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Role of the various types of transitions in three-photon absorption in InAs

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The frequency dependences of the three-photon absorption (TPA) probability and the linear-circular dichroism (LCD) associated with TPA in InAs are investigated theoretically and experimentally. The role of the various types of transitions involved in TPA is analyzed. The obtained experimental data are in satisfactory agreement with the results of a calculation performed in the two-band model with allowance for both allowed-allowed-allowed and allowed-forbidden-forbidden transitions.

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1. The observation of three-photon absorption (TPA) of CO_2-laser radiation in indium arsenide is reported in Ref. 1. During the study of the polarization characteristics of the TPA probability, there was observed—in accord with the predictions made in Refs. 2 and 3—a large linear-circular dichroism (LCD), $\Lambda = W_l^{(3)}/W_c^{(3)}$, near the TPA edge ($W_{l,c}^{(3)}$ are the TPA probabilities for linear and circular polarizations, respectively). The theoretical results of Refs. 2 and 3 were obtained in the lowest approximation in the parameter $\eta = (3\hbar\omega - E_g)/\hbar\omega \ll 1$ (E_g is the forbidden-band width) with allowance for only the allowed-allowed-allowed (A-A-A) transitions. In the indicated approximation Λ is frequency independent. Experiment[1] has, however, shown that a small increase in the crystal temperature (299–307 K) leads to a sharp decrease in the magnitude of Λ_{exp}. It is natural to assume that such a character of the dependence $\Lambda(T)$ is connected with the temperature variation of $E_g(T)$ and the strong dependence of the LCD near the TPA edge on the excess energy $(3\hbar\omega - E_g)$. In view of this, it was of interest to calculate the quantities $W_l^{(3)}$ and $W_c^{(3)}$ in the case of an arbitrary value of η, experimentally investigate the frequency dependences of the TPA probability and the magnitude of the LCD in InAs, and, comparing the theoretical and experimental results, analyze the relative contributions of the various types of transitions involved in TPA.

2. In the two-band model three-photon transitions are forbidden in A_3B_5 crystals for circularly polarized light in the case when $3\hbar\omega \approx E_g$.[2] This is due to the impossibility of changing by more than two units a component of the angular momentum of an electron in an interband transition in the vicinity of the $k=0$ point. If we do not restrict ourselves to the two-band model, and take into consideration transitions with intermediate states in other bands, then the indicated forbiddenness is, generally speaking, lifted. In the three-band model, in which, besides the conduction band c and three valence subbands v_i $(i=1,2,3)$, the higher-lying conduction band $\bar{c}$ is taken into consideration for A-A-A transitions, i.e., in the lowest approximation in the parameter $\eta \ll 1$, the expression for the TPA probability has the following form:1) [3]

$$W^{(3)}(\mathbf{e},\omega) = W^{(1)}(3\omega)\frac{\sigma^3(\omega)}{\sigma(3\omega)}\sum_\alpha [\,|a_1(\mathbf{ee})e_\alpha + a_2 e_\alpha^3|^2 + {}^3/_4|a_3 e_\alpha(e_{\alpha+1}^2 - e_{\alpha+2}^2)|^2], \quad (1)$$

where

$$a_1 = \frac{1}{2\hbar\omega}\left[-\frac{P_{cv}^2}{E_g-\hbar\omega}\frac{\Delta+4\hbar\omega}{2\hbar\omega(\Delta+2\hbar\omega)} + \frac{P_{\bar{c}c}^2}{E_{\bar{c}c}-2\hbar\omega}\right] - a_2 - \frac{1}{2}a_3,$$

$$a_2 = \frac{P_{\bar{c}v}^2}{E_{\bar{c}v}-\hbar\omega}\frac{\Delta+4\hbar\omega}{2\hbar\omega(\Delta+2\hbar\omega)},$$

$$a_3 = \frac{P_{\bar{c}v}^2}{E_{\bar{c}v}-\hbar\omega}\frac{\Delta}{3\hbar\omega(\Delta+2\hbar\omega)},$$

$W^{(1)}$ is the one-photon absorption probability, $\mathbf{e}$ is the light-polarization vector, $\sigma(\omega)=2\pi\hbar e^2 j/\omega m^2 c\varkappa^{1/2}$, j is the light intensity in kW/cm^2-sec, $\varkappa$ is the high-frequency permittivity, and α assumes the values x, y, z.

In the case when the laser radiation propagates along one of the principal axes ([001]) of the crystal and the polarization vector of the linearly polarized light is parallel to one of the other two axes ([100] or [010]), the magnitude of the LCD is, according to (1), equal to

$$\Lambda=4(a_1+a_2)^2/(a_2^2+{}^3/{}_4a_3^2). \tag{2}$$

In InAs the $\bar{c}$ band is 3.9 eV above the top of the valence band.[4] This is greater by an order of magnitude than the quantity $E_g\approx 0.35$ eV. Therefore, the contribution of the transitions in which the $\bar{c}$ band participates should be small. The indicated conclusion can be verified by estimating the quantity Λ from the formula (2). The matrix elements P_{cv}, $P_{\bar{c}v}$, and $P_{\bar{c}c}$ were computed within the framework of the $\mathbf{k}\cdot\mathbf{p}$ method in the three-band model with the use of experimental values for the effective masses of the electrons and holes.[5] The value thus obtained for Λ turned out to be ~5×10^3 for the following values of the effective electron and hole masses:

$$m_c=0.024m_0, \quad m_{vl}=0.026m_0, \quad m_{vh}=0.41m_0.$$

Consequently, the relative contribution of the transitions with intermediate states in the $\bar{c}$ band is indeed small, and, in computing the TPA probability, we can restrict ourselves to the two-band model.

The greatest value obtained for Λ in an experiment under conditions that approximately corresponded to the TPA edge turned out to be equal to 27. To explain the discrepancy between Λ_{theor} and Λ_{exp}, as well as the extremely critical dependence of Λ_{exp} on $3\hbar\omega-E_g$, let us take into account the fact that for $k\neq 0$ the forbidden transitions can play an important role.

3. Within the framework of the two-band model, besides A-A-A transitions of the v-c-v-c type (for linearly polarized light), allowed-forbidden-forbidden (A-F-F) transitions of the type v-v-v-c, v-v-c-c, and v-c-c-c, which can occur for both linear and circular polarizations of the radiation, also contribute to the TPA, since for A-F-F transitions the excess angular momentum that arises in the absorption of three circularly polarized photons is transferred to the intraband motion of the electrons. As the quantity $3\hbar\omega-E_g$ increases, the role of the A-F-F transitions becomes more important, and the value of Λ should decrease, since the contribution to $W^{(3)}$ of the A-F-F transitions near the TPA edge is proportional to k^5 and becomes comparable to the contribution from the A-A-A transitions when there is even a small departure from the edge.

Let us give the results of the calculation of the dependences $W^{(3)}_{l,c}(\hbar\omega)$ that takes both the A-A-A and the A-F-F transitions into account. The calculation was carried out in the Kane model with allowance for the nonparabolicity. In this case we considered, for simplicity, the two limiting cases: $\Delta\gg E_g$ and $\Delta=0$ (Δ is the magnitude of the spin-orbit splitting of the valence band). Since the dependences $\Lambda(\hbar\omega)$ for the indicated cases turned out to be similar, the obtained results are also applicable when $\Delta\sim E_g$ (in InAs the quantity $\Delta=0.43$ eV).

For $\Delta\gg E_g$ or $\Delta=0$ the energy spectrum of the electrons and the matrix elements of the interband and intraband optical transitions and, hence, the TPA probability can be computed for an arbitrary value of the parameter η. In particular, for $\Delta\gg E_g$ the expressions for $W^{(3)}_l$ and $W^{(3)}_c$ have the following form:

$$W_l^{(3)}=A\left(\frac{x-1}{x}\right)^{1/2}[f_1(x)(24a^2+3b^2+8ab+4c^2)+f_2(x)(4d^2+6dp+7{,}5p^2+24q^2+8qs+3s^2)], \tag{3}$$

$$W_c^{(3)}=A\left(\frac{x-1}{x}\right)^{1/2}\{f_1(x)[4(a-b)^2+10c^2]+f_2(x)[3(d-p)^2+4(q-s)^2]\}, \tag{4}$$

where

$$A=\frac{81\sqrt{6}}{140\pi}\left(\frac{2\pi e^2 j}{\omega m_0 c\varkappa^{1/2}}\right)^3\frac{P_{cv}^3}{\hbar E_g^2}, \quad x=\frac{3\hbar\omega}{E_g},$$

$$f_1(x)=[(4x-3)(5x-3)]^{-2}, \quad f_2(x)=\frac{1}{972x^6}\left(\frac{x+1}{x}\right)^{1/2},$$

$$a=\frac{15-23x}{24}, \quad b=\frac{20x-21}{3}, \quad c=2(1-x),$$

$$d=\frac{3x}{4}\frac{85x^2-117}{9-x^2},$$

$$p=-\frac{2s}{x}=\frac{3(8x^2-9)}{x}, \quad q=\frac{3x^2}{8}\frac{45-13x^2}{9-x^2}.$$

In Fig. 1 we have plotted $W_{l,c}$ as a function of the quantity x according to the formulas (3) and (4). The nonmonotonic nature of the dependence $W_l(x)$ in the region $1<x<1.2$ is noteworthy.

4. The experimental study of the frequency dependence of $W^{(3)}_l$ and Λ was accomplished in experiments on the photoconductivity associated with three-photon excitation. The measurements were carried out on samples of n-InAs ($n=1.5\times 10^{16}$ cm^{-3}). The source of the exciting radiation was a Q-switched CO_2 laser with a diffraction grating mounted in its resonant cavity. It was then possible to obtain generation at four wavelengths: 10.6, 10.2, 9.5, and 9.2 μ. Since the intensities of the emissions with different frequencies differed from each other, we measured at each wavelength the dependence of the photoconductivity ($\Delta\sigma_l$) on the intensity of linearly polarized pumping radiation (Fig. 2). Then, with allowance for these dependences, the $\Delta\sigma_l$ values obtained at the various wavelengths were reduced to values for one density, and the curve $\Delta\sigma_l(\hbar\omega)$ was plotted (Fig. 3). The indicated dependence should reflect the behavior of $W^{(3)}_l(\hbar\omega)$. In Fig. 3 we show, for

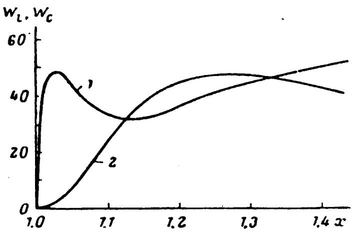

FIG. 1. Dependences on the ratio $x=3\hbar\omega/E_g$ of three-photon absorption probabilities computed for InAs in the two-band model for linear (curve 1) and circular (curve 2) polarizations of the pumping radiation.

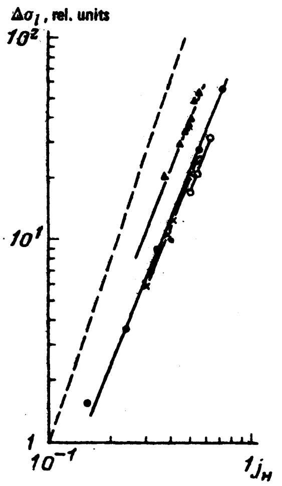

FIG. 2. Dependences, measured in an n-InAs sample ($n=1.6\times10^{16}$ cm^{-3}), of three-photon photoconductivity on the intensity of the linearly polarized pumping radiation: ×—$\lambda=10.6\mu$, ○—$\lambda=9.5\mu$, ●—$\lambda=10.2\mu$, ▲—$\lambda=9.2\mu$. The dashed line corresponds to the relation $(\Delta\delta_l)\sim j^3$.

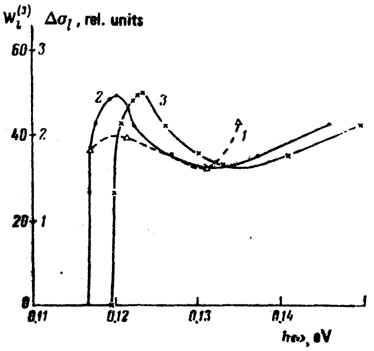

FIG. 3. Frequency dependence of the three-photon photoconductivity in InAs at 300 K (curve 1) and the dependence on $\hbar\omega$ of the TPA probability, $W_l^{(3)}$, constructed for two values of E_g/300 K: 0.350 eV (curve 2) and 0.359 eV (curve 3).

comparison with the experimental points, the dependence $W_l^{(3)}(\hbar\omega)$, plotted for two different values of E_g (300 K).[2)] [5,6] It can be seen that the experimental dependence is qualitatively similar to the theoretical dependence, the agreement being better at $E_g=0.350$ eV (300 K).

The smooth variation of the quantity $x=3\hbar\omega/E_g$, on which the TPA probability depends, could have been realized because of the variation of E_g with temperature. But the variation of temperature leads also to the variation of a number of other parameters that have influence on $\Delta\sigma$, parameters like the lifetime τ_n, the mobility, the dark-carrier concentration, etc. Therefore, a comparison of the dependences $\Delta\sigma_l(x)$ and $W_l^{(3)}(x)$ is difficult in this case. These parameters do not enter into the expression for the magnitude of the LCD (Λ is a function of x only) and, thus, the comparison of Λ_{theor} with Λ_{exp} can be carried out with a minimum number of additional assumptions.

We measured the temperature variation of $\Delta\sigma_l/\Delta\sigma_c=\Lambda_{\text{exp}}$ at two wavelengths (10.6 and 9.5 μ) in the range 298–370 K, and the two dependences were then constructed as functions of x. For the conversion we used the values

$$E_{g0}=0.434\text{ eV [5, 6]},\quad \partial E_g/\partial T=2.8\cdot10^{-4}\text{ eV/deg [6]},$$
$$\partial E_g/\partial T=2.46\cdot10^{-4}\text{ eV/deg [5]}.$$

In Fig. 4 we show the obtained experimental dependences $\Lambda_{\text{exp}}(x)$, together with the theoretical dependence, which was constructed in accordance with the formulas (3) and (4). It can be seen from the figure that the experimental points obtained at the two different wavelengths fall nicely on a monotonic curve. As predicted by theory, near the TPA edge, the quantity Λ_{exp} falls off very sharply with increasing x and then decreases smoothly to unity. As for the frequency dependence of $\Delta\sigma_l$ (Fig. 3), a better agreement with theory is obtained in the case when the values of x are found on the basis of the data on E_g given in Ref. 6. But the two $\Lambda_{\text{exp}}(x)$ dependences turn out all the same to be shifted toward the region of lower values of x as compared to the theoretical curve $\Lambda(x)$. It should also be noted that the behavior of the experimental $\Lambda(x)$ curve near the TPA edge is more critical than follows from the performed theoretical calculation.

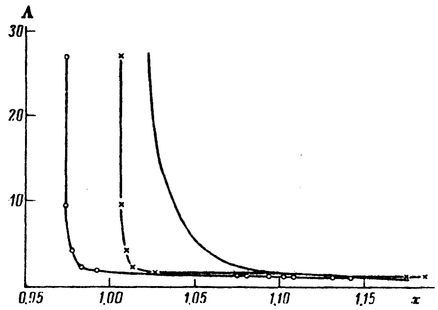

FIG. 4. The dependence $\Lambda(x)$; the continuous curve is a theoretical curve; the points: ○—$\Lambda(x)$ for $\partial E_g/\partial T=2.8\times10^{-4}$ eV/deg, ×—$\Lambda(x)$ for $\partial E_g/\partial T=2.46\times10^{-4}$ eV/deg; $E_{g0}=0.434$ eV.

The above-indicated facts may be connected with the influence of the electron-phonon interaction in the course of the three-photon transition.

In conclusion, let us note that the investigation of the LCD associated with TPA is an extremely convenient technique, and it may turn out to be useful in the analysis of the nature of three-photon transitions in other semiconductors.

1)In the two-band model the constant-energy surfaces for the electrons and holes in the straight-band A_3B_5 semiconductors are spherically symmetric, and the angular-momentum component conservation law is fulfilled in optical transitions. The inclusion of the $\bar{c}$ band in the three-photon transition scheme leads to the explicit allowance for the cubic anisotropy of the crystalline potential, as a result of which the selection rules change.

2)The values of the forbidden-band width, E_g, of indium arsenide (300 K) given in different papers lie in the range 0.31–0.36 eV. For the construction of the dependence $W_l^{(3)}(\hbar\omega)$ we used the most reliable data obtained from experiments on edge absorption and processed with allowance for the nonparabolic nature of the bands,[6] as well as from experiments on magnetoabsorption.[5,7]

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Electromagnetic field absorption in superconducting films.

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The low-frequency dispersion of electromagnetic field absorption, connected with the relaxation of the order parameter and the excitation distribution function, is investigated. It is shown that on passage of a direct current, a sharp peak of the absorption of the high-frequency field near the threshold frequency $\omega = 2\Delta$ appears at a current density much lower than the critical density.

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1. INTRODUCTION

The energy relaxation time τ_ϵ in superconductors is large. Therefore, inelastic collisions can usually be neglected in an approximation that is linear in the alternating field. However, at low frequencies, such neglect is not always possible and can lead, in the dynamic limit as $\omega \to 0$ to a difference from the static limit obtained at $\omega = 0$. In superconducting films, in the presence of a direct current, these two limiting values, for example for the correction to the order parameter, differ from one another by a value of Δ/T close to the transition temperature.[1] As will be shown below, such a difference leads to strong dispersion in the absorption at frequencies $\omega \sim \Delta/T\tau_\epsilon$. Strong dispersion arises also at $\omega\tau_\epsilon \sim 1$.

The low-frequency dispersion is connected with processes of relaxation of the excitation distribution function. Energy relaxation is effected in films only because of interaction of the electrons with phonons.

One more peculiarity arises when current flows in a film. It is known that there is a kink in the absorption at the frequency $\omega = 2\Delta$. At low temperatures, the presence of a weak current $j \ll j_c$ leads to the appearance of a sharp maximum in the absorption at a frequency close to 2Δ.

2. DISPERSION OF THE ABSORPTION AT LOW FREQUENCIES

We limit ourselves below to superconductors with small free path length $l \ll v/T, d$ (d is the film thickness, v is the velocity on the Fermi surface). We also assume that the film thickness d satisfies the condition $d < \xi(T)$, where $\xi(T)$ is the correlation length of the superconductor.

At low frequencies ($\omega \ll \Delta$) it is convenient to describe the properties of the superconductor by means of the kinetic equations.[2,3] Below, we choose a special gauge, in which the scalar potential $\varphi = 0$; the vector potential and the order parameter have the form

$$\mathbf{A}(\mathbf{r}, t) = \mathbf{A}(\mathbf{r}) + \mathbf{A}_1 e^{-i\omega t}, \quad \Delta(\mathbf{r}, t) = \Delta + \Delta_1 e^{-i\omega t},$$

respectively. Of the two distribution functions f and f_1, only the function f differs from zero in the chosen gauge:

$$f = \operatorname{th}(\varepsilon/2T) + \tilde{f} e^{-i\omega t},$$

where

$$-\frac{i\omega}{2}(g^R - g^A)\tilde{f} + I_1^{ph}\left(\operatorname{th}\frac{\varepsilon}{2T} + \tilde{f}\right)$$
$$= -i\omega \frac{\partial}{\partial \varepsilon} \operatorname{th}\left(\frac{\varepsilon}{2T}\right)\left\{\Delta_1 \frac{(F^R - F^A)}{2} - ie^2(\mathbf{A}\mathbf{A}_1)D((g^R)^2 - (g^A)^2)\right\}. \quad (1)$$

In Eq. (1), $g^{R,A}$ and $F^{R,A}$ are the static Green's functions, satisfying the set of equations

$$(g^{R,A})^2 = (F^{R,A})^2 + 1,$$
$$2e^2\mathbf{A}^2 D g^{R,A} F^{R,A} + i\Delta g^{R,A} - i\varepsilon F^{R,A} = 0, \quad (2)$$

where $D = vl_{tr}/3$ is the diffusion coefficient.

The collision integral I_1^{ph} is determined by the expression[2]

$$I_1^{ph}(f) = \frac{i\nu g^2}{16}\int\frac{d\varepsilon_1}{2\pi}\int d\Omega_{p_1}[(g_\varepsilon^R - g_\varepsilon^A)(g_{\varepsilon_1}^R - g_{\varepsilon_1}^A) - (F_\varepsilon^R - F_\varepsilon^A)(F_{\varepsilon_1}^R - F_{\varepsilon_1}^A)]$$
$$\times\{(f(\varepsilon) - f(\varepsilon_1))D_{p-p_1}(\varepsilon_1 - \varepsilon) + (1 - f(\varepsilon)f(\varepsilon_1))[D^R_{p-p_1}(\varepsilon_1 - \varepsilon) - D^A_{p-p_1}(\varepsilon_1 - \varepsilon)]\}. \quad (3)$$

where $\nu = mp/2\pi^2$ is the density of states on the Fermi surface. At thermodynamic equilibrium, the photon Green's functions are

$$D_k^R(\omega) = D_k^{A*}(\omega) = -\omega^2(\mathbf{k})/[\omega^2(\mathbf{k}) - (\omega + i\delta)^2],$$
$$D_k(\omega) = \operatorname{cth}(\omega/2T)[D_k^R(\omega) - D_k^A(\omega)]. \quad (4)$$

We limit ourselves below to the region of tempera-

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