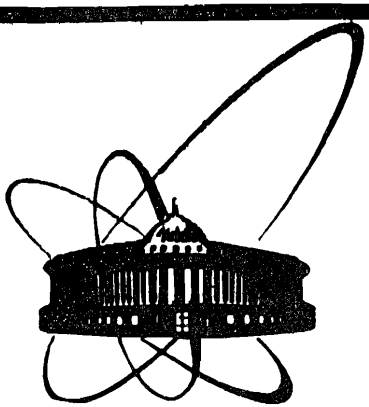

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B.Esser, A.Selloni*

MULTIPHONON COUPLING OF OPTICAL TRANSITIONS IN DISORDERED SYSTEMS AND INVERSE PARTICIPATION RATIO

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* Permanent address: International School for Advanced Studies, 34014 Trieste, Italy

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In the last years a considerable progress has been achieved in understanding the localization problem in disordered systems (see e.g. /1/). Most of the work is concerned with the influence of localization on transport properties because of the drastic influence of the localized to extended states transition on the electrical conductivity. The electronic wavefunctions, however, enter also in such expressions as the familiar coupling functions of optical transitions to multiphonon processes /2/. Therefore, when considering optical transitions involving localized states with varying degree of localization - and hence varying amount of network relaxation - some influence of the localized to extended states transitions on the optical properties can be expected. In this short note we show that in some approximation the coupling function for multiphonon processes is related to the inverse participation ratio of the electronic states /3/ which displays a critical behaviour at the mobility adge /4/. Using this connection we consider the temperature dependence of the absorption tail.

We start from the following expression of the absorption coefficient $\alpha(\omega,T)$ for optical transitions coupled to vibrations

$$\alpha(\omega,T)=\frac{(2\pi e)^2}{3ncm_0^2\omega V}\sum_{f,i}|\langle f|\hat{\bar{p}}|i\rangle|^2 C_{fi}^{T}(\omega-\omega_{fi}). \quad (1)$$

Here i (f) denotes the initial (final) states of the transitions, $\hat{\bar{p}}$ is the momentum operator and $C_{fi}^{T}(\omega-\omega_{fi})$ describes the temperature dependent coupling of the electronic states to the phonon system. The notation in front of the sum is standard. The function $C_{fi}^{T}(\omega-\omega_{fi})$ is taken in the form

$$C_{fi}^{T}(\omega-\omega_{fi})=\frac{1}{2\pi}\int_{-\infty}^{+\infty}ds\, e^{is(\omega-\omega_{fi})+\chi_{fi}^{T}(s)} \quad (2)$$

where ω_{fi} is the difference of the electronic energies and $\chi_{fi}^{T}(s)$ is given by /2/

$$\chi_{fi}^{T}(s)=\frac{1}{V}\sum_{q}\frac{\omega_q}{2}\left(\Delta_q^{fi}\right)^2\left[(N_q+1)(e^{-i\omega_q s}-1)+N_q(e^{+i\omega_q s}-1)\right] \quad (3)$$

Here ω_q and N_q are the frequency and occupation number of the q-th phonon mode, respectively. The coupling is due to the difference Δ_q^{fi} in the equilibrium positions, q_o^i and q_o^f of the normal coordinates for the initial and final states. The $q_o^{f,i}$ are expressed in terms of the deformation potentials $D_{f,i}$ and electronic wavefunctions $\Psi_{f,i}$ as

$$q_{qo}^{f,i}=\frac{D_{f,i}}{\omega_q^2}\int d^d\bar{x}\, e^{i\bar{q}\bar{x}}|\Psi_{f,i}(\bar{x})|^2. \quad (4)$$

In what follows we consider optical transitions from the localized states of the valence band tail to extended states of the conduction band. This problem can be important, for instance, for the interpretation of the absorption tail in amorphous Silicon, where the influence of disorder on the conduction band states is known to be much less relevant than for the valence band states (see,e.g./5/). Then $q_o^f\sim\delta_{q,o}$ and the shift Δ_q^{fi} is due to q_o^i which varies for different tail states because $\Psi_i(\bar{x})$ becomes increasingly delocalized approaching the mobility edge. In particular, for the initial energies E_i crossing the mobility edge and hence $\Psi_i(\bar{x})$ entering the extended state region we have $q_o^i\sim\delta_{q,o}\Delta_{q,o}^{fi}\to 0$. In order to express this behaviour of the coupling in the simplest form we introduce an average phonon frequency by setting $\omega_q=\omega_o$ in (3). Inserting (4) into (3) we obtain

$$\chi_i^{T}(s)=\frac{D^2}{2\omega_0^3}f(s,T)P(E_i), \quad (5)$$

where

$$f(s,T)=(N_o+1)(e^{-i\omega_o s}-1)+N_o(e^{+i\omega_o s}-1) \quad (6)$$

and

$$P(E_i)=\frac{1}{V}\sum_q\left[\int d^d\bar{x}\, e^{i\bar{q}\bar{x}}|\Psi_i(\bar{x})|^2\right]^2. \quad (7)$$

By interchanging the q and x integrals it is easily seen that (6) is equivalent to

$$P(E_i)=\int d^d\bar{x}\,|\Psi_i(\bar{x})|^4, \quad (8)$$

which is the continuous representation of the inverse participation ratio /3,4/; $P(E_i)$ is approximately the inverse volume over which the electronic density $|\psi_i(\bar{x})|^2$ is effectively spread, e.g. $P(E_i) \sim \sim [\chi(E_i)]^3$, $\chi(E_i)$ being the inverse localization radius. Approaching the mobility edge from the region of localized states one obtains $\chi(E_i) \longrightarrow 0$ and correspondingly $P(E_i) \longrightarrow 0$. Considering the influence of the E_i dependence of $P(E_i)$ on the tail of $\alpha(\omega,T)$ we assume a smooth energy dependence of the $\hat{p}$-matrix elements for the localized to extended transitions and take them out of the sum in (1). Then eq. (1) is equivalent to

$$\alpha(\omega,T) = \frac{A}{\omega}\int dE_1 \int dE_2\, \rho_v(E_1)\rho_c(E_2)\, C_{21}^{T}(\omega - E_2 + E_1), \tag{9}$$

where the initial and final densities of states (dos), $\rho_v(E_1)$ and $\rho_c(E_2)$, respectively, were introduced. Now the following model assumptions are made. The valence band tail dos is taken as

$$\rho_v(E_1) = N_v e^{-(E_1 - E_v)/\Delta_v} \tag{10}$$

and the dos of the conduction band as

$$\rho_c(E_2) = N_c\sqrt{E_2 - E_c}. \tag{11}$$

Here E_v and Δ_v are the mobility edge and the tailing parameter, respectively, and E_c is the conduction band edge. In order to perform the E_1 integral analytically, the E_1 dependence of $P(E_1)$ is assumed to be of the form

$$P(E_1) = \frac{1}{\vartheta_0}\frac{E_1 - E_v}{\Delta_\ell}, \quad E_1 \geq E_v, \tag{12}$$

i.e. the critical index is 1, ϑ_0 and Δ_ℓ are parameters having the dimensions of volume and energy, respectively. Inserting (2) with (5) into (9), performing the E_1 integration, using (10) and (12) we obtain

$$\alpha(\omega,T) = \frac{A'}{\omega}\int dE_2 \rho_c(E_2)\int_{-\infty}^{+\infty}\frac{ds}{1 - is\Delta_v - af(s,T)}\, e^{is(\omega - E_2 + E_v)}, \tag{13}$$

where

$$a = \frac{\Delta_v D_v^2}{2\vartheta_0\Delta_\ell\omega_0^3} \tag{14}$$

and $f(s,T)$ is given by (6). For no phonon coupling, i.e. $a=0$, the s-integral in (13) is easily done using the pole $s_0 = -\frac{i}{\Delta_v}$. Then by inserting (11) into (13) an optical tail is obtained with the same tailing energy Δ_v as the valence dos (10). For $a > 0$ the position of the pole s_0 is modified by the function $f(s,T)$ resulting in a temperature dependent optical tail. We consider this modification for a small coupling $0 < a \ll 1$ where the contribution of other poles is negligible. By setting $s=-it, t > 0$, we solve the equation $1 - t\Delta_v - af(-it) = 0$ iteratively. For small a we can restrict ourselves to the 1st iteration, and employing the explicit form of $f(s,T)$, eq.(6), we obtain

$$s_1 = -\frac{i}{\Delta_v}\left\{1 - a\left[(N_0+1)\left(e^{-\frac{\omega_0}{\Delta_v}} - 1\right) + N_0\left(e^{+\frac{\omega_0}{\Delta_v}} - 1\right)\right]\right\}. \tag{15}$$

Closing the integration contour in the lower plane and performing the E_2 integral with (11) we find

$$\alpha(\omega,T) \sim e^{\frac{\omega - E_g}{\Delta(T)}}, \tag{16}$$

where $E_g = E_c - E_v$ and $\Delta(T)$ is the temperature dependent tailing energy

$$\Delta(T) = \Delta_v\left[1 - a\left(1 - e^{-\frac{\omega_0}{\Delta_v}}\right) + \frac{2a\left(\mathrm{ch}\frac{\omega_0}{\Delta_v} - 1\right)}{e^{\omega_0/T} - 1}\right]. \tag{17}$$

In conclusion we note the following points:

1. By using a constant coupling, i.e. neglecting the E_i dependence of the coupling function (5) we obtain an expression for $\alpha(\omega,T)$ different from (13). We have earlier investigated this constant coupling numerically and obtained parallel shifts of the optical tail to lower frequencies with increasing T, i.e. for this case $\Delta(T) \cong$ const., in contrast with the experimental results /5/. On the other hand, the E_i - dependence of the coupling strength, originating in our case from the inverse participation ratio P(E) in (5), gives rise to a T - dependent tailing energy (15) in agreement with the form used for fitting the experimental data /5/. Hence our calculation is a strong indication - at least within our model - - that the varying degree of localization of the tail states has to be taken into account to describe optical properties in disordered systems properly.

2. Considering the $T \to 0$ limit in (17) we find that the optical tailing energy $\Delta(T=0) < \Delta_V$ — the characteristic energy of the dos tail (10). This effect is due to the larger amount of network relaxation for deeper tail states implying a greater Stokes shift for these states. This effect shifts the optical transitions originating from the deeper tail states to higher energies compared to the smaller shifts of the states nearby the edge having a weaker network relaxation and hence a weaker Stokes shift. At T=0 this results in an optical tail narrower than the dos tail. Obviously this effect is due to the energy dependence of P(E) implying different Stokes shifts. Hence, by considering an energy dependent coupling we have indicated another mechanism why dos tails are usually broader than the optical tails (/6/, chapter 5).

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Эссер Б., Зеллони А. E17-87-441

Многофононная связь оптических переходов
в неупорядоченных системах
и обратная величина участия

Рассматривается влияние электронных состояний с разной степенью локализации на электрон-фононную функцию связи оптических переходов. Функция связи выражается через обратную величину объема, участвующего в формировании локализованного состояния. Вычисляется температурная зависимость хвоста коэффициента поглощения.

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Multiphonon Coupling of Optical Transitions
in Disordered Systems
and Inverse Participation Ratio

Optical transitions involving localized states with a varying degree of localization and electron-phonon coupling are considered. The coupling function is shown to be related to the inverse participation ratio displaying a critical behaviour at the mobility edge. Using this connection the temperature dependence of the absorption tail is obtained in a form consistent with experimental data.

The investigation has been performed at the Laboratory of Theoretical Physics, JINR.

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