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## Multiphonon mechanism of light scattering in glasses

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**Abstract.** The absorption of electromagnetic waves and Raman scattering in glasses are calculated. The principal scattering mechanism considered is the interaction of the electromagnetic waves with atoms tunnelling in strongly fluctuating double-well potentials which are characteristic of glasses. The temperature and frequency dependences of the absorption coefficient and the cross section of the small frequency shift Raman scattering are obtained. The results are fitted to the experimental data.

### 1. Introduction

Absorption of electromagnetic waves in glasses in the temperature range from liquid helium to room temperature is usually interpreted in terms of the relaxation interaction of the wave with some localized objects within the glassy samples (see, e.g., reviews in Phillips (1981)). On the other hand an important role can be played by the interaction of the electromagnetic wave with the rigid double-well potentials (DWP) in glasses due to multiphonon processes (Fleurov and Trakhtenberg (1986), see also Goldanskii *et al* (1989)). According to this mechanism tunnelling by the particle in such a DWP is essentially renormalized due to the strong fluctuations of the potential barrier. As a result a multiphonon packet is involved in the interaction of the absorbed particle with the DWP (for the phonon see Fleurov and Trakhtenberg (1986); for the neutron, Fleurov and Levanda (1992); the photon is to be considered in this paper). The multiphonon absorption of the electromagnetic wave is similar in many respects to the multiphonon ultrasonic attenuation considered in Fleurov and Trakhtenberg (1986). There is always a competing relaxation mechanism (Jäckle *et al* 1976) which usually appears to be stronger. However the higher temperatures and higher phonon (or photon in this paper) frequencies favour the multiphonon mechanism. There are two reasons for this: first, the fluctuations of the barrier become stronger with a temperature rise causing a rapid increase in the intensity of the multiphonon transitions; second the relaxation interaction is always proportional to the factor  $[1 + (\tau\omega)^2]^{-1}$  where  $\tau$  is the relaxation time and  $\omega$  is the frequency. Therefore the relaxation interaction becomes weak in the limit  $\tau\omega \gg 1$  which is easily achievable in the THz region. Then the multiphonon interaction would become the leading mechanism determining both the temperature and frequency dependence of the absorption. A similar situation takes place for the attenuation of the electromagnetic waves where the two mechanisms—relaxation and multiphonon—compete, the latter being stronger at higher temperatures and frequencies.

The multiphonon interaction can also be important for the Raman scattering of light. This is an inelastic scattering process when the incident photon is absorbed and then re-emitted with another frequency. The small frequency shift ( $\leq 20 \text{ cm}^{-1}$ ) scattering is usually attributed to two different processes, i.e. the single-phonon and Brillouin scattering processes (Hayes and Loudon 1978). However, they are not able to account for the whole scattering intensity (Winterling 1975, Theodorakopoulos and Jäckle 1976, Fontana *et al* 1989). We believe that both the relaxation mechanism (Jäckle 1981, and references therein) and the multiphonon mechanism (to be discussed in this paper) contribute to this excess scattering. The two mechanisms differ in their temperature and frequency shift dependences. As we shall see later the multiphonon mechanism, in contrast to the relaxation mechanism, does not predict a maximum in the temperature in the Raman scattering cross section. The presence or absence of such a maximum can be a good indication in favour of one of these mechanism under certain experimental conditions.

In this paper we will study the interaction of the electromagnetic waves with rigid DWPs in glasses. Some general relationships describing the interaction of the electromagnetic waves with the DWPs are considered in section 2. The cross section of the Raman scattering due to the multiphonon interaction of the light with the rigid DWPs is discussed in section 3 and the absorption of the electromagnetic wave is calculated in section 4.

## 2. Interaction of the electromagnetic waves with rigid DWPs

A glass sample interacting with the electromagnetic field is described by the Hamiltonian

$$\hat{H} = \sum_{\mathbf{r}_j} \frac{1}{2m} \left( -i\hbar\nabla_{\mathbf{r}_j} - \frac{e}{c} \mathbf{A}(\mathbf{r}_j, t) \right)^2 + \sum_{\mathbf{R}_i} \frac{1}{2M_i} \left( -i\hbar\nabla_{\mathbf{R}_i} + \frac{Z_i e}{c} \mathbf{A}(\mathbf{R}_i, t) \right)^2 + V(\{\mathbf{r}_j\}, \{\mathbf{R}_i\}). \quad (2.1)$$

Here the first two terms stand for the kinetic energy of the electrons and ions interacting with the electromagnetic field  $\mathbf{A}$ .  $M_i$  and  $m$  are the masses of the  $i$ th ion and electrons, respectively,  $e$  is the electron charge,  $c$  is the light velocity,  $Z_i$  is the ion valency.  $V(\{\mathbf{r}_j\}, \{\mathbf{R}_i\})$  includes all sorts of interactions in the electron-ion system of the sample. The following notation will be used for the parts of the Hamiltonian describing the interaction between the electromagnetic field and the glass:

$$\hat{H} = \hat{H}^{AA} + \hat{H}^{AP_e} + \hat{H}^{AP_N} \quad (2.2)$$

where

$$\hat{H}^{AA} = \frac{e^2}{2c^2} \left\{ \frac{1}{m} \sum_j \mathbf{A}(\mathbf{r}_j)^2 + \sum_i \frac{Z_i^2}{M_i} \mathbf{A}(\mathbf{R}_i)^2 \right\}$$

$$\hat{H}^{AP_e} = \frac{ie\hbar}{c} \frac{1}{m} \sum_j \mathbf{A}(\mathbf{r}_j) \nabla_{\mathbf{r}_j},$$

$$\hat{H}^{AP_N} = -\frac{ie\hbar}{c} \sum_i \frac{Z_i}{M_i} \mathbf{A}(\mathbf{R}_i) \nabla_{\mathbf{R}_i}.$$

Considering long wave electromagnetic waves (i.e. the wavelength is larger than the characteristic dimension of the DWPs) we can neglect the space coordinate dependence of the vector potential and use the following expression for the quantized vector potential

$$A(t) = \sum_k \left( \frac{\hbar c^2}{2\epsilon V \omega_k} \right)^{1/2} e_{\mathbf{k}} (\hat{a}_{\mathbf{k}} e^{i\omega_k t} + \hat{a}_{\mathbf{k}}^{\dagger} e^{-i\omega_k t}) \quad (2.3)$$

where  $\omega_k$  is the frequency of the  $k$ th mode;  $\epsilon$  is the dielectric constant of the glass;  $e_{\mathbf{k}}$  is the vector of the polarization of the light;  $\hat{a}_{\mathbf{k}}$  and  $\hat{a}_{\mathbf{k}}^{\dagger}$  are the creation and annihilation operators of the photons,  $V$  is the sample volume. The orthogonality of the different states of the DWP will also allow us in what follows to disregard the contribution of the  $\hat{H}^{AA}$  part of the Hamiltonian (2.2).

Now we need the wavefunctions of the unperturbed states of the glass without interaction with the electromagnetic field (2.2). Using the double adiabatic approximation they can be written in the form:

$$\Phi_{m,\alpha,n}(\{\mathbf{r}_k\}, \{x_l\}, \{\rho_j\}) = \varphi_m(\{\mathbf{r}_k\}; \{x_l\}, \{\rho_j\}) \psi_{\alpha,m}(\{x_l\}; \{\rho_j\}) \Psi_{n,\alpha,m}(\{\rho_j\}). \quad (2.4)$$

Here  $\{\mathbf{r}_k\}$  and  $m$  stand for the electron coordinates and the electronic quantum states. The coordinates  $\{x_l\}$  and  $\{\rho_j\}$  and the quantum numbers  $\alpha$  and  $n$  correspond to the local anharmonic modes and harmonic phonons, respectively. They are obtained from the atomic coordinates  $\{\mathbf{R}_i\}$  by a transformation (Karpov *et al* 1982, Fleurov 1989, see also the review of Galperin *et al* (1989))

$$\{\mathbf{R}_i^{\alpha}\} \rightarrow \{x_l\}, \{\rho_j\}. \quad (2.5)$$

The double adiabatic approximation only works well (Fleurov and Trakhtenberg 1986, Goldanskii *et al* 1989) for rigid DWPs which will be considered later. The standard procedure results in the following equation for the operator of the non-adiabatic interaction between the states

$$\hat{L}_{mam'\alpha'} = \Lambda_{mam'\alpha'}^e + \Lambda_{mam'\alpha'}^N \quad (2.6)$$

where

$$\Lambda_{mam'\alpha'}^N = - \int \psi_{m\alpha} [\hat{T}_N(\{\rho_j\}), \psi_{m'\alpha'}] \delta_{mm'} \{dx_l\}$$

$$\Lambda_{mam'\alpha'}^e = - \int \psi_{m\alpha} \psi_{m'\alpha'} \varphi_m [\hat{T}_N(\{\rho_j\}) + \hat{T}(\{x_l\}), \varphi_{m'}] \{dx_l\} \{d\mathbf{r}_j\}.$$

$\hat{T}_N(\{\rho_j\})$  and  $\hat{T}(\{x_l\})$  are the kinetic energy operators of the harmonic degrees of freedom and those of the DWPs, respectively.

In the calculations that follow we shall need the matrix elements

$$\mathbf{P}_{mm'}^l(x_l, \{\rho_j\}) = -i\hbar \langle \varphi_m(\{\mathbf{r}_j\}, x_l, \{\rho_j\}) | \sum_j \frac{\partial}{\partial \mathbf{r}_j} | \varphi_{m'}(\{\mathbf{r}_j\}, x_l, \{\rho_j\}) \rangle_{\{\mathbf{r}_j\}} \quad (2.7)$$

of the electron momentum. The subscript  $\{\mathbf{r}_j\}$  indicates the integration over the electronic coordinates. It is calculated independently for each DWP,  $l$ , and describes

a transition between the electronic states  $m$  and  $m'$ . It can be done assuming that different DWPs are local entities, do not interact strongly with each other and are treated independently. We shall be interested later only in the transitions between the ground and lowest excited electronic states and we will omit the electronic indices in order to simplify the notation.

The transitions between two states of the  $l$ th DWP are determined by the matrix

$$\Pi_{\alpha\beta}(\{\rho_j\}) = \langle \psi_\alpha(x_l; \{\rho_j\}) | \mathbf{P}_{\alpha\beta}^l(x_l, \{\rho_j\}) | \psi_\beta(x_l; \{\rho_j\}) \rangle_{x_l} = \mathbf{p}_u^l \delta_{\alpha\beta} + \mathbf{p}_d^l \sigma_{\alpha\beta}^z + \mathbf{p}_n^l \sigma_{\alpha\beta}^x \quad (2.8)$$

depending on the phonon configuration  $\{\rho_j\}$ . The subscript  $x_l$  denotes integration over this variable and  $\sigma^x$  and  $\sigma^z$  are the Pauli matrices.

As usual we start here with the non-diagonal *localized* representation (see, e.g., Phillips (1987); this choice is discussed in Fleurov and Levanda (1992)) in which each function  $\psi_\alpha(x; \{\rho_j\})$  describes the tunnelling 'particle' localized in one of the two wells of the DWP. Then the coefficients in the transition amplitude (2.8) can be represented in the form

$$\begin{aligned} \mathbf{p}_u, \mathbf{p}_d &\approx \mathbf{p}_0 \\ \mathbf{p}_n &\simeq \mathbf{p}_0 \exp[-\frac{1}{2}J(\{\rho_j\})] \end{aligned} \quad (2.9)$$

where

$$|\mathbf{p}_0| \approx \hbar/a_{\text{DWP}}.$$

$a_{\text{DWP}}$  is the characteristic scale of the DWP and  $J(\{\rho_j\})$  is the transparency parameter of the DWP barrier for the tunnelling particle.

A relatively weak dependence of the coefficient  $\mathbf{p}_0$  on the phonon coordinates  $\{\rho_j\}$  is neglected compared with the strong dependence of the exponential function. The latter appears to be due to the small overlap of the wavefunctions  $\psi_\alpha(x; \{\rho_j\})$  of the different wells of the DWP and has the same form as the amplitude

$$\Delta(\{\rho_j\}) = \hbar\nu \exp[-\frac{1}{2}J(\{\rho_j\})] \quad (2.10)$$

of the tunnelling transitions between these two wells.

Now the transition to the *diagonal* representation is made by means of the canonical transformation (Fleurov and Trakhtenberg 1986, Fleurov and Levanda 1991)

$$\mathbf{T} = \frac{1}{\sqrt{2\Delta E(T)}} \begin{pmatrix} \sqrt{\Delta E(T) + \epsilon_a} & \sqrt{\Delta E(T) - \epsilon_a} \\ -\sqrt{\Delta E(T) - \epsilon_a} & \sqrt{\Delta E(T) + \epsilon_a} \end{pmatrix} \quad (2.11)$$

where

$$\Delta E(T) = \sqrt{\Delta_1(T)^2 + \epsilon_a^2}$$

$$\Delta_1(T) = \frac{\text{Tr}[e_{\text{ph}}(\{q_j\})\Delta(\{q_j\})]}{\text{Tr}[e_{\text{ph}}(\{q_j\})]}$$

$e_{\text{ph}}(\{q_j\})$  is the density matrix of the phonon subsystem which depends on the deviations  $q_j = \rho_j - \rho_j^{(0)}$  of the phonon variables from their equilibrium values  $\rho_j^{(0)}$ .

The asymmetry parameter  $\epsilon_a$  is the difference between the energies of the particle in each well obtained in the localized representation.

We are interested here in the study of the multiphonon interaction which is due only to the third term in equation (2.8) and is strongly dependent exponentially on the phonon coordinates. The other addends in equation (2.8) do not contain this strong dependence; however, we shall keep them here since they will be used in the calculation of both the Raman scattering and the absorption of the electromagnetic waves. After the transformation (2.11) the transition amplitude (2.9) takes the form

$$\begin{aligned} \tilde{\Pi}_{\alpha\beta}(\{\rho_j\}) = & \mathbf{p}_u \delta_{\alpha\beta} + \mathbf{p}_d \left( -\frac{\Delta_1(T)}{\Delta E(T)} \sigma_{\alpha\beta}^x + \frac{\epsilon_a}{\Delta E(T)} \sigma_{\alpha\beta}^z \right) \\ & + \frac{\mathbf{p}_0}{\hbar\nu} \Delta(\{\rho_j\}) \left( \frac{\Delta_1(T)}{\Delta E(T)} \sigma_{\alpha\beta}^z + \frac{\epsilon_a}{\Delta E(T)} \sigma_{\alpha\beta}^x \right) \end{aligned} \quad (2.12)$$

that will be used later on. The term proportional to  $\delta_{\alpha\beta}$  has been omitted in a similar expression in our previous paper (Fleurov and Levanda 1992) where the multiphonon neutron scattering is discussed. It is unimportant there since it does not contribute to the multiphonon interaction with rigid DWPs in the first order of the perturbation theory. Here, however, we shall deal with second-order processes where the interference of this term with the term proportional to the exponential function  $\Delta(\{\rho_j\})$  makes the principal contribution. As for the second term, proportional to  $\mathbf{p}_d$ , it contributes to the relaxation mechanism.

### 3. Raman scattering of the electromagnetic wave in the glass

In this section the double-differential cross section for the Raman scattering in the glass due to the interaction of the electromagnetic field (2.3) with the DWPs is calculated. It describes the interaction of the electromagnetic wave with the DWPs which causes a scattering of a wave with frequency  $\omega_1$  into a solid angle  $d\Omega$  with a new frequency  $\omega_2$ . In this case the direct interaction of the electromagnetic wave with the ions can be neglected due to their large masses and the interaction with the electrons described by the operator  $\hat{H}^{APe}$  plays the dominant role.

The average cross section per DWP is

$$\begin{aligned} \frac{d^2\sigma_m(\omega_1, \omega_2)}{d\Omega d\omega_2} = & \frac{2\pi}{\hbar} \frac{V^2 \epsilon^2 \omega_2^3}{c^4 \omega_1 N_1 N_{\text{DWP}}} \\ & \times Av_i \sum_{f, \text{DWP}} \left| \sum_{m'\alpha'n'} \frac{\langle \Phi_{m''\alpha''n''} | \hat{H}^{APe} | \Phi_{m'\alpha'n'} \rangle \langle \Phi_{m'\alpha'n'} | \hat{H}^{APe} | \Phi_{m\alpha n} \rangle}{E_{m'\alpha'n'} - E_{m\alpha n} - \hbar\omega_1} \right|^2 \\ & \times \delta(\hbar\omega_1 - \hbar\omega_2 - \delta E) \end{aligned} \quad (3.1)$$

where  $N_1$  is the number of the incident photons in the scattering volume,  $V$ ; and  $N_{\text{DWP}}$  is the number of rigid DWPs in the scattering volume; to make the notation shorter the changes in the photon numbers are not denoted explicitly in equation (3.1). Here the averaging over the initial  $\Phi_{m\alpha n}$ , ( $i$ ), and summation over the final  $\Phi_{m''\alpha''n''}$ , ( $f$ ), states are implied. Summation over all DWPs participating in the process should also be carried out in (3.1). Raman scattering is the process which first appears to

second order in the electric field since a photon with the frequency,  $\omega_1$ , is absorbed and then a photon with another frequency,  $\omega_2$ , is emitted. This process is accompanied by a corresponding change in the energy  $\delta E$  of the glass. It may be due to a change in the electronic, phonon or DWP quantum numbers. However, here we are interested in a frequency range corresponding to energies which are much smaller than the characteristic electronic energies. Therefore, we may assume that the initial and final electronic states coincide,  $m = m'' = 0$ .

The operators  $-i\hbar\nabla_{\mathbf{r}_j}$ , in  $\hat{H}^{APc}$  necessarily change the electronic quantum numbers  $m$  and the intermediate electronic state differs from the ground state  $m' \neq 0$ . The leading contribution of the first excited electronic state in the cross section (3.1) will be considered later. Substituting equation (2.12) into (3.1) one obtains three terms which are proportional to the zeroth, first and second order of the exponential function  $\exp[-J(\{\rho_j\})]$ . The zeroth-order term is responsible here for the relaxation processes and does not contribute to the multiphonon processes at all while the second-order term is small compared with the first-order term. Therefore only the first-order term will be taken into account.

The value of the energy denominator in the equation is determined mainly by the electronic energy,  $E_e$ , the changes in the phonon and DWP energies giving only relatively small corrections. Therefore, we may substitute it by a constant,

$$E_{m'\alpha'n'} - E_{m\alpha n} - \hbar\omega_1 \simeq E_e - \hbar\omega_1 \quad (3.2)$$

and remove it from the sums.

Now we use equation (2.2) for the interaction and equation (2.12) for the matrix elements of the operator of the electron momenta. The result is

$$\frac{d^2\sigma_m(\omega_1, \omega_2)}{d\Omega d\omega_2} = \frac{1}{N_{DWP}} \sum_{DWP} \left\{ \left( \frac{\Delta_1(T)}{\Delta E(T)} \right)^2 \tilde{\sigma}(0) + \left( \frac{\epsilon_a}{\Delta E(T)} \right)^2 \frac{1}{\cosh(\beta\Delta E/2)} \Sigma(\Delta E) \right\} \quad (3.3)$$

where

$$\begin{aligned} \tilde{\sigma}(E) &= \frac{\pi\omega_2^2}{2\hbar c^4\omega_1^2} \left( \frac{p_0^2}{\hbar\nu} \right)^2 \left( \frac{e}{m} \right)^4 \frac{1}{(E_e - \hbar\omega_1)^2} \\ &\times Av_i \sum_f |\langle \Psi_{0\alpha'n'}(\{\rho_j\}) | \Delta(\{\rho_j\}) | \Psi_{0\alpha n}(\{\rho_j\}) \rangle|^2 \\ &\times \delta(E + \Delta E_{ph} - \hbar\omega_1 + \hbar\omega_2) \end{aligned} \quad (3.4)$$

and

$$\Sigma(\Delta E) = \tilde{\sigma}(\Delta E)e^{-\beta\Delta E/2} + \tilde{\sigma}(-\Delta E)e^{\beta\Delta E/2}.$$

Equation (3.4) for the quantity  $\tilde{\sigma}(\Delta E)$  coincides to within the prefactor with the equation for the rate constant for the tunnelling transitions through the fluctuating barrier as calculated in Fleurov and Trakhtenberg (1986) (see also Goldanskii *et al*

1989). The principal details of these calculations are given in Fleurov and Levanda (1992). Using these results the Raman cross section can be represented as

$$\frac{d^2\sigma_m(\omega_1, \omega_2)}{d\Omega d\omega_2} = \frac{1}{N_{\text{DWP}}} \frac{\hbar^2}{4c^4} \left(\frac{\omega_2}{\omega_1}\right)^2 \left(\frac{p_0^2}{\hbar\nu}\right)^2 \left(\frac{e}{m}\right)^4 \times \frac{1}{(E_e - \hbar\omega_1)^2} \sum_{\text{DWP}} \frac{g(T)}{\tau(T)} \exp\{-\frac{1}{2}\beta\hbar(\omega_2 - \omega_1)\}. \tag{3.5}$$

Here

$$g(T) = \cosh^{-1} \left( \frac{\Delta E(T)}{2kT} \right) \left[ 1 + 8 \left( \frac{\Delta_1(T)}{\Delta E(T)} \right)^2 \sinh^2 \left( \frac{\Delta_1(T)}{4kT} \right) \right].$$

The quantity  $\tau(T)$  in equation (3.5) is the multiphonon relaxation time in a rigid DWP. The calculation carried out in our previous papers (Fleurov and Trakhtenberg (1986), see also Fleurov and Levanda (1992)) assumes that the fluctuations in the DWP barrier are mainly caused by its interaction with only one phonon mode characterized by the frequency  $\Omega$  and reduced mass  $M$ . The equation obtained there reads

$$\frac{1}{\tau(T)} = \frac{\nu^2}{\Omega\sqrt{\varphi_1\varphi_2\varphi_3}} \exp(-J_0 + \varphi_1) \tag{3.6}$$

where

$$\begin{aligned} \varphi_1(T) &= \frac{R}{4} \left[ \tanh\left(\frac{\beta\hbar\Omega}{4}\right) + \frac{R_1}{2} \right]^{-1} \\ \varphi_2(T) &= \frac{\sinh(\beta\hbar\Omega/4)/\cosh^3(\beta\hbar\Omega/4)}{2[\tanh(\beta\hbar\Omega/4) + R_1/2]} \\ \varphi_3(T) &= \left[ 1 + \frac{R_1}{2} \coth\left(\frac{\beta\hbar\Omega}{4}\right) \right] \left[ 1 + \frac{R_1}{2} \tanh\left(\frac{\beta\hbar\Omega}{4}\right) \right]. \end{aligned}$$

The two dimensionless parameters in equation (3.6) are

$$R = \frac{\hbar}{2M\Omega} \left( \frac{\partial J}{\partial q} \right)^2 \quad R_1 = \frac{\hbar}{2M\Omega} \frac{\partial^2 J}{\partial q^2}.$$

Equation (3.5) enables an analysis of the temperature and frequency dependence of the cross section of the Raman scattering due to the multiphonon interaction of the light with the rigid DWPs. If we average over the DWPs in the manner described in Fleurov and Levanda (1992) assuming a broad distribution of the asymmetry parameter,  $\epsilon_a$ , and a narrow distribution of the tunnelling parameter,  $J_0$ , then the principal factors determining the behaviour of the cross section are the relaxation time  $\tau(T)$  and exponential function. The latter also determines the dependence of the cross section (3.1) on the frequency shift  $\omega_2 - \omega_1$  in the scattering process.

For a frequency shift which is not too large, when this formula holds best of all, the temperature dependence of the double-differential cross section is determined mainly by the relaxation time  $\tau(T)$  for which we have equation (3.6). Using this expression



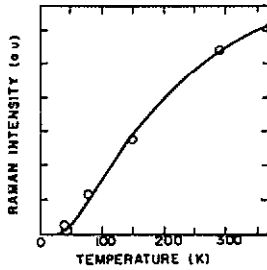


Figure 1. The temperature dependence of the Raman scattering in vitreous silica for the frequency shift of 150 GHz. The circles show the experimental data of Buchenau *et al* (1988). The theoretical results are shown by the full curve.

we may try and fit the double-differential cross section (3.1) to the experimental data of Buchenau *et al* (1988) (see figure 1). The measurements were made on vitreous silica at relatively high temperatures and correspond to the photon frequency change of 160 GHz. The fitting was done using the parameters:  $R = 500$ ,  $R_1 = 4.5$  and  $\Omega/2\pi = 0.64$  THz. These are the same parameters that we used in our multiphonon approach to neutron scattering (Fleurov and Levanda 1992) and to the ultrasonic attenuation (Fleurov and Trakhtenberg 1986).

#### 4. Absorption of the electromagnetic waves in the glass

Absorption of the electromagnetic waves in the glass can be treated in the way similar to that for the phonon attenuation (Fleurov and Trakhtenberg 1986, Goldanskii *et al* 1989). We shall consider here the frequencies of the electromagnetic waves which are not too high and do not cause transitions to higher electronic states. Now both the  $\hat{H}^{AP_e}$  and  $\hat{H}^{AP_N}$  parts of the interaction Hamiltonian (2.2) contribute to the absorption of the electromagnetic wave. The operator  $\hat{H}^{AP_N}$  is smaller than the operator  $\hat{H}^{AP_e}$  due to the difference between the electron and ion masses. However, the operator  $\hat{H}^{AP_N}$  does not change the electronic state of the system and for this reason it contributes to the lowest order. As for the larger operator  $\hat{H}^{AP_e}$  it changes the electronic state and therefore one needs weak non-adiabatic processes returning the system to its initial electronic state. As will be seen later the two contributions are of the same order.

The absorption coefficient (or the inverse free path length for the photons in the glass) due to the rigid DWPs is determined by the equation

$$l_m^{-1} = l_+^{-1} - l_-^{-1} \quad (4.1)$$

where + and - denote absorption or emission of a photon. The inverse lengths in equation (4.1) are given by the equation

$$l_{\pm}^{-1} = \frac{1}{cN_1} \frac{2\pi}{\hbar} \sum_{\text{DWP}} Av_i \sum_f |\langle \Psi_{n_i, \alpha, 0}(\{\rho_j\}) | \mathcal{R}_{\alpha\beta}(\{\rho_j\}) | \Psi_{n_f, \beta, 0}(\{\rho_j\}) \rangle|^2 \delta(\delta E \mp \hbar\omega_1) \quad (4.2)$$

where the effective amplitude of the process is calculated to the second order of the perturbation theory:

$$\begin{aligned} \mathcal{R}_{\alpha\beta}(\{\rho_j\}) = & \langle \varphi_0 \psi_{\alpha,0} | \hat{H}^{APN} | \varphi_0 \psi_{\beta,0} \rangle \\ & + \langle \varphi_0 \psi_{\alpha,0} | \sum_{\gamma,n''} \frac{\hat{H}^{APc} |\varphi_1 \psi_{\gamma,1}\rangle |\Psi_{n'';\gamma,1}\rangle \langle \Psi_{n'';\gamma,1} | \langle \varphi_1 \psi_{\gamma,1} | \hat{L}}{E_{1\gamma n''} - E_{0\alpha n} \mp \hbar\omega_1} \\ & + \sum_{\gamma,n''} \frac{\hat{L} |\varphi_1 \psi_{\gamma,1}\rangle |\Psi_{n'';\gamma,1}\rangle \langle \Psi_{n'';\gamma,1} | \langle \varphi_1 \psi_{\gamma,1} | \hat{H}^{APc}}{E_{1\gamma n''} - E_{0\alpha n}} | \varphi_0 \psi_{\beta,0} \rangle. \end{aligned} \quad (4.3)$$

Equation (4.3) implies integration over the electron and DWP coordinates. The integration over the phonon coordinates  $\{\rho_j\}$  is carried out in equation (4.2).

We have a convenient expression for the matrix (2.12) produced by averaging the interaction of the electromagnetic field with the electrons,  $\hat{H}^{APc}$ , over the electronic and DWP variables. Similar equations produced by two other interactions,  $\hat{H}^{APN}$  and  $\hat{L}$  are now needed.

First, the ion contribution (operator  $\hat{H}^{APN}$ ) is considered. Using the standard procedure applied both in this and the previous paper the corresponding interaction of the electromagnetic wave with the  $l$ th DWP can be written as

$$\mathbf{D}'_{\alpha\beta}(\{\rho_j\}) = \langle \psi_{\alpha;0} | \sum_i \frac{Z_i \hbar \nabla_{R_i}}{M_i} | \psi_{\beta;0} \rangle = \frac{1}{M_r} (\mathbf{d}'_u \delta_{\alpha\beta} + \mathbf{d}'_d \sigma^z_{\alpha\beta} + \mathbf{d}'_n \sigma^x_{\alpha\beta}). \quad (4.4)$$

The bra and ket vectors in equation (4.4) represent the electronic and DWP parts of the wavefunctions (2.4). Integration over all coordinates except for the phonon ones is implied. The electronic state does not change and is assumed to be the ground one.  $M_r$  is a reduced mass associated with the DWP which also accounts for the valences  $Z_i$  of the atoms. Using the localized representation for the DWP wavefunctions one can write (e.g., (2.9))

$$\begin{aligned} \mathbf{d}_d & \approx \mathbf{d}_0 \\ \mathbf{d}_n & \approx \mathbf{d}_0 \exp[-\frac{1}{2} J(\{\rho_j\})] \\ |\mathbf{d}_0| & \approx \theta^{-1/2} p_0 \end{aligned} \quad (4.5)$$

where  $\theta = \sqrt{m/M}$ ,  $m$  is the electron mass and  $M$  an atomic mass. Comparing equations (4.5) with equations (2.9) one sees that the parameter  $\theta$  is the ratio of the characteristic atomic and electronic momenta. This parameter also characterizes the scale of the non-adiabatic interaction to be considered later. Only the term proportional to the exponential function  $\Delta(\{\rho_j\})$  is kept in equation (4.4) since this operator appears to the first order in the effective amplitude (4.3).

Now the canonical transformation (2.11) converts the matrix (4.4) into the matrix

$$\tilde{\mathbf{D}}_{\alpha\beta}(\{\rho_j\}) = \frac{1}{M_r} \frac{\mathbf{d}_0}{\hbar\nu} \Delta(\{\rho_j\}) \left( \frac{\Delta_1(T)}{\Delta E(T)} \sigma^z_{\alpha\beta} + \frac{\epsilon_a}{\Delta E(T)} \sigma^x_{\alpha\beta} \right) \quad (4.6)$$

describing the interaction of the electromagnetic wave directly with the atoms of the DWP.

A similar procedure can be applied to the non-adiabaticity operator (see equation (2.6)). If the integration only with respect to the electronic and the DWP variables is carried out in the expression for  $\Lambda_{m\alpha m'\beta}^e$  then one gets the operator

$$L_{\alpha\beta}(\{\rho_j\}) = L_u \delta_{\alpha\beta} + L_d \sigma_{\alpha\beta}^z + L_n \sigma_{\alpha\beta}^x \quad (4.7)$$

where

$$L_n = L_0 \exp[-\frac{1}{2}J(\{\rho_j\})]$$

$$L_u, L_d \approx L_0 \approx p_0^2/M\theta^{1/2}.$$

The operator  $L_{\alpha\beta}$  describes the non-adiabatic transition with a change of the electronic state which is not denoted explicitly in the equation (4.7) in order to reduce notations. The canonical transformation (2.11) converts (4.7) into

$$\begin{aligned} \tilde{L}_{\alpha\beta}(\{\rho_j\}) = & L_u \delta_{\alpha\beta} + L_d \left( \frac{\Delta_1(T)}{\Delta E(T)} \sigma_{\alpha\beta}^z + \frac{\epsilon_a}{\Delta E(T)} \sigma_{\alpha\beta}^x \right) \\ & + \frac{L_0}{\hbar\nu} \Delta(\{\rho_j\}) \left( \frac{\Delta_1(T)}{\Delta E(T)} \sigma_{\alpha\beta}^z + \frac{\epsilon_a}{\Delta E(T)} \sigma_{\alpha\beta}^x \right). \end{aligned} \quad (4.8)$$

Now the effective interaction amplitude  $\mathcal{R}_{\alpha\beta}(\{\rho_j\})$  is simplified using the assumption that the energy denominators in the equation (4.3) are determined mainly by the electronic gap  $E_e$  (see (3.2)) and does not depend on the phonon and DWP quantum numbers. Then one can make use of the standard equation

$$\sum_{n''} |\Psi_{1,\gamma,n''}(\{\rho_j\})\langle\Psi_{1,\gamma,n''}(\{\rho'_j\})| = \prod_j \delta(\rho_j - \rho'_j) \quad (4.9)$$

in order to get rid of the phonon functions  $\Psi_{1,\gamma,n''}(\{\rho_j\})$  (see definition (3.2)) in the equation for the effective amplitude (4.3).

Having all three possible interactions described by equations (2.12), (4.6) and (4.8) we can calculate the effective amplitude. In fact we only need the part proportional to the exponential function  $\Delta(\{\rho_j\})$  which is responsible for the multiphonon processes.

$$\mathcal{R}_{\alpha\beta}^{\pm}(\{\rho_j\}) = \sqrt{\frac{e^2 \hbar N_1}{2\epsilon V \omega_1}} \frac{R_{\pm}}{\hbar\nu} \Delta(\{\rho_j\}) \left( \frac{\Delta_1(T)}{\Delta E(T)} \sigma_{\alpha\beta}^z + \frac{\epsilon_a}{\Delta E(T)} \sigma_{\alpha\beta}^x \right) \quad (4.10)$$

where

$$R_{\pm} = \frac{1}{M_r} d_0 + \frac{1}{m} (p_0 L_u + p_u L_0) \left( \frac{1}{E_e} + \frac{1}{E_e \mp \hbar\omega_1} \right). \quad (4.11)$$

In order to verify that two terms in equation (4.11) are of the same order we have to use the estimates (2.9), (4.5) and (4.7) for the quantities present in  $R_{\pm}$ . As for the electronic energy it can be estimated using the equation

$$E_e \approx p_0^2/2m.$$

Then one gets  $R_{\pm} \approx p_0^2/\sqrt{\theta}M_r$ . Now substituting expressions (4.10) into equations (4.1) and (4.2) and carrying out the standard procedure used in our approach the multiphonon absorption coefficient is

$$I_m^{-1} = \frac{e^2}{2\hbar\nu^2 c\epsilon V} \sum_{\text{DWP}} \frac{g(T)}{\tau(T)} \frac{1}{\omega_1} (R_+^2 e^{\beta\hbar\omega_1/2} - R_-^2 e^{-\beta\hbar\omega_1/2}). \quad (4.12)$$

Equation (4.12) is similar to the equations obtained for the Raman scattering (3.5) and the equation for the neutron scattering cross section obtained in Fleurov and Levanda (1992) and uses the same notation. Together with expression (3.6) for the relaxation time  $\tau(T)$  it gives a complete description of the photon absorption in the glass caused by the multiphonon interaction of the light with the DWPs. It differs from the absorption due to the relaxation mechanism which always contains an additional factor  $[1 + (\tau(T)\omega)]^2$ . That is why the temperature and frequency dependence of the multiphonon and the relaxation absorption coefficients differ and can be distinguished experimentally. Using the results of the analysis parallel to that made for the sound attenuation (Fleurov and Trakhtenberg 1986) one can say that the multiphonon mechanism of the electromagnetic wave attenuation is most important at temperatures approaching room temperature and frequencies close to the Debye frequencies.

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