}) \ . \tag{11}$$

One can see from (12) that taking into account the short-range order leads to a negative contribution to the attenuation growing rapidly with frequency. The function $c_K(k)$ appears as a constant in this frequency range and owing to this contribution the bump on the attenuation curve can, in certain conditions, arise.

For the purpose of obtaining numerical estimations we have calculated the attenuation curve using $c_K(k)$ where the function $1/(r_1^{-2} + k^2)$ has been inserted for $S(k)$. In Fig. 2 we display the results of this calculation on plots of $\Gamma(\epsilon)/\epsilon$ versus ω/sr_1^{-1}. In so doing we use $\xi(\epsilon)$ in the form of $\xi = p\epsilon^{5/2}$,[21] where p is proportional to the concentration of QV's. Depending on z we find two lines of behavior for this curve. At $z < 20$ there is one maximum (curve *a*), which becomes steeper as p increases.

At $z > 20$ two maxima can appear (curve *b*). The first one falls in the region of weak scattering and is due to the influence of short-range order. The shape of the attenuation curve in the vicinity of this maximum is described by (11) and the value ω_m depends on both correlation radii r_1 and r_2 and on the parameter p, characterizing QV's. As z and p increase this bump flattens out and disappears eventually (curve *c*). The second maximum of this attenuation curve lies in the strong scattering region and can be described by Eq. (9) and the last line of Eq. (8). Let us note that the position of this maximum $\omega_m^{(2)}$ is determined by the condition $\omega_m^2 = sr_2^{-1}\xi^{-1/2}(\omega_m^2)$ rather than just the correlation radius r_2. We would like to emphasize that curve *b* can appear only if ω_m belongs to the weak scattering region $\xi \ll 1$ (computer simulations show that this curve does not appear at $\xi > 0.2$).

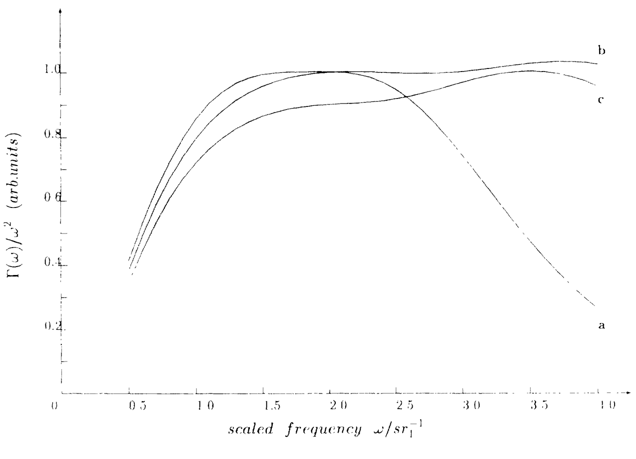

FIG. 2. The attenuation curves obtained using Krivoglaz's function and $\xi = p\epsilon^{5/2}$ at various z and p.

III. NUMERICAL CALCULATIONS AND COMPARISON WITH EXPERIMENTAL DATA

It is obvious from Fig. 2 that curves *a* and *b* are in rather good qualitative agreement with the experimental results of Ref. 10. By this means we show that taking into account phonon-QV coupling and medium- and short-range order, in principal, one can obtain attenuation curves which are similar to the experimental ones, even though QV's are not active in the infrared region. However, to compare the obtained results with the experimental data quantitatively is a more difficult problem.

First of all, we do not know, which phonons, in point of fact, are responsible for the attenuation. Indeed, it can be predicted with certainty that longitudinal phonons make little contribution to the attenuation (due to their large velocity) if it is known that light and phonons interact in accordance with the model of charge defects. But in the context of the model of induced dipoles, the coupling parameter of light with longitudinal phonons can be sufficiently greater than with transversal ones.[7,14] Therefore the contribution of longitudinal phonons in such a case may be the major cause of this effect.

The question—which phonons make the most contribution to the attenuation—is very important. Indeed, we know from various experimental results[14,21,22] that transversal phonons are very strongly scattered at frequencies >500 GHz and the maximum on the curve $\Gamma(\epsilon)/\epsilon$ for SiO_2 falls within the region of strong scattering of transversal phonons. Therefore if one believes that transversal phonons do mainly contribute to attenuation, then we have to conclude that nothing but curve *a* in Fig. 2 is consistent with experiments. Using the experimental data by Ghivelder and Phillips[10] and our results, we estimate the values r_1, r_2, and $\xi(\omega_m)$ in this case. We find $r_1 = 12$ Å, $r_2 = 0.8$ Å, and one can regard these values as quite acceptable for medium- and short-range order accordingly. It is more important to estimate $(kl)^{-1}$ at ω_m [l is the transversal phonon mean free path, which is directly determined by $\xi(\omega_m)$], since one can compare the result with the data of independent experiments. We obtain $(kl)^{-1}$ to be approximately equal to 0.66 while this value from measurements of heat conductivity[22] is 0.8. These estimations show that the situation considered is not unreasonable.

However, if we admit that longitudinal phonons are most important for the attenuation, the situation looks rather different. One may expect that longitudinal phonons are scattered significantly weaker than transversal ones and ω_m can fall in the weak scattering region of longitudinal phonons. In such a case curve *b* in Fig. 2 will most likely agree with observations.

By this means, although we have demonstrated that inactive in infrared QV's can influence the attenuation due to their interaction with phonons and can give rise to the attenuation curves, which are quantitatively similar to the experimental ones, a lot of questions are left unanswered. To clear up the situation new experimental investigations are in order. It would be of use to investigate how the shape of the attenuation curve depends on the parameter p in the expression $\xi = p\epsilon^n$ (n can be equal to $\frac{5}{2}$ or $\frac{3}{2}$ depending on the model of QV's). This value can be determined from the measurements of the specific heat.[27] This investigation could be carried out with the samples prepared under different conditions. Curves a and b display dissimilar behavior versus the value p. If we deal with curve a, the maximum thereof should become sharper and the position of the maximum should move to a lower frequency if p increases. Unlike this, curve b flattens out in the vicinity of the first maximum with increasing p.

It is of great interest also to observe light-induced phonon emission from glasses. By means of such observations one could obtain a lot of information concerning light-phonon interactions in glasses and the part played by QV's in this process as well.

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