

INTERBAND ABSORPTION SPECTRA OF DISORDERED SEMICONDUCTORS
 IN THE COHERENT POTENTIAL APPROXIMATION

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The interband absorption spectra of a cellular disordered system with Gaussian site-diagonal randomness are calculated in the coherent potential approximation. Two characteristic features of the spectra are the relaxation of the k-selection rule and the exponential tail of the absorption edge (the Urbach tail). An interband stochastic correlation of site-energies induces the electron-hole vertex correction which leads to a shift of the absorption edge.

I. Introduction

The interband optical transition in disordered semiconductors reflects the nature of the electronic states in disordered systems. In the first place, the optical k-selection rule is supposed to relax due to the phase incoherence of the wave functions. Although the assumption of partial or complete relaxation of the k-selection rule has provided [1,2] a basis for the derivation of E^2 -rule of the absorption edge in amorphous semiconductors, there have been few theoretical attempts to consolidate the foundation of the assumption. In the next place, the localization of the wave functions should induce the low energy tail of the optical absorption edge. Thirdly, it is of particular interest for a certain class of phenomena to investigate the effect of the interband correlation of randomness, such as parallel or antiparallel potential fluctuations of the conduction and valence bands.

In this paper, the interband absorption spectra of a system with cellular disorder are calculated in the coherent potential approximation (CPA) [3] to clarify the above-mentioned points. The electron-hole Coulomb interaction will be neglected for simplicity.

II. Model

Let us consider a two-band system described by the tight-binding Hamiltonian

$$H = \sum_{\mu=1,2} \sum_n (\sum_{\nu} \epsilon_{n\nu} a_{n\nu}^* a_{n\nu} + \sum_{n \neq m} \sum_{\nu} v_{nm\nu} a_{n\nu}^* a_{m\nu}), \quad (1)$$

where μ is the band index (1 for conduction, 2 for valence) and $n(m)$ is the site index on a regular lattice. We confine ourselves to the case of site-diagonal randomness, assuming a joint Gaussian distribution of the two energies $(\epsilon_{n1}, \epsilon_{n2})$ on each site. The parameters which characterize the randomness are $W_{\mu} = \sqrt{\langle (\epsilon_{n\mu} - \langle \epsilon_{n\mu} \rangle)^2 \rangle}$, the mean

deviations, and $\gamma = \langle (\epsilon_{n1} - \langle \epsilon_{n1} \rangle) (\epsilon_{n2} - \langle \epsilon_{n2} \rangle) \rangle / (W_1 W_2)$, the correlation coefficient, where $\langle \dots \rangle$ denotes an ensemble average. In particular, $\gamma = +1$ and -1 correspond to parallel and antiparallel fluctuations of site-energies, respectively.

III. Two-particle CPA

When $\gamma \neq 0$, the averaged two-particle (electron-hole) Green function $\langle g_1 g_2 \rangle$ required for absorption spectra cannot be decoupled into $\langle g_1 \rangle \langle g_2 \rangle$. That is, the vertex correction arises. If we intend to apply the CPA to the two-particle Green function, the vertex correction must be counted in the spirit of the original one-particle CPA. This two-particle extension of the CPA was performed [4] by Velicky in his paper on the conductivity of binary alloys. The procedure in our case is: First, the coherent potential for each band is determined by the usual self-consistent condition $\langle t_\mu \rangle = 0$, where t_μ is the single site scattering t-matrix. Next, the two-particle Green function is obtained by solving the Bethe-Salpeter equation with the fully site-diagonal irreducible part

$$\Lambda = \frac{1}{\langle t_1 t_2 \rangle - 1 + G_1 G_2}, \quad (2)$$

where G_μ 's are the site-diagonal elements of the one-particle Green functions.

IV. Results and Discussion

Let us rewrite the second term in the parentheses of eq.(1) into the k-representation form $\sum_k E_\mu(k) a_{k\mu}^* a_{k\mu}$ and assume a simplified model of the band structure $E_\mu(k)$: (i) semi-elliptic density of states $\frac{1}{N} \sum_k \delta(E - E_\mu(k)) = \frac{2}{\pi B_\mu} \sqrt{B_\mu^2 - E^2}$ ($-B_\mu \leq E \leq B_\mu$) and (ii) similarity in dispersion of the two bands, i.e., $E_2(k) = \mp \frac{B_2}{B_1} E_1(k)$, where the minus sign corresponds to a direct gap and the plus sign, to an indirect gap.

For the moment, we confine ourselves to the case of the uncorrelated randomness ($\gamma = 0$). The absorption band broadens with increasing randomness, as shown in Fig.(1). In the case of the direct gap, the calculated spectra show nearly quadratic energy dependence at the absorption edge (Fig.(2)). This absorption edge reflects the convolution of the density of states of the band edges because of the relaxation of the k-selection rule. In the case of the indirect gap, the relaxation of the k-selection rule causes Lorentzian broadening of the absorption band into the originally forbidden region (Fig.(1b)).

Another important feature of the calculated spectra is the low energy tail of the edge. As shown in Fig.(3), the exponential energy dependence $I(E) \propto \exp(-\frac{E}{T})$ holds fairly well over several decades of the absorption intensity as far as the strength of the randomness is small compared with the band widths. This exponential absorption tail results from the exponential tails of the density of states due to an interplay between the transfer energy and the Gaussian-distributed site-energies, essentially the same mechanism as was considered [5,6] for the temperature dependent Urbach tail of the exciton absorption band in crystals (the thermal distribution of the energy of the localized exciton coupled linearly to harmonic lattice vibrations is necessarily a Gaussian). In this way, the

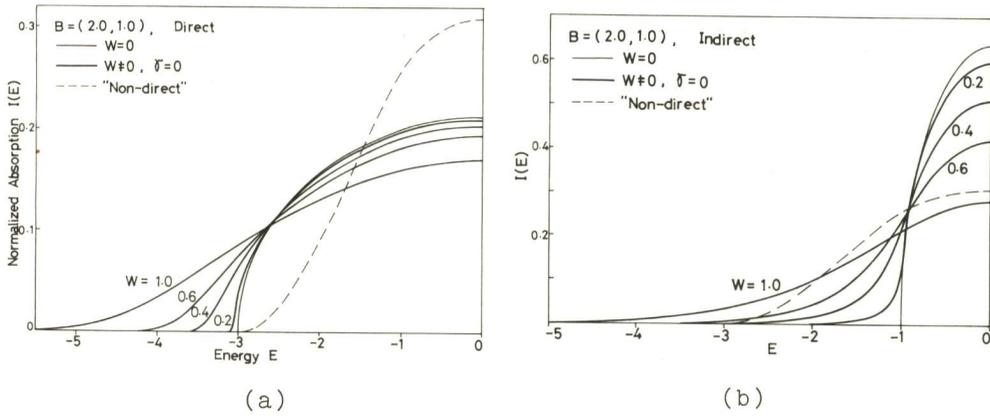


Fig.1 Interband absorption spectra calculated for $B_1=2, B_2=1$ and $\gamma=0$ with $W_1=W_2=W$ as a parameter in the case of (a) direct gap and (b) indirect gap: Broken curves demonstrate the convolution of the density of states of the original two bands

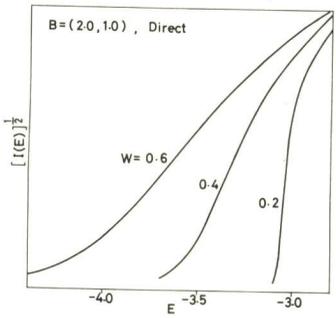


Fig.2 Semi-square-root plot of calculated absorption edges

Urbach rule for random lattice can be derived, on the assumption of Gaussian-distributed energy, both for the exciton and the band-to-band transition, and hence, presumably, irrespective of the magnitude of the electron-hole binding energy. The Gaussian distribution of potential energy can be expected, because a large number of sources of randomness may contribute to the potential.

The tailing parameter Γ of the exponential tail is approximately proportional to

$$E_{char.} = \max\left(\frac{W_1^2}{B_1}, \frac{W_2^2}{B_2}\right),$$

with a proportionality coefficient $0.3 \sim 0.7$. $E_{char.}$ characterizes also the extent of the relaxation of the k-selection rule (the width of the Lorentzian broadening).

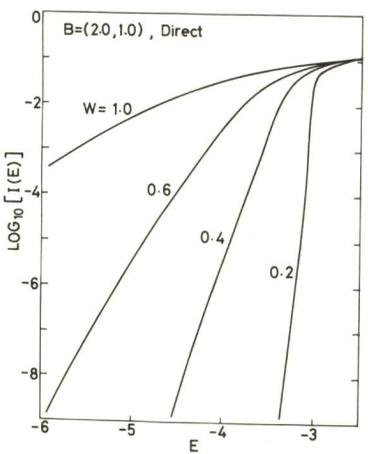


Fig.3 Semi-logarithmic plot of calculated absorption edges

We now turn to the effect of the interband correlation of randomness. The spectra calculated for three cases $\gamma=+1, 0$ and -1 at a fixed value of W are given in Fig.(4). A positive correlation narrows the absorption band, while a negative one broadens it. By comparing Fig.(4a) with Fig.(1a), the effect can better be considered as a parallel shift of the edge: a blue shift by a positive correlation and a red shift by a negative correlation. It should be emphasized that there are practically no changes in the tailing parameter Γ of the exponential tail (see Fig.(4b)).

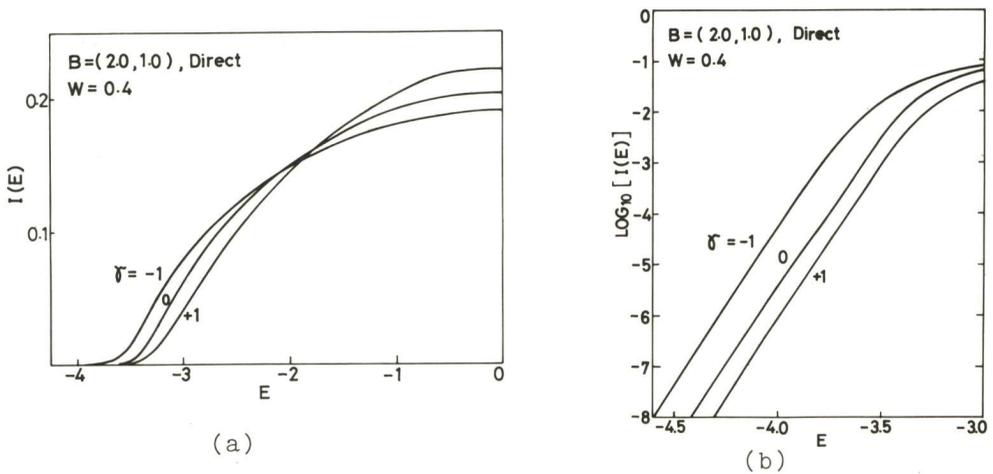


Fig.4 Calculated absorption spectra for a fixed value of W with γ as a parameter in the case of direct gap: (b) is the semi-logarithmic plot of the tail part of the spectra in (a)

In chalcogenide glasses, sufficient irradiation of a sample by band gap light causes a red shift of its absorption edge [7-9]. This phenomenon is known as the photo-structural change. The above-mentioned result suggests that a part of the photo-structural change may be associated with a negative interband correlation of randomness, i.e., an increase in the antiparallel component of potential fluctuations. This negative correlation can be understood as follows: Before a photo-created pair of an electron and a hole recombines (most probably at the same site, say, n), they will distort the surrounding lattice so as to stabilize themselves, namely, to decrease ϵ_{n1} and increase ϵ_{n2} , which contributes to negative correlation. After the electron-hole pair has recombined, this lattice distortion with its negative correlation will be frozen in some metastable configuration [9] because of the freedom characteristic of amorphous networks.

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