Absorption of light by glasses in the far infrared

L. I. Deĭch
Institute of Physics, Siberian Division, Russian Academy of Sciences, 660036 Krasnoyarsk, Russia

(Submitted 1 October 1992)
Pis'ma Zh. Eksp. Teor. Fiz. **56**, No. 10, 508–510 (25 November 1992)

An explanation is proposed for the deviation from the Debye shape of the absorption curve of glasses in the far infrared.

At present, it is generally accepted that the main features of the dynamics of glasses in the terahertz frequency region are associated with quasilocal oscillations (QO).[1] There is also a connection between the appearance of a maximum in the absorption curve, represented in the form $\Gamma(\omega)/\omega$ (Γ is the absorption coefficient and ω is the frequency), in the far infrared, and the density of states of the quasilocal oscillations.[2] However, data on inelastic neutron scattering in α-SiO_2 (Ref. 3) show that the symmetry of quasilocal oscillations in this material does not permit them to be active in infrared absorption.[2] In the present paper we analyze the effect of quasilocal oscillations on the infrared spectra in such a case.

At the core of our treatment is the QO–phonon interaction and also of short-range and medium-range order in the spatial distribution of the photon–phonon coupling parameter. The only difference in the expression for the absorption in comparison with the Debye model in the given case consists of the necessity of taking into account the renormalization of the phonon Green's function. If we assume that the main contribution to the absorption comes from phonons of any polarization, then we can write for the absorption $\Gamma(\epsilon)$

$$\Gamma(\epsilon) \propto \sqrt{\epsilon} \int c(k) \operatorname{Im} G(\epsilon,k) d^3k, \tag{1}$$

where $\epsilon=\omega^2$. The effective photon–phonon coupling parameter $c(k)$ is proportional to the spectral density $S(k)$ of the inhomogeneities of the real coupling parameter. The proportionality coefficient depends on the model of this interaction and is equal either to a constant[4] or to k^2 (Refs. 5 and 6).

Treating the quasilocal oscillations as in Ref. 7 in terms of resonance scattering of phonons in a random pseudopotential, we write the renormalized Green's function in the form $G(k)=[\epsilon-s^2k^2-\Sigma(\epsilon)]^{-1}$, where s is the speed of sound, and the mass operator Σ takes into account the effect of the quasilocal oscillations. We ignore its real part, and write its imaginary part in the form $\operatorname{Im}\Sigma=\epsilon\xi(\epsilon)$, where, following Ref. 7, we assume $\xi=p\xi^{5/2}$.

In order to obtain a general representation of the behavior of the integral (1), it is convenient to consider first its asymptotic behavior for the special case of the function $c_0(k)$, which behaves like k^2 for $k \ll k_c$ and decays faster than k^{-2} for $k \gg k_c$ (k_c characterizes the inhomogeneity scale $r_c \approx k_c^{-1}$). In this case it is not difficult to obtain

 © 1992 American Institute of Physics

$$\frac{A(\epsilon)}{\epsilon}=\begin{cases} c_0(k)+a_0\sqrt{\epsilon\xi} & \epsilon\ll\epsilon_c=s^2k_c^2, \quad \xi\ll 1 \\ c_0(k)+a_\infty\xi\epsilon^{-3/2} & \epsilon\gg\epsilon_c, \quad \xi\ll 1 \\ a_\infty\xi^{-1}\epsilon^{-3/2} & \epsilon\gg\epsilon_c, \quad \xi\gg 1 \end{cases}, \tag{2}$$

where we have assumed that ϵ_c lies in the weak scattering region $\xi\ll 1$. Now we can obtain analogous expressions for the two other types of functions $c(k)$: $c_1(k)$, which is proportional to const for $k\ll k_c$ and decays faster than k^{-2} for $k\gg k_c$, and $c_2(k)$, which, is proportional to k^2 for small values of k and proportional to const for $k\gg k_c$:

$$\frac{A(\epsilon)}{\epsilon}=c_{1(2)}(k)+\begin{Bmatrix} \mp a_0^{1(2)}\sqrt{\epsilon\xi} & \epsilon\ll\epsilon_c \\ \pm a_\infty^{1(2)}\xi\epsilon^{-3/2} & \epsilon\gg\epsilon_c \end{Bmatrix} \xi\ll 1, \tag{3}$$

where the superscripts refer to $c_1(k)$ and the subscripts refer to $c_2(k)$. As a nontrivial fact it should be noted that in the case under consideration the quasilocal oscillations can give a negative contribution to the absorption. In the strong scattering region the asymptotic behavior of the absorption curve for the functions of the type c_2 has the form

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

whereas in the case of the functions c_1 the absorption in this limiting case behaves the same as for c_0.

These results allowed us to carry out a more detailed study of the absorption curve. For this purpose it was convenient to choose the function $c(k)$ in the form

$$C_{SHL}(k)=(1-z^{-3})^{-1}[zS_0(k/z)-z^{-3}S_0(k)], \tag{5}$$

where S_0 is assumed to be a function of the type c_1. This function is characterized by two correlation radii r_1 and r_2, $z=r_1/r_2\geqslant 1$, and can be considered as a generalization of the Schlömann function used to describe experimental results in the charge- defect model.[4] The latter model is obtained from Eq. (5) in the limit $r_2\Rightarrow 0$. This function is useful because it can obtain different forms of the absorption curves characteristic for each of the above-introduced functions c as functions of the order parameter z. In addition, taking into account $r_2\neq 0$ means taking into account the short-range order, which is completely natural from a physical point of view and turns out to be extraordinarily important for the problem under consideration.

Using Eqs. (2)–(4), we obtain any asymptotic limit of the absorption curve for the function c_{SHL}; however, of greatest interest is the intermediate asymptotic limit $sr_1^{-1}\ll\omega\ll sr_2^{-1}$ ($\xi\ll 1$), which has the form

$$\frac{A(\epsilon)}{\epsilon}\propto c_{SHL}(k)-\xi(a_\infty\epsilon^{-3/2}+z^{-1}a_0\sqrt{\epsilon}). \tag{6}$$

Clearly, taking short-range order into account led to the appearance of the negative contribution, which grows rapidly with frequency, in relation (6). Under certain conditions this term can lead to the appearance of a maximum in the curve $A(\epsilon)/\epsilon$.

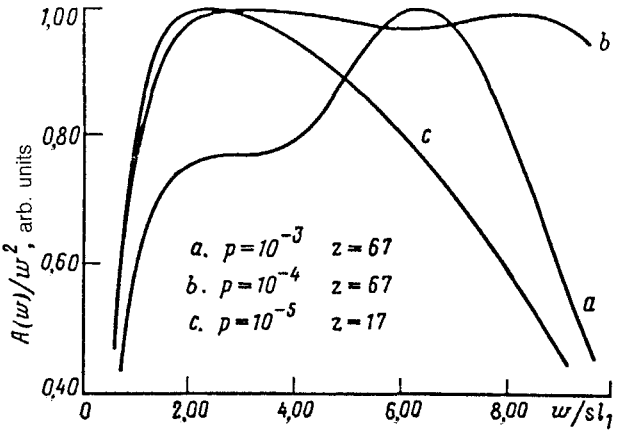

FIG. 1. Different shapes of the absorption curves of $\Gamma(\epsilon)/\epsilon$ for the function $c_{SHL}(k)$.

In order to obtain numerical estimates, we calculated the absorption curve using the function $c_{SHL}(k)$, where S_0 was chosen to correspond to the exponential correlation function. Figure 1 shows the dependence of the function $A(\epsilon)/\epsilon^2$ on the normalized frequency ω/sr_1^{-1}. We found two types of absorption curves, depending on the parameter z. For $z \leqslant 20$ $A(\epsilon)/\epsilon^2$ has one maximum, which lies in the weak scattering region $\xi \ll 1$ (curve c). Its position is determined largely by the correlation length r_1. With increase of the parameter p and decrease of z, this maximum becomes progressively sharper. For $z \geqslant 20$ two maxima are possible: one, as before, in the weak scattering region, and the second for $\xi \gg 1$ (curve b). The shape of the curve in the vicinity of the first maximum can be described by expression (6), and its position is determined by the parameter p and by both the two correlation lengths. With increase of p and z, the curve becomes flatter until it disappears altogether (curve a). The position of the second maximum is given by the expression $\omega_m \cong sr_2^{-1}\xi^{-1}(\omega_m)$, and with increase of p it shifts toward lower frequencies, thereby becoming more pronounced.

As an illustration, we have estimated $r_{1(2)}$ and $\xi(\omega_m)$ for curve b, using experimental data from Ref. 2 and our own results. For the correlation lengths we obtained the completely acceptable numbers 100 Å and 1.4 Å for medium-range order and short-range order, respectively. The parameter $\xi(\omega_m)$ turned out to be roughly 0.025, while an estimate of this same quantity based on measurements of the thermal conductivity[8] gave roughly 0.006.

Thus, the theory of absorption in the far infrared by amorphous SiO_2 proposed in this paper leads to reasonable estimates of the main parameters and allows one to explain how quasilocal oscillations affect the absorption curve when they do not interact with the light directly and when their density of states does not contribute directly to the absorption. Note also that our results do not vary qualitatively if some other sufficiently rapidly increasing function $\xi(\epsilon)$ is chosen.

[1]Yu. M. Galperin, V. G. Karpov, and V. I. Kozub, Adv. Phys. **38**, 669 (1989).
[2]L.. Ghivelder and W. A. Philips, J. Noncryst. Sol. **109**, 280 (1989).
[3]U. Buchenau, M. Prager *et al.*, Phys. Rev. **B34**, 5665 (1986).
[4]E. Schlömann, Phys. Rev. **135**, A413 (1964).
[5]E. Whalley, J. Chem. Soc. Faraday Trans. II **68**, 662 (1972); D. D. Klug and E. Whalley, Physica A **156**, 376 (1989).
[6]L. I. Deich and V. A. Igantchenko, Waves in Random Media **1**, 133 (1991).
[7]Yu. M. Gal'perin, V. G. Karpov, and V. N. Solov'ev, Zh. Eksp. Teor. Fiz. **94**, 373 (1988) [Sov. Phys. JETP **67**, 2386 (1988)].
[8]J. E. Graebner, B. Golding, and L. C. Allen, Phys. Rev. B **34**, 5696 (1986).

Translated by P. F. Schippnick