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LIGHT ABSORPTION BY ELECTRONS OF NONMETALLIC CRYSTALS IN AN ELECTRIC FIELD

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The effect is determined of a strong electric field on the optical absorption coefficient due to the conduction electrons of a nonmetallic crystal (semiconductor, insulator).

THERE are several mechanisms for the absorption of infrared radiation in semiconductors: 1) intrinsic absorption; 2) absorption due to local states; 3) lattice absorption; 4) free-carrier absorption.[1] Recently attention has been given to the effect of external fields on these mechanisms, e.g., on the intrinsic absorption.[2,3] This interest is due in particular to the fact that semiconductor devices (p-n junctions, transistors) operate under conditions which create strong electric fields in them. However, no theory has yet been given which takes into account the effect of a field on the remaining three mechanisms. In connection with the prospective use of semiconductors to amplify and generate infrared radiation,[4] the carrier absorption mechanism merits particular study, since it can be a competing process.

In the absence of a field, the mechanism under consideration consists of the absorption of a photon and the transition of an electron between levels of a single conduction band (in an n-type semiconductor). However, by virtue of the selection rules,[1] a direct transition in a single band is forbidden, so that it only occurs through intermediate states, with the participation of lattice vibrations or impurity centers. From the viewpoint of perturbation theory this process is of the second order. However, because the selection rules mentioned are determined by the wave functions of the electrons in the perfect lattice, it can be anticipated that they cease to apply when the wave functions are significantly changed owing to the strict periodicity of the lattice field being destroyed. The character of the optical absorption by the electron should also change. We shall consider changes of this type, assuming that the periodicity is destroyed by an external homogeneous electric field. As a result, we will show that under defined conditions a sufficiently strong field can so change the electron states that direct transitions (in the first order of perturbation theory), with simultaneous absorption (emission) of a photon, become possible between levels of the conduction band as altered by the field.

It is convenient to expand the wave functions in terms of Wannier functions:[5]

$$\psi(r) = \sum_{n,m} \Phi_n(\mathbf{r}_m)\, w_n(\mathbf{r}-\mathbf{r}_m), \qquad (1)$$

where $w_n(\mathbf{r}-\mathbf{r}_m)$ is the Wannier function of the n-th band localized at the lattice site $\mathbf{m}(l_x, l_y, l_z)$. The coefficients Φ_n are determined from the system of equations[5]

$$(E_n^0(\mathbf{p}) + \mathbf{rF} - E)\,\Phi_n = -\sum_{n'\neq n} \mathbf{F}\mathbf{r}_{nn'}\Phi_{n'}. \qquad (2)$$

Here F/e is the electric field strength, $\mathbf{r}_{nn'}$ are the coordinate matrix elements, and $E_n^0(\mathbf{p})$ is the operator obtained from the expression for the energy in the n-th band as a function of the quasimomentum in the absence of the field, by replacing $\hbar\mathbf{k}$ by the momentum operator p.

In the zero-order approximation, we neglect the effect of the energy bands on one another by discarding the right-hand side of (2). This means that we ignore the Zener effect,[6] which occurs in very large fields of the order of 10^6 v/cm. Of the equations obtained for the separate bands, we will consider below only the equation for the conduction band.

In order to determine Φ_n and E, an explicit form of $E_n^0(\hbar\mathbf{k})$ is needed, which forces us to use an approximate model. With a view to revealing the conditions under which the effect of the field on the states will be significant, we will use the "strong coupling" approximation,[7] which, for a simple cubic lattice, gives:

$$E_n^0 = E_n - 2\alpha_n \sum_{i=1}^{3} \sin^2\left(\frac{a_i k_i}{2}\right), \qquad (3)$$

where $\mathbf{a}_i$ are the primitive translations, $2\alpha_n$ is the width of the band. Limiting the summation in (3) to nearest neighbors only corresponds to assuming that the band is narrow.[7] If we put

$$\Phi_n(r) = \int \varphi(p) \exp(-i\,\mathbf{rp}/\hbar)\, d\tau_p,$$

then we obtain from (2)

$$\varphi(p) = A \exp\left[\frac{i}{\hbar}\sum_{s=1}^{3}\left(\frac{C_s p_s}{F_s} - \alpha_n \frac{\sin ak_s}{aF_s}\right)\right]. \tag{4}$$

Using the series expansion of $\exp(-iz\sin\varphi)$ in terms of Bessel functions,[8] we reduce Φ_n to the form

$$\Phi_n(r) = AJ_{k'}(-\alpha_n/aF_x)\, J_{k''}(-\alpha_n/aF_y)\, J_{k'''}(-\alpha_n/aF_z),$$

$$k' = C_1/aF_x + x/a, \qquad k'' = C_2/aF_y + y/a,$$
$$k''' = C_3/aF_z + z/a. \tag{5}$$

The requirement that the wave function be finite can only be satisfied when J_k is a Bessel function of the first kind, and k', k'', k''' are whole numbers. The energy spectrum of the electrons in the band then becomes

$$E = E_n - 3\alpha_n + \sum_i aF_i s_i, \tag{6}$$

where s_1, s_2, s_3 are whole numbers replacing C_i/aF_i, and the normalized wave function is

$$\psi_{s_1s_2s_3}(r) = \sum_{l_i} J_{s_x-l_x}\left(\frac{-\alpha_n}{aF_x}\right) J_{s_y-l_y}\left(\frac{-\alpha_n}{aF_y}\right) J_{s_z-l_z}\left(\frac{-\alpha_n}{aF_z}\right)$$
$$\times w_n(\mathbf{r}-\mathbf{r}_m). \tag{7}$$

Since the external field does not change the number of states in the band, the limits of the variables s_i are given by the relationships

$$-N_i/2 \leqslant s_i \leqslant N_i/2, \tag{8}$$

where N_i is the number of elementary cells in the crystal in the i axis direction.

The wave function (7) and the discrete spectrum (6) which have been obtained correspond to finite motion of the electron. We emphasize that this result is essentially related to the assumption that transitions of electrons from one band to another can be neglected. The possibility that the energy of an electron in the crystal in a homogeneous field can be quantized has been discussed under similar assumptions by I. Lifshitz and Kaganov, using a different method,[9] for an arbitrary dispersion law $E_n(\hbar\mathbf{k})$ as applied to metals. This quantization cannot, however, give observable effects in metals. In fact, for the quantum character of the states to be displayed, it is necessary that the amplitude of the periodic (in the quasi-classical sense) motion, which is of the order $2\alpha_n/F$, shall be smaller than the mean free path. Even for large current densities $j = 10^2$ amp/cm^2, the attainable fields F/e in metals amount to $10^{-8} - 10^{-6}$ cgs esu, so that $2\alpha_n/F \approx 10^4$ cm, which greatly exceeds the mean free path.[9] In semiconductors and insulators fields of $F/e \approx 10^4$ cgs esu can be realized when the energy gap separating off the conduction band is sufficiently large. If, moreover, the width of the band is small, so that, for example, $2\alpha_n \sim 0.1$ ev, then $2\alpha_n/F$ is 10^{-6} cm. The mean free path in a number of semiconductors (Ge, Si) attains $10^{-5}-10^{-4}$ cm, which greatly exceeds the amplitude of the periodic motion.[10]

Assuming that all the required conditions are satisfied in our model, we will calculate the optical absorption coefficient due to electrons of the band. The interaction of an electron with light is given in the coordinate representation (1) by the operator

$$\delta V = e\,\mathbf{Ap}/mc, \tag{9}$$

where $\mathbf{p}$ is the momentum operator, $\mathbf{A}$ is the vector potential:

$$\mathbf{A} = (\Omega\varepsilon)^{-1/2}\sum_{\sigma,t} q_\sigma \xi_{\sigma t} \exp(i\,\sigma\mathbf{r}), \tag{10}$$

where σ is the photon wave vector, $\xi_{\sigma t}$ are the unit polarization vectors, and Ω is the volume of the crystal. The matrix element of the transition from the state s_x, s_y, s_z to the state s'_x, s'_y, s'_z is

$$\langle\psi_{s'}|\,\delta V\,|\psi_s\rangle = \frac{e}{mc\sqrt{\Omega\varepsilon}}\sum_{\sigma,l,l'} q_\sigma \prod_{i=1}^{3} J_{s_i-l_i}\left(\frac{-\alpha_n}{eF_i}\right) J_{s'_i-l'_i}\left(\frac{-\alpha_n}{aF_i}\right)$$
$$\times \int w_n(\mathbf{r}-\mathbf{r}_l)(\xi_\sigma\mathbf{p})\, e^{i\sigma\mathbf{r}} w_n(\mathbf{r}-\mathbf{r}_{l'})\, d\tau. \tag{11}$$

The integral in (11) admits the following transformation:

$$\int w_n(\mathbf{r}-\mathbf{r}_l)(\xi_\sigma\mathbf{p})\, e^{i\sigma\mathbf{r}} w_n(\mathbf{r}-\mathbf{r}_{l'})\, d\tau$$
$$= e^{i\sigma\mathbf{r}_l}\int w_n(\boldsymbol{\rho})(\xi_\sigma\mathbf{p})\, e^{i\sigma\mathbf{p}} w_n(\boldsymbol{\rho}-(\mathbf{r}_{l'}-\mathbf{r}_l))\, d\tau.$$

Since the region in which the function $w_n(\rho)$ is localized has linear dimensions of the order of the lattice constant, a, but the wavelength of light $\lambda \gg a$, then we put $e^{i\sigma\cdot\rho} = 1$ in the integral. The matrix element (11) can only be non-zero if account is taken of the overlapping of the Wannier functions for neighboring cells.

Taking the cubic symmetry into account, we obtain the relations

$$A_0 = \int w_n(\boldsymbol{\rho})\, p_x w_n(\boldsymbol{\rho}\pm\mathbf{a}_1)\, d\tau = \int w_n(\boldsymbol{\rho})\, p_y w_n(\boldsymbol{\rho}\pm\mathbf{a}_2)\, d\tau$$
$$= \int w_n(\boldsymbol{\rho})\, p_z w_n(\boldsymbol{\rho}\pm\mathbf{a}_3)\, d\tau;$$

$$A_1 = \int w_n(\boldsymbol{\rho})\, p_x w_n(\boldsymbol{\rho}\pm\mathbf{a}_2)\, d\tau = \int w_n(\boldsymbol{\rho})\, p_x w_n(\boldsymbol{\rho}\pm\mathbf{a}_3)\, d\tau. \tag{12}$$

The fundamental formula determining the absorption coefficient μ is

$$\mu = \overline{P}\sqrt{\varepsilon}/c, \tag{13}$$

$$\overline{P} = \frac{2\pi}{\hbar}\sum_{s,\, s'} |\langle\psi_{s'}|\delta V|\psi_s\rangle|^2 \rho(E_{s'}) f(E_s) \tag{14}$$

where $\overline{P}$ is the total absorption probability, calculated for 1 sec, $f(E_s)$ is the electron distribution function (Maxwell-Boltzmann) normalized to the number of electrons N, ϵ is the optical dielectric constant, and $\rho(E)$ is the density of electronic states on an energy scale.

Because of the complexity of (11), calculation using formula (14) presents great difficulty for an arbitrary flux direction. We therefore evaluate μ for the particular case when the light flux is directed along the x axis of the crystal. We will also assume that the radiation is unpolarized, so that

$$\overline{\xi}_{\sigma z} = \overline{\xi}_{\sigma y} = 0, \qquad \overline{\xi^2_{\sigma y}} = \overline{\xi^2_{\sigma z}} = \frac{1}{2}.$$

In the optical frequency region considered, $\omega \approx 10^{14}$ and for all attainable fields $\hbar\omega \gg |\alpha_n \sin(\sigma a/2)|$. With these assumptions we obtain

$$\mu = \frac{2\pi ne^2 (A_0 + 2A_1)^2}{m^2 c\omega \sqrt{\varepsilon}\, aF} J^2_{\hbar\omega/\alpha F}\left(\frac{\alpha_n}{aF}\sin\frac{\sigma a}{2}\right) \tag{15}$$

$(n = N/\Omega)$. Because also $\sigma\alpha \ll 1$, then when $\alpha_n \sim 0.1 - 1$ ev we have, for all frequency ranges, $\hbar\omega \gg aF$. This inequality permits us to use an asymptotic form of the function $J_p(x)$,[8] and to bring (15) to the final form

$$\mu = \frac{ne^2 (A_0 + 2A_1)^2}{mc \sqrt{\varepsilon}\, \hbar\omega^2} (1 - e^{-\hbar\omega/kT}) e^{-F_0(\omega)/F},$$

$$F_0(\omega) = \frac{\hbar\omega}{2a} \ln\left(\frac{\hbar\omega}{\alpha_n \sin(\sigma a)}\right). \tag{16}$$

The multiplier $(1 - e^{-\hbar\omega/kT})$ takes the induced radiation into account. For small fields $(F < F_0)$, the absorption μ is small, and disappears as $F \to 0$. For fields $F > F_0$, the quantity μ tends to saturation. It is of interest to note that the frequency dependence of the absorption μ is the inverse of that which was observed under the experimental conditions used by V. Vavilov and Britsyn[3] where the intrinsic absorption mechanism apparently dominated: with increasing frequency the absorption gets smaller. This fact can serve to separate the contribution of free carriers to the absorption in a field from other contributions in the total absorption coefficient.

We remark finally that the effect discussed should be expected in semiconductors with impurity bands the width of which is small, and that at sufficiently low temperatures the applicability conditions of the calculations given improve.

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