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PHYSICS

1967

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Abstract

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UDC 539.293:535+535.215.1

PHYSICS

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ON THE RELATIVE ROLE OF DIRECT AND INDIRECT TRANSITIONS IN OPTICAL ABSORPTION

(Presented by Academician A. A. Lebedev on 23 IV 1966)

For photons of energy $\hbar\omega$, the contribution to the optical conductivity $\sigma(\omega)$ of direct transitions between a pair of bands n and s, separated by a direct energy gap E_g, neglecting broadening effects, is determined by the expression

$$\omega\sigma_d(\omega) = C\int \frac{2}{(2\pi)^3}|M_{ns}(\mathbf{k})|^2\delta(\omega_{ns}(\mathbf{k})-\omega)\,d^3k, \tag{1}$$

where C is a constant; $M_{ns}(\mathbf{k})$ is the matrix element for the direct transition of an electron from the state with energy $E_s(\mathbf{k})$ to the state $E_n(\mathbf{k})$; $\hbar\omega_{ns} = E_n(\mathbf{k}) - E_s(\mathbf{k})$. The integration is carried out over the Brillouin zone. Theoretical calculations (^{1,2}) show that $M_{ns}(\mathbf{k})$ depends only weakly on $\mathbf{k}$, and when integrating expression (1) it may, to a good approximation, be assumed that $|M_{ns}|^2 = M_d^2 = \text{const}$. Then

$$\omega\sigma_d(\omega) = CM_d^2\rho_{ns}(\omega), \tag{2}$$

where $\rho_{ns}(\omega)$ is the joint density of states for the pair of bands under consideration. The quantity ρ_{ns} has a discontinuity at critical points $\mathbf{k}_c$ satisfying the condition $\nabla_k\omega_{ns}(\mathbf{k}_c) = 0$, and thereby determines the entire structure in the optical spectra beyond the absorption edge (^{1}). This idea has proved very fruitful and in a short time has led to significant advances in understanding the optical properties, as well as the band structure, of solids over a broad energy interval (^{3}).

Such a generally accepted interpretation scheme assumes that the magnitude of absorption is also determined by direct transitions with conservation of $\mathbf{k}$. The contribution of indirect transitions is usually considered negligibly small and is not taken into account in calculations. Here we present some considerations which show that the latter assertion is apparently not always valid, and that cases are possible in which the role of indirect transitions may be significant or even predominant.

Without specifying for the time being the detailed mechanism of the indirect absorption process, let us assume that the matrix element for indirect interband transitions, as in the case of direct transitions, is a weak function of $\mathbf{k}$. Then the contribution of indirect transitions between bands n and s to the optical conductivity is approximately expressed in the form

$$\omega\sigma_i(\omega) = CM_i^2 \int \rho_n(E_s + \hbar\omega)\rho_s(E_s)\, dE_s, \tag{3}$$

where M_i^2 is the square of the matrix element for indirect transitions, averaged over all states of the pair of bands under consideration; ρ_n and ρ_s are the densities of states in the corresponding bands, and the integration is performed over the initial states.

From a comparison of expressions (2) and (3) it is seen that, despite the large difference in the magnitudes M_d^2 and M_i^2, a comparison of the contributions of direct and indirect transitions is in principle permissible because of the quadratic dependence of the second on the densities of states. Physically this result can be understood as follows. For a fixed value of $\hbar\omega$ in indirect

in transitions all states contained in an energy band of width $(2\hbar\omega - E_g)$ are able to participate, whereas the number of states separated by a direct energy interval $\hbar\omega$ is very limited. As $\hbar\omega$ increases, the difference in the number of states for direct and indirect transitions will grow and may compensate the difference in the probabilities of elementary transitions M_d^2 and M_i^2. Thus there may exist a value $\hbar\omega = \hbar\omega_t$ at which the contributions of direct and indirect transitions to the optical absorption become equal. An approximate value of the threshold $\hbar\omega_t$ is conveniently estimated from the condition that the areas under the curves $\omega\sigma_d$ and $\omega\sigma_i$ in the range from E_g to $\hbar\omega_t$ are equal. Assuming, for simplicity, $\rho_n = \rho_s = \rho_0 = \text{const}$, one easily obtains

$$\hbar\omega_t \approx E_g + \frac{N}{\rho_0}\frac{M_d^2}{M_i^2}, \tag{4}$$

where N is the ratio of the number of states participating in direct transitions to the total number of states in the band $(2\hbar\omega_t - E_g)$. The use of realistic models for the densities of electronic states in the bands, under the assumption of constant matrix elements, has little effect on the result. Since ρ_0 is usually very large, while N may be much less than unity, $\hbar\omega_t$ may not greatly exceed the magnitude of the forbidden gap E_g. In this case, for all values $\hbar\omega \to \hbar\omega_t$, the absorption may be due mainly to the indirect excitation process. Then, as is evident from formula (3), the structure in the densities of band states will be reproduced in the optical spectra, creating in them the principal structure, whereas the fine structure of the spectra is due to transitions at critical points.

A consideration analogous to that given above makes it possible to conclude that, in photoelectron emission as well, there may exist a threshold for indirect transitions that differs little from the magnitude of the photoelectric work function. From this point of view, the recent work of Spicer and co-workers on measurements of photoemission in a number of semiconductors (^{4}) and metals (^{5}) is of great interest. The results of the measurements are well explained on the basis of the assumption that, in all the materials studied, external photoelectrons are produced predominantly by an indirect absorption process. Thus, for example, the densities of the initial and final states calculated by the authors under this assumption from the energy distributions of emitted electrons agree well with the results of available band calculations. The most important result of Spicer for us is that calculations of the optical constants carried out by formula (3), using the densities of states found from photoemission, reproduce the main structure in optical spectra measured independently by other authors. This result cannot be explained by reference to specific features of photoemission. Consequently, under certain conditions, conservation of k is not an essential selection rule for optical transitions in general. The question of the possibility of such conditions must be answered by a detailed quantitative calculation taking into account the real band structure and the various mechanisms of nonconservation of k in optical transitions.

We express our gratitude to K. Ya. Shtivelman and B. I. Beglov for discussion of the questions touched upon here.

Received
18 IV 1966

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Note: Figure translations are in progress. See original paper for figures.

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