Anisotropy of two-photon absorption in cubic direct-band-gap semiconductors

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The two-photon absorption (TPA) cross section in zinc-blende-structure semiconductors has been analyzed, both analytically and numerically. The warping of the valence band is shown to result in a dependence of the TPA on the orientation of the polarization plane of the light with respect to the crystal axes, i.e., in a linear dichroism. In InSb and GaAs, the relative differences in the TPA cross sections for light polarized along ⟨100⟩ and ⟨111⟩ axes, depending on the light wavelength, are 0.02–0.05 and 0.13–0.20, respectively. The linear-circular dichroism of TPA, even in narrow-band-gap semiconductors, is noticeably influenced by the presence of a split-off valence subband.

I. INTRODUCTION

The two-photon absorption (TPA) cross section in semiconductors has been investigated in a number of papers, both theoretically and experimentally. It is the main mechanism of absorption of sufficiently intense light in the frequency range $\hbar\omega < \varepsilon_g < 2\hbar\omega$ (ω is the light frequency; ε_g is the energy gap). For zinc-blende-structure semiconductors, the most detailed theoretical results on TPA were obtained in the isotropic-band approximation for some sufficiently simple models of the band structure (see, e.g., Refs. 1–4 and references therein). The main characteristics investigated were the frequency dependence of the absorption cross section and the linear-circular dichroism, i.e., the difference in the cross sections for linearly and circularly polarized light.

In the present paper we investigate, both analytically and numerically, the dependence of TPA on light polarization in direct-band-gap cubic semiconductors for realistic models of the band structure. The emphasis is on the dependence of the TPA cross section on the orientation of the polarization plane of the light with respect to the crystal-symmetry axes. Such a dependence is a specifically nonlinear effect. In linear optics, cubic crystals are well known to be isotropic (apart from the effects of spatial dispersion) and, in particular, one-photon absorption is independent of light polarization. For TPA the situation is absolutely different. It follows from the symmetry arguments that in the general case the decrease of the light intensity $I=(cn/2\pi)|\mathbf{E}|^2$ [the electric field strength $\mathcal{E}(z,t)=\mathbf{E}(z)\exp(-i\omega t)+\mathbf{E}^*(z)\exp(i\omega t)$, n is the refractive index] due to TPA is given by the equation

$$-(dI/dz)_{\mathrm{TPA}}=A_1|\mathbf{E}|^4+A_2\mathbf{E}^2\mathbf{E}^{*2}+A_3\sum_{\kappa}|E_\kappa|^4\ ,\quad(\kappa=x,y,z)\ , \tag{1}$$

where E_κ are the projections of $\mathbf{E}$ on the fourfold-rotation-symmetry axes (the ⟨100⟩ axes).

The first two terms in Eq. (1) are not specifically "crystalline"; they refer to TPA in isotropic media as well (cf. Ref. 5 where similar terms in the free energy of radiation in a transparent isotropic medium were demonstrated to give rise to self-induced rotation of the ellipse of light polarization). In the case of TPA these terms result in a difference in the absorption cross sections K_{lin} and K_{circ} for linearly ($\mathbf{E}^*\|\mathbf{E}$) and circularly ($\mathbf{E}^2=0$) polarized light beams, respectively.[1] This difference is called linear-circular dichroism. It is obvious from (1) that

$$K_{\mathrm{lin}}/K_{\mathrm{circ}}=1+A_2/A_1\ ,\quad(A_3=0)\ . \tag{2}$$

The last term in (1) is "pinned" to crystal-symmetry axes and contains a dependence of TPA on the orientation of the electric field $\mathbf{E}$ with respect to these axes, i.e., *a linear dichroism* (although somewhat different from linear dichroism in linear optics, cf. Ref. 6). In particular, the ratio $K_{100}:K_{110}:K_{111}$ of the absorption cross sections for light polarized linearly along the ⟨100⟩, ⟨110⟩, and ⟨111⟩ axes is obvious from (1) to be

$$K_{100}:K_{110}:K_{111}=(A_1+A_2+A_3):(A_1+A_2+\tfrac{1}{2}A_3):(A_1+A_2+\tfrac{1}{3}A_3)\ . \tag{3}$$

Such dichroism gives rise to a specific rotation of the polarization plane of light; this is also a general symmetry feature of nonlinear optics of cubic crystals that was predicted and observed for various mechanisms of optical nonlinearity.[7]

We note that, in contrast to the case of a one-beam TPA considered in the present paper, in the case of absorption of photons with different frequencies from two

polarized beams the TPA cross section depends on the relationship between their polarizations and even in an isotropic medium[8] for fixed polarization of one of the beams, the absorption of the other beam displays a linear dichroism.

In Sec. II below we give an analytic theory of TPA near the absorption threshold ($2\hbar\omega-\varepsilon_g \ll \varepsilon_g$) for large spin-orbit splitting of the valence band and for small but finite warping of the hole constant-energy surfaces. In Sec. III the numerical results for two semiconductors, InSb and GaAs (opposite in relative magnitude of the spin-orbit splitting of the valence band), are given; they have been obtained using the (8×8) anisotropic Kane model. Section IV contains concluding remarks.

II. ABSORPTION CROSS SECTION IN THE PARABOLIC-BAND APPROXIMATION

In the zinc-blende-structure semiconductors for $\hbar\omega<\varepsilon_g<2\hbar\omega$ TPA is due mainly to the valence-band to conduction-band transitions accompanied by light-induced intra-conduction-band or intra- or inter-valence-subband virtual transitions.[1-3] A direct way of calculating the TPA cross section, i.e., the coefficients $A_{1,2,3}$, is based on numerical evaluation of the matrix elements of the transitions and of the electron-dispersion law using the standard for narrow-gap semiconductors formulation[9] of the $\mathbf{k}\cdot\mathbf{p}$ method,

$$A_1|\mathbf{E}|^4+A_2\mathbf{E}^2\mathbf{E}^{*2}+A_3\sum_\kappa|E_\kappa|^4=\frac{4\pi e^4}{m^4\omega^3}\sum_{i,f}\int\frac{d\mathbf{k}}{(2\pi)^3}|M_{if}(\mathbf{k},\mathbf{E})|^2\delta(\varepsilon_f(\mathbf{k})-\varepsilon_i(\mathbf{k})-2\hbar\omega)\ ,$$
$$M_{if}(\mathbf{k},\mathbf{E})=\sum_j(f|\mathbf{E}\cdot\hat{\mathbf{p}}|j)(j|\mathbf{E}\cdot\hat{\mathbf{p}}|i)/[\varepsilon_i(\mathbf{k})+\hbar\omega-\varepsilon_j(\mathbf{k})]\ . \tag{4}$$

Here, m is the free-electron mass, and i and f denote the initial and final states of a light-absorbing electron in the valence and conduction bands, respectively; the states $|i)$, $|f)$ refer to one and the same value of the electron wave vector $\mathbf{k}$. We note that virtual transitions to both occupied and empty electron states $|j)$ are permitted (such transitions correspond simply to mixing of the electron states by the electric field of light). The states $|i)$ and $|f)$ can be interpreted also as the hole and electron states.[3]

To gain insight into the anisotropy of TPA it is important to obtain analytic expressions for the parameters $A_{1,2,3}$. This can be done for

$$2\hbar\omega-\varepsilon_g \ll \varepsilon_g\ , \tag{5}$$

where the energies of the electrons and holes involved are small compared to the energy gap, and thus the effects of nonparabolicity of the dispersion law can be neglected and the dipole matrix element of the valence- to conduction-band transition can be assumed independent of the electron wave vector $\mathbf{k}$. The analytic results in what follows refer to semiconductors with large spin-orbit splitting Δ. For such semiconductors only two valence subbands, those of heavy and light holes, come into play. For $\hbar\omega\ll\Delta$ the virtual transitions via the split-off subband can be ignored and the denominators in (4) can be replaced by either $-\hbar\omega$ [if $|j)$ is a conduction-band state] or $\hbar\omega$ (in the opposite case). The latter means that we neglect $|\varepsilon_h(\mathbf{k})-\varepsilon_l(\mathbf{k})|$ compared to $\hbar\omega$, where $\varepsilon_h(\mathbf{k})$, $\varepsilon_l(\mathbf{k})$ are the energies of heavy and light holes, respectively.

The Hamiltonian of holes is of the form

$$H(\mathbf{k})=\frac{\hbar^2}{m}\left[\tfrac{1}{2}(\gamma_1+\tfrac{5}{2}\gamma_2)\mathbf{k}^2-\gamma_3(\mathbf{k}\cdot\mathbf{J})^2+(\gamma_3-\gamma_2)\sum_\kappa k_\kappa^2J_\kappa^2\right], \tag{6}$$

where J_x, J_y, J_z are the (4×4) matrices of the angular momentum $\frac{3}{2}$, and γ_1, γ_2, γ_3 are the Luttinger parameters. It is the last term in (6) that is anisotropic and is "pinned" to the crystal-symmetry axes, and it is this term that is thus responsible for the linear dichroism of TPA.

For many actual semiconductors the anisotropy is weak,

$$|\gamma_3-\gamma_2| \ll \gamma_3\ . \tag{7}$$

If (7) is fulfilled, the linear dichroism of TPA is small, $|A_3| \ll A_1$. To lowest order in $(\gamma_3-\gamma_2)/\gamma_3$ there are two additive contributions to the A_3 term in (4). The first comes from the anisotropy of the energy of holes in the δ function in (4), i.e., from the anisotropy of the density of states. It can be found by calculating the transition matrix elements $M_{if}(\mathbf{k},\mathbf{E})$ to zeroth order in $(\gamma_3-\gamma_2)/\gamma_3$. The second contribution comes from the anisotropic term in $M_{if}(\mathbf{k},\mathbf{E})$ itself and can be calculated in the neglect of warping of the hole constant-energy surfaces.

To find the coefficients $A_{1,2,3}$ it is convenient first to sum the squared absolute values of the matrix elements $M_{if}(\mathbf{k},\mathbf{E})$ over the degenerate initial and final states, i.e., over orientations of the electron spin in the conduction band and over two degenerate heavy- and light-hole states. To first order in $(\gamma_3-\gamma_2)/\gamma_3$ the corresponding sum over the states with the same $\varepsilon_i(\mathbf{k})$, $\varepsilon_f(\mathbf{k})$ can be written as

$$\sum_{\varepsilon_{i,f}(\mathbf{k})=\text{const}}|M_{if}(\mathbf{k},\mathbf{E})|^2=\tfrac{2}{3}(mPk/\hbar\omega)^2\mu_\eta(\mathbf{n},\mathbf{E})\ ,$$
$$\mu_\xi(\mathbf{n},\mathbf{E})\simeq\mu_\xi^{(0)}(\mathbf{n},\mathbf{E})+\mu_\xi^{(1)}(\mathbf{n},\mathbf{E})\ ,\quad \mathbf{n}=\mathbf{k}/k\ . \tag{8}$$

Here, P is the Kane parameter, $P=-i(\hbar/m)\ (s|\hat{\mathbf{p}}_x|x)$ [$|s)$ and $|x)$ are the orbital conduction- and valence-band wave functions for $k=0$]. The subscript ξ takes on two values, h and l, for the transitions giving rise to the creation of heavy or light hole, respectively. The term $\mu^{(0)}$ in (8) corresponds to the isotropic-band approximation, and $\mu^{(1)}$ gives the anisotropy-induced correction.

A. The "isotropic" matrix elements for two-photon-induced transitions

The calculation of $\mu_{h,l}^{(0)}(\mathbf{n},\mathbf{E})$ is quite straightforward. It is convenient to choose the z axis parallel to $\mathbf{n}=\mathbf{k}/k$ (cf. Ref. 9) and to take the wave functions of the heavy ($J_z=\pm\frac{3}{2}$) and light ($J_z=\pm\frac{1}{2}$) holes in the form

$$\begin{aligned}
&|v,\tfrac{3}{2}\rangle=-\frac{1}{\sqrt{2}}|x+iy\rangle\uparrow\ ,\\
&|v,-\tfrac{3}{2}\rangle=\frac{1}{\sqrt{2}}|x-iy\rangle\downarrow\ ,\\
&|v,\tfrac{1}{2}\rangle=\frac{1}{\sqrt{6}}[2|z\rangle\uparrow-|x+iy\rangle\downarrow]\ ,\\
&|v,-\tfrac{1}{2}\rangle=\frac{1}{\sqrt{6}}[2|z\rangle\downarrow+|x-iy\rangle\uparrow]\ .
\end{aligned} \tag{9a}$$

The wave functions of the conduction band $|c,\alpha\rangle$, $\alpha\pm\frac{1}{2}$, are of the form

$$|c,\tfrac{1}{2}\rangle=i|s\rangle\uparrow\ ,\quad |c,-\tfrac{1}{2}\rangle=i|s\rangle\downarrow\ . \tag{9b}$$

The matrix element of the operator $\mathbf{E}\cdot\hat{\mathbf{p}}/m$ for a conduction-band to conduction-band transition is diagonal in spin projection and equals $\hbar\mathbf{k}\cdot\mathbf{E}/m_c$ where m_c is the conduction-band effective mass. The interband matrix elements of $\mathbf{E}\cdot\hat{\mathbf{p}}/m$ on the functions (9a) and (9b) are expressed in the standard way in terms of the Kane parameter P [actually, their proportionality to P has been used in (8)]. For the valence-band to valence-band transitions with a given $\mathbf{k}$ the operator $\mathbf{E}\cdot\hat{\mathbf{p}}/m$ takes on the form

$$(\mathbf{E}\cdot\hat{\mathbf{p}}/m)_v=-\hbar^{-1}\mathbf{E}\cdot d\mathcal{H}/d\mathbf{k}=-(\hbar\mathbf{k}/m)\left[(\gamma_1+\tfrac{5}{2}\gamma_2)(\mathbf{n}\cdot\mathbf{E})-\gamma_3[(\mathbf{n}\cdot\mathbf{J})(\mathbf{E}\cdot\mathbf{J})+(\mathbf{E}\cdot\mathbf{J})(\mathbf{n}\cdot\mathbf{J})]+2(\gamma_3-\gamma_2)\sum_\kappa n_\kappa E_\kappa J_\kappa^2\right] \tag{10}$$

[the minus sign in front of (10) reflects the fact that the electric charge of a hole is positive or, in other terms, the velocity of an electron in the valence band is $-\hbar^{-1}d\mathcal{H}/d\mathbf{k}$; this fact was not taken into account in Ref. 3)].

One can show using Eqs. (4) and (8)–(10) that

$$\begin{aligned}
&\mu_h^{(0)}(\mathbf{n},\mathbf{E})=\tfrac{3}{2}\gamma_h\gamma_l|\mathbf{E}\cdot\mathbf{n}|^2(|\mathbf{E}|^2-|\mathbf{n}\cdot\mathbf{E}|^2)+3\gamma_3^2(|\mathbf{E}|^4-|\mathbf{n}\cdot\mathbf{E}|^4-\tfrac{1}{2}|\mathbf{E}^2-(\mathbf{n}\cdot\mathbf{E})^2|^2)\ ,\\
&\gamma_h=\gamma_1-2\gamma_3+m/m_c\ ,\quad \gamma_l=\gamma_h+4\gamma_3\ ,\\
&\mu_l^{(0)}(\mathbf{n},\mathbf{E})=\tfrac{1}{2}\gamma_l^2|\mathbf{n}\cdot\mathbf{E}|^2(|\mathbf{E}|^2+3|\mathbf{n}\cdot\mathbf{E}|^2)+\tfrac{9}{2}\gamma_3^2|\mathbf{E}^2-(\mathbf{n}\cdot\mathbf{E})^2|^2+6\gamma_3\gamma_l\mathrm{Re}\{(\mathbf{n}\cdot\mathbf{E}^*)^2[\mathbf{E}^2-(\mathbf{n}\cdot\mathbf{E})^2]\}\ .
\end{aligned} \tag{11}$$

B. General formalism. The "anisotropic" part of the matrix elements

A rather general expression for the matrix elements of two-photon transitions that allows for the anisotropy of holes can be obtained by applying the projection-operator technique[10] and by making use of symmetry arguments. Because of the last term in the Hamiltonian of holes (6) the "true" wave functions of heavy and light holes are superpositions of the function (9a). The projection operators for heavy and light holes, $\Lambda_h(\mathbf{n})$ and $\Lambda_l(\mathbf{n})$, respectively, are given by the expressions

$$\Lambda_h(\mathbf{n})=[H(\mathbf{k})-\varepsilon_l(\mathbf{k})]/[\varepsilon_h(\mathbf{k})-\varepsilon_l(\mathbf{k})]\ ,\quad \Lambda_l(\mathbf{n})=1-\Lambda_h(\mathbf{n})\ . \tag{12a}$$

To zeroth order in $(\gamma_3-\gamma_2)/\gamma_3$, $\Lambda_{h,l}(\mathbf{n})=\Lambda_{h,l}^{(0)}(\mathbf{n})$ with

$$\Lambda_h^{(0)}(\mathbf{n})=\tfrac{1}{2}(\mathbf{n}\cdot\mathbf{J})^2-\tfrac{1}{8}\ ,\quad \Lambda_l^{(0)}(\mathbf{n})=\tfrac{9}{8}-\tfrac{1}{2}(\mathbf{n}\cdot\mathbf{J})^2\ . \tag{12b}$$

With the aid of the projection operators the combinations of the matrix elements $\mu_{h,l}(\mathbf{n},\mathbf{E})$ of Eq. (8) can be written as

$$\mu_\xi(\mathbf{n},\mathbf{E})=\tfrac{3}{2}(mPk)^{-2}\sum_{\alpha,\beta}\left|\sum_{\beta'}[(c,\alpha|\mathbf{E}\cdot\hat{\mathbf{p}}|v,\beta')(v,\beta'|\mathbf{E}\cdot\hat{\mathbf{p}}\Lambda_\xi(\mathbf{n})|v,\beta)-\delta_{\beta\beta'}(c,\alpha|\mathbf{E}\cdot\hat{\mathbf{p}}\Lambda_\xi(\mathbf{n})|v,\beta)(c,\alpha|\mathbf{E}\cdot\hat{\mathbf{p}}|c,\alpha)]\right|^2 \tag{13}$$

($\xi=h,l$). In obtaining (13) we have used the completeness of the set (9a) in the limit of strong spin-orbit splitting where virtual transitions to the split-off subband can be neglected. This completeness makes it possible also to rewrite the sum over β' in (13) as

$$-m\hbar^{-1}\sum_{\beta'}(c,\alpha|\mathbf{E}\cdot\hat{\mathbf{p}}|v,\beta')(v,\beta'|\mathbf{E}\cdot d\mathcal{H}/d\mathbf{k})\Lambda_\xi(\mathbf{n})|v,\beta)\ ,$$

with

$$\mathcal{H}=\frac{\hbar^2k^2}{2m_c}+H(\mathbf{k})\ . \tag{14}$$

It is quite straightforward then to sum over the conduction-band states, i.e., over α, in (13). The result is of the invariant form

$$\begin{aligned}
\mu_\xi(\mathbf{n},\mathbf{E})&=(m/\hbar^2k)^2\\
&\times\mathrm{Tr}[\Lambda_\xi(\mathbf{n})(\mathbf{E}^*\cdot d\mathcal{H}/d\mathbf{k})L(\mathbf{E})(\mathbf{E}\cdot d\mathcal{H}/d\mathbf{k})]\ ,
\end{aligned} \tag{15}$$

where the trace is taken over the valence-band states [it is simply the trace of the corresponding (4×4) matrix] and

$$L(\mathbf{E})=\tfrac{3}{2}(\hbar/mP)^2\sum_{\beta,\beta'}|v,\beta)\times\sum_{\alpha}(v,\beta|\mathbf{E}^*\cdot\hat{\mathbf{p}}|c,\alpha)(c,\alpha|\mathbf{E}\cdot\hat{\mathbf{p}}|v,\beta')\times(v,\beta'|\ . \tag{16}$$

Allowing for the explicit form of the functions (9a), (9b) one can easily show $L(\mathbf{E})$ to be of the form

$$L(\mathbf{E})=\tfrac{1}{2}\{\tfrac{9}{4}\mathbf{E}^*\cdot\mathbf{E}-(\mathbf{E}^*\cdot\mathbf{J})(\mathbf{E}\cdot\mathbf{J})+\tfrac{3}{2}i[\mathbf{E}^*\times\mathbf{E}]\cdot\mathbf{J}\}\ . \tag{17}$$

The single assumption made in obtaining (15) was, in essence, the independence of the operator L as given by (16) of the wave vector $\mathbf{k}$. This is a consequence of the interband matrix elements of the operator $\hat{\mathbf{p}}$ being independent of $\mathbf{k}$ for $2\hbar\omega-\varepsilon_g=(\hbar^2k^2/2m_c)+\varepsilon_\xi(\mathbf{k})\ll\varepsilon_g$. We note that Eq. (15) holds also for more general models of the valence band, e.g., for arbitrary spin-orbit splitting; the explicit form of $L(\mathbf{E})$ differs from (17) for such models, but $L(\mathbf{E})$ is still isotropic by symmetry arguments.

The Hamiltonian $\mathcal{H}$ in (14) and (15) can be put into a form of a linear combination of the zeroth-order projection operators $\Lambda^{(0)}_{h,l}(\mathbf{n})$ [Eq. (12b)]:

$$\mathcal{H}=\frac{\hbar^2}{m}\left[\tfrac{1}{2}\gamma_h k^2\Lambda^{(0)}_h(\mathbf{n})+\tfrac{1}{2}\gamma_l k^2\Lambda^{(0)}_l(\mathbf{n})+(\gamma_3-\gamma_2)\sum_{\kappa}k_\kappa^2[\Lambda^{(0)}_h(\boldsymbol{\kappa})-\Lambda^{(0)}_l(\boldsymbol{\kappa})]\right] \tag{18}$$

$\boldsymbol{\kappa}$ are unit vectors along the fourth-order crystal axes. It follows from (12), (15), and (18) that the evaluation of $\mu_{h,l}(\mathbf{n},\mathbf{E})$ is reduced to the calculation of

$$G(\mathbf{n};\mathbf{n}_1,\mathbf{n}_2;\mathbf{E})=\mathrm{Tr}[\Lambda^{(0)}_h(\mathbf{n})\Lambda^{(0)}_h(\mathbf{n}_1)L(\mathbf{E})\Lambda^{(0)}_h(\mathbf{n}_2)]\ . \tag{19}$$

By making use of (12b) and (17) one obtains from (19)

$$G(\mathbf{n};\mathbf{n}_1,\mathbf{n}_2;\mathbf{E})=g(\mathbf{n};\mathbf{n}_1,\mathbf{n}_2;\mathbf{E})+g(\mathbf{n};\mathbf{n}_2,\mathbf{n}_1;\mathbf{E}^*)\ ,$$

$$g(\mathbf{n};\mathbf{n}_1,\mathbf{n}_2;\mathbf{E})=\tfrac{3}{32}\{[1+(\mathbf{n}_1\cdot\mathbf{n}_2)^2+6(\mathbf{n}\cdot\mathbf{n}_1)^2]|\mathbf{E}|^2-3[1-(\mathbf{n}_1\cdot\mathbf{n}_2)^2]|\mathbf{E}\cdot\mathbf{n}|^2-2[1+3(\mathbf{n}\cdot\mathbf{n}_1)^2]|\mathbf{E}\cdot\mathbf{n}_2|^2 \tag{20}$$
$$-[3(\mathbf{n}\cdot\mathbf{n}_1)(\mathbf{n}\cdot\mathbf{n}_2)-(\mathbf{n}_1\cdot\mathbf{n}_2)][\mathbf{n}_1\times\mathbf{n}_2]\cdot[\mathbf{E}^*\times\mathbf{E}]-6[(\mathbf{n}\cdot\mathbf{n}_1)(\mathbf{n}_1\cdot\mathbf{n}_2)+(\mathbf{n}\cdot\mathbf{n}_2)][\mathbf{n}_2\times\mathbf{n}]\cdot[\mathbf{E}^*\times\mathbf{E}]\}\ .$$

Equations (15) and (18)–(20) make it possible to calculate the matrix elements $\mu_{h,l}(\mathbf{n},\mathbf{E})$ for arbitrary warping of the valence band. In the case of isotropic dispersion law ($\gamma_3=\gamma_2$) the result coincides with expression (11). The first-order correction in $\gamma_3-\gamma_2$ gives the anisotropic addition $\mu^{(1)}_{h,l}(\mathbf{n},\mathbf{E})$ in (8). It is rather clumsy, but the quantities of interest are the values of $\mu^{(1)}_{h,l}(\mathbf{n},\mathbf{E})$ averaged over n (integrated over the orientations of $\mathbf{n}$). It is substantial that, if we are not interested in small corrections $\sim(\gamma_3-\gamma_2)/\gamma_3$ to A_1, A_2, then only the terms proportional to $\sum_\kappa|E_\kappa|^4$ should be taken into account in the averaged $\mu^{(1)}_{h,l}$. The final result for the corresponding dichroic contributions, $\langle\mu^{(1)}_{h,l}(\mathbf{E})\rangle_d$, is quite compact:

$$\langle\mu^{(1)}_{h,l}(\mathbf{E})\rangle_d=(\gamma_3-\gamma_2)[-2\gamma_h-4\gamma_3\pm\tfrac{8}{5}\gamma_3\pm\tfrac{1}{7}(\gamma_h\gamma_l+3\gamma_3^2)/\gamma_3]\sum_\kappa|E_\kappa|^4\ , \tag{21}$$

where the + and − signs refer to heavy and light holes, respectively. The same result for $\langle\mu^{(1)}_{h,l}(\mathbf{E})\rangle$ was obtained also directly from Eq. (13) by expanding $\Lambda_{h,l}(\mathbf{n})$ to first order in $(\gamma_3-\gamma_2)/\gamma_3$ and by taking into account the anisotropic term in $(\mathbf{E}\cdot\hat{\mathbf{p}}/m)_v$ in Eq. (10).

C. Explicit expressions for the absorption coefficients

It follows from Eqs. (4), (8), (11), and (21) that the TPA parameters A_1, A_2, A_3 can be written as

$$A_i=8e^4P^2(2m)^{1/2}(2\hbar\omega-\varepsilon_g)^{3/2}a_i/3\pi\hbar^7\omega^5\ ,$$
$$a_i=a_i^{(h)}+a_i^{(l)}\ ,\quad i=1,2,3\ . \tag{22}$$

where $a_i^{(h)}$ and $a_i^{(l)}$ describe the contributions from the transitions resulting in the birth of heavy and light holes, respectively. To zeroth order in $(\gamma_3-\gamma_2)/\gamma_3$ the parameters $a_1^{(h,l)}$, $a_2^{(h,l)}$, that are responsible for the "isotropic" part of TPA, take the form

$$a_1^{(h)}=\tfrac{3}{10}(\gamma_h\gamma_l+8\gamma_3^2)\gamma_h^{-5/2}\ ,\quad a_2^{(h)}=-\tfrac{1}{3}a_1^{(h)}\ ,$$
$$a_1^{(l)}=(\tfrac{11}{30}\gamma_l^2-\tfrac{4}{5}\gamma_3\gamma_l+\tfrac{3}{5}\gamma_3^2)\gamma_l^{-5/2}\ , \tag{23}$$
$$a_2^{(l)}=(\tfrac{1}{10}\gamma_l^2+\tfrac{8}{5}\gamma_3\gamma_l+\tfrac{9}{5}\gamma_3^2)\gamma_l^{-5/2}\ .$$

The parameters $a_3^{(h,l)}$, that are responsible for linear dichroism of TPA, to lowest order in $(\gamma_3-\gamma_2)/\gamma_3$ are given by the expression

$$a_3^{(\xi)}=(\gamma_3-\gamma_2)(\pm\tfrac{1}{7}\gamma_\xi^3\gamma_3^{-1}-\tfrac{8}{7}\gamma_\xi^2\mp\tfrac{29}{35}\gamma_\xi\gamma_3+\tfrac{6}{7}\gamma_3^2)\gamma_\xi^{-7/2} \tag{24}$$

(the upper and lower signs refer to $\xi=h$ and $\xi=l$, respectively).

Equations (1)–(3) and (22)–(24) give the absorption cross section as a function of the polarization of the light in an explicit form and describe both the linear-circular dichroism of TPA that is inherent to the spherical-band approximation, and the specific linear dichroism which is a feature of cubic symmetry and is due to warping of the constant-energy surfaces of holes. It is evident that both

types of dichroism depend crucially on the band-structure parameters of a semiconductor.

The ratios (2) and (3) that characterize the dichroisms take on an extremely simple form when the reciprocal heavy-hole mass vanishes and the light-hole and conduction-band masses coincide:

$$\gamma_1=2\gamma_3\ ,\quad m/m_c=4\gamma_3\ ,\quad (m_h^{-1}=0\ ,\ m_l=m_c)\ . \tag{25a}$$

The linear dichroism is seen from (1) and (3) to be determined by the ratio $a_3/(a_1+a_2)$. According to the explicit expressions (23), (24), and (25a) at the threshold of TPA,

$$a_3/(a_1+a_2)\simeq -0.4(\gamma_3-\gamma_2)/\gamma_3\ . \tag{25b}$$

It is seen from (25b) that for typical values $(\gamma_3-\gamma_2)/\gamma_3\sim 0.1$ the absorption cross section should differ by several percent for different orientations of the light polarization plane with respect to crystal symmetry axes.

Remarkably, the linear-circular dichroism is practically absent in the case (25a): $K_{\rm lin}/K_{\rm circ}=0.98$ with neglect of terms $\sim(\gamma_3-\gamma_2)/\gamma_3$. This results from the heavy- and light-hole contributions to $K_{\rm lin}/K_{\rm circ}$ cancelling each other. We note that it is not due to the symmetry and is quite unusual, generally speaking. For example, for an isotropic medium containing homogeneously distributed over orientations two-photon-absorbing dipoles with a matrix element of the transition proportional to $(\mathbf{E}\cdot\mathbf{d})^2$, where $\mathbf{d}$ is the dipole moment, one can easily show $K_{\rm lin}/K_{\rm circ}$ equal to $\frac{3}{2}$. It would be expected therefore that the linear-circular dichroism will be highly sensitive to the effects of virtual transitions to other bands (first of all, to the split-off valence subband) and also to the deviations from the approximation (25a).

III. NUMERICAL RESULTS

The analysis of the frequency dependence of the TPA as a whole and of the TPA dichroism in particular has been done for several zinc-blende-structure semiconductors numerically. A realistic model of the band structure of such semiconductors is the (8×8) Kane model. In this model not only the heavy- and light-hole subbands, but also the split-off one are taken into account. We have neglected in the (8×8) matrix Hamiltonian $\tilde{H}$ the terms arising because of the lack of the inversion symmetry and ignored also the difference in the matrix elements of $\tilde{H}$ due to the difference in the bases of orbital wave functions for Γ_8 and Γ_7 valence subbands.

The numerical evaluation of the matrix elements of two-photon transitions in (4) was based on diagonalization of $\tilde{H}$. For a given direction of $\mathbf{k}=k\mathbf{n}$ the actual absolute values k were found from the energy-conservation law (we note that several transitions can happen in a multisubband system for a given $\mathbf{n}$). The matrix elements of $\hat{\mathbf{p}}=(m/\hbar)d\tilde{H}/d\mathbf{k}$ on the eigenstates of $\tilde{H}$ were then calculated, weighted allowing for the light polarization and averaged over $\mathbf{n}$.

The TPA parameters a_1, a_2, a_3 versus relative energy excess

$$\delta=(2\hbar\omega-\varepsilon_g)/\varepsilon_g \tag{26}$$

for two actual semiconductors, InSb and GaAs, are plotted in Figs. 1 and 2. The data for CdHgTe alloys are qualitatively the same as those for InSb. The parameters in the figure captions are given in the notations of Ref. 11 (in view of the above approximation the parameters with and without primes in Ref. 11 were assumed coincident). To avoid ambiguity we use for the intra-valence-band parameters of $\tilde{H}$ the notations γ_{1K}, γ_{2K}, γ_{3K} instead of γ_1, γ_2, γ_3 in Ref. 11. The parameters γ_1, γ_2, γ_3 in Eq. (6) and the conduction-band effective mass m_c are related to γ_{1K}, γ_{2K}, γ_{3K} and the "bare" conduction-band mass $m/(2F+1)$ by the expressions

$$\begin{aligned}&\gamma_1=\gamma_{1K}+\tfrac{1}{3}(\varepsilon_p/\varepsilon_g)\ ,\\ &\gamma_i=\gamma_{iK}+\tfrac{1}{6}(\varepsilon_p/\varepsilon_g)\quad (i=2,3)\ ,\\ &\frac{m}{m_c}=2F+1+\tfrac{1}{3}\varepsilon_P\left[\frac{2}{\varepsilon_g}+\frac{1}{\varepsilon_g+\Delta}\right]\quad \varepsilon_p=2mP^2/\hbar^2\ .\end{aligned} \tag{27}$$

It is seen from Fig. 1 that, for InSb, the frequency dependences of the "isotropic" dimensionless parameters of TPA, a_1 and a_2, are monotonous, while a_3 is nearly independent of frequency. The values of a_1 and a_3 near the absorption threshold ($\delta\ll 1$) are close to those that result from the explicit expressions (23) and (24). The frequency dependence of a_1 that characterizes the polarization-independent TPA is in good agreement with analytical results for the isotropic (6×6) Kane model with the infinite heavy-hole mass [cf. (25a)] and with the same values of ε_p, ε_g as those in Fig. 1. The analytical expressions could be obtained, e.g., along the lines of Ref.

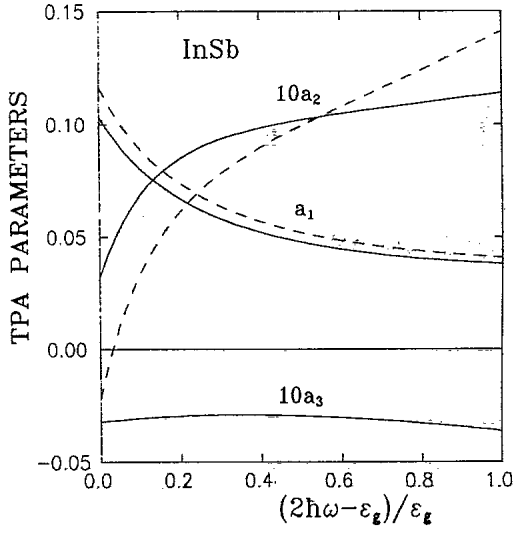

FIG. 1. Frequency dependence of the dimensionless parameters of two-photon absorption for the (8×8) Kane model (Ref. 11) with the parameter values $\varepsilon_g=0.2352$ eV, $\varepsilon_p=23.2$ eV, $\Delta=0.803$ eV, $\gamma_{1K}=3.25$, $\gamma_{2K}=-0.2$, $\gamma_{3K}=0.9$, $F=-0.2$ that correspond (Ref. 12) to InSb. The analytical results for the isotropic (6×6) Kane model with infinite heavy-hole mass and with the same $\varepsilon_p/\varepsilon_g$ are shown as dashed lines.

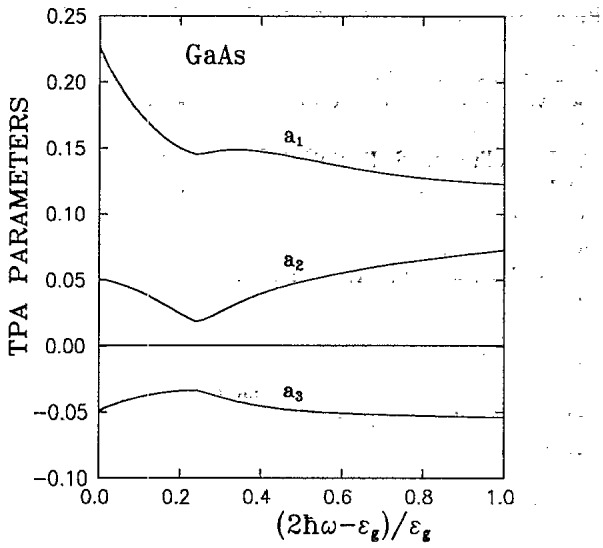

FIG. 2. Frequency dependence of the dimensionless parameters of two-photon absorption for the (8×8) Kane model (Ref. 11) with the parameter values $\varepsilon_g=1.5$ eV, $\varepsilon_p=23$ eV, $\Delta=0.35$ eV, $\gamma_{1K}=2.54$, $\gamma_{2K}=-0.15$, $\gamma_{3K}=0.72$, $F=0$ that correspond approximately to GaAs.

1, and our analytical result for $K_{\rm lin}/K_{\rm circ}$ coincided with that in Ref. 1.

The difference in the numerical and analytical values of the parameter a_2 is much more pronounced (for small δ they are of opposite signs). This is due to the fact that the contributions to a_2 due to transitions from the light- and heavy-hole subbands nearly compensate each other for $\delta \ll 1$; therefore, virtual transitions to the split-off subband contribute to a_2 noticeably although the splitting Δ is large. As a consequence, the ratio $K_{\rm lin}/K_{\rm circ}$ exceeds 1 for all frequencies, and it approaches 1.3 for $\delta \to 1$.

It follows from the data shown in Fig. 1 that the linear dichroism parameter $a_3/(a_1+a_2)$ increases monotonously in absolute value with the light frequency (primarily because of the decrease of a_1). It is -0.03 for $\delta \to 0$ and -0.07 for $\delta \to 1$. This means that the TPA cross section, depending on the frequency, should vary by 2–5 % with the rotating light polarization plane for light propagating along a $\langle 110 \rangle$ axis.

It would be expected that the linear-circular dichroism will increase if the spin-orbit splitting Δ decreases, and will be strongly changed if real transitions from the split-off subband come into play. A plausible small-Δ semiconductor is GaAs. This semiconductor is interesting also because of warping of the hole constant-energy surfaces being comparatively large: $(\gamma_3-\gamma_2)/\gamma_3 \simeq 0.25$. The values of the parameters a_2, a_3 that characterize the "polarization-dependent" TPA are seen from Fig. 2 to exceed, in the case of GaAs, those in InSb by nearly an order of magnitude, while the values of a_1 differ by a factor of 2. The linear-circular dichroism as defined by (2) varies in GaAs within the interval (1.16, 1.59), while the linear dichroism parameter $a_3/(a_1+a_2)$ changes monotonously from -0.18 to -0.27 as δ increases from 0 to 1. The latter means that the ratio K_{100}/K_{111} of the TPA cross sections for light polarized along the $\langle 100 \rangle$ and $\langle 111 \rangle$ axes amounts to 0.80–0.87, depending on the light frequency.

The feature obvious from Fig. 2 is the nonmonotony of the frequency dependence of the parameters a_1, a_2, a_3. The bending occurs for $2\hbar\omega=\varepsilon_g+\Delta$, i.e., at the threshold of two-photon transitions from the split-off subband; the corresponding contribution is proportional to $(2\hbar\omega-\varepsilon_g-\Delta)^{3/2}$ near the threshold.

We note that the Kane model does not describe warping of the conduction band that is substantial in GaAs for electron energies of order of the $\Gamma-L$ spacing. Although the photon energies for direct two-photon transitions to the L valleys exceed ε_g this warping gives rise to additional anisotropy of TPA for $\delta \sim 1$.

IV. CONCLUSION

It follows from the results of the present paper that both linear-circular and linear dichroisms of the two-photon absorption are of utmost sensitivity to the interrelation between the band-structure parameters of direct-band-gap zinc-blende-structure semiconductors. The difference in the TPA cross sections for different light polarizations is quite large. This makes it possible to determine the ratios of the band-structure parameters, in particular warping of the constant-energy surfaces, in one-laser-beam experiments without external fields.

We note in conclusion that the dichroism considered in the present paper is inherent not only to bulk cubic semiconductors, but also to the cubic-semiconductor-based quantum-well structures for light propagating along the confinement direction (in addition to the difference in the absorption cross sections for the beams polarized parallel and perpendicular to this direction[13]). It can be even stronger because of the splitting of the valence band. The corresponding results will be discussed elsewhere.

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