PAPER

Strong-field induced optical anisotropy in semiconductors with cubic structure: theory

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Abstract

A mechanism pertaining to an anomalous optical anisotropy under a strong field in semiconductors having cubic structure is proposed. The valence-conduction band transitions of valence electrons located around atomic bonds re-distribute electrons between bands and, consequently, produce symmetry defects in local electronic density. The optical anisotropy is due to these symmetry defects, which are the sources for localized and non-compensated dipole moments randomly distributed within the crystal. The quantum time-dependent probability amplitude method, combined with classical electrodynamics, demonstrate that the non-compensated dipole moments destroy the optical isotropy of a crystal. To compare the scheme with an experiment, the theory is applied to GaAs single crystal. Within a strong-field IR regime, the predicted four-fold symmetry of optical anisotropy, along with its amplitude, correlate well with an experiment.

1. Introduction

The interaction of an electromagnetic wave with matter remains one of the most intriguing problem in solid state physics. This interaction depends on the magnitude of the field, and leads to many effects, from the classical Laue scattering [1, 2] and the second harmonic generation [3, 4], to the damage of the sample [5–7]. In a weak field, the electromagnetic wave-crystal interaction mechanisms are described in numerous studies (see e.g. [8–13]), but the strong field studies [14–16] are still rare. Currently, due to both fast-developing techniques of ultrafast terahertz pulse spectroscopy [17–21] and the non-linear optics applications [22, 23], the problem of interaction of strong-field wave with crystals, becomes especially important.

It is known that strong-field laser irradiation with an electric field amplitude, \( E_0 > MV/m \), excites electronic states in metals, semiconductors and dielectrics far beyond plasmonic oscillations [18, 24–26]. In semiconductors, due to multiphoton absorption, if the photon energy is smaller than the bandgap, \( h\omega < \Delta E_g \), the non-resonant excitation of an electronic system induces the transition of electrons from the valence band to the conduction band (\( VB \rightarrow CB \)) [24]. Due to this transition, the correction, \( \Delta n_A \), to the equilibrium electron density in \( CB \) can affect both the electric properties of a material, e.g. the conductivity, \( \sigma \), and, consequently, the optical behaviors [18, 26].

Moreover, recent studies show that the strong-field laser irradiation can dramatically modify the fundamental symmetry-related optical properties of a semiconductor. For example, the experiment demonstrated that the bulk optical response of a single-crystal GaAs to strong laser field is different from the expected one [16]. In an experimental setup, the linearly-polarized strong field, \( E \sim GV/m \), 1900 nm IR laser beam was passed through the 600 \( \mu \)m thick GaAs crystal. The laser beam induces the \( VB \rightarrow CB \) transitions, and the laser transmission, \( T \), was measured. The incident laser beam was normal to the single-crystal plane (100), and \( T \) data were collected as a function of crystal rotation of polarization direction by the angle \( \theta \) with [001] axis (figure 1). The rotation of the crystal is equivalent to the rotation of the electric field vector, \( \vec{E} \), in the (100) plane (figure 1), and the laser transmission was obtained for \( 0 < \theta < 360^\circ \). The transmission, \( T \), is shown in figure 2, and \( T \) is a periodic function of the angle \( \theta \), \( T(\theta) = T_0 + A \sin^2(2\theta) \), with four-fold periodicity and the fitting parameter, \( A > 0 \). As follows from Fresnel theory, this behavior of \( T \) is very different from expected in crystals with a cubic
symmetry transmission, \( T = \text{const}. \) Consequently, this dramatic discrepancy should be explained. Indeed, any kind of dependence of \( T \) on \( \theta \), which is different from \( T = \text{const} \), is an obvious violation of a well-known principle of the optical isotropy of cubic crystals [1].

In other words, in a strong field, the single-crystal GaAs demonstrates a birefringence behavior which is forbidden in cubic structures according to Ref. [1]. It is important to remind that the optical isotropy in cubic lattices is based on unshakable arguments of lattice symmetry [1]. Therefore, to explain the birefringence in cubic GaAs, one should assume that the cubic symmetry of either the lattice or the electron distribution was broken.

This idea has motivated our current paper and our method has been specifically applied to the GaAs induced anisotropy. We will compare the theoretical predictions with the experimental data presented in [16].

The structure of the paper is as follows.

In section 2, the proposed scheme is considered in more detail to explain the physics underlying the formal calculations. In section 3, the quantum time-dependent probability amplitude method is utilized to obtain the time-breathing mode for the populations of the electrons distributed between the VB and the CB. The goal of this section is to obtain number \( (\text{or the density}) \) of affected atomic bonds and the amplitudes of the excited dipole moments. In section 4, we analyze the symmetry-driven orientations for the randomly distributed in a crystal containing excited dipole moments. The power radiated back by the oscillating dipole moments is obtained and the transmission, \( T \), is calculated. The explicit expressions for the optical parameters, along with the optical anisotropy, are compared with the experiment [16]. In section 5, we discuss the results, and compare the theory with the alternative explanations of the effect.
2. Optical anisotropy and band transitions

In an ideal cubic lattice, the induced dipole moment, $\vec{d}_i$ is parallel to the electric field vector, $\vec{E}$ ($\vec{d}_i || \vec{E}$). To the contrary, in a birefringent crystal, the electric vector and the induced dipole moment are not collinear anymore: $\vec{d}_i \neq || \vec{E}$ (see figure 3). Hence, in a scenario with optical anisotropy present, there must be induced dipole moments, $\vec{d}_i$, which are noncollinear with the electric field.

We associate the origin of the dipole moments $\vec{d}_i$ with the well-known dipole nature of the VB $\rightarrow$ CB transitions [13]. In GaAs, the sp$^3$ valence electron density is located predominantly at the GaAs atomic bonds. Consequently, strong-field-induced VB $\rightarrow$ CB transitions should leave behind the local, and non-compensated dipole moments, $\vec{d}_i$, randomly distributed in space. These dipole moments are due to a re-distribution of the electron density, and the crystal, in terms of an electron density distribution, locally loses its cubic symmetry. We will call the defected electronic configuration in vicinity of the affected GaAs atomic bond a symmetry defect. Hence, in the GaAs crystal with defected cubic symmetry for the electron density, in accordance with the optical principles [1], one should expect the birefringence signature in transmission-reflection curves.

The optical effect of these randomly distributed induced local dipole moments is based on the following picture. In GaAs, each As atom (4s$^2$4p$^3$) is bound with four Ga atoms (4s$^2$4p$^1$) by means of a shared valence sp$^3$ electron. The GaAs atomic bonds form the tetrahedron, as shown in figure (4), with 4 different space orientations. In a single-mode radiation field, $\vec{E} = E_0 \cos(\omega t)$, the valence electrons in each of 4 atomic bond orientations experience the VB $\rightarrow$ CB transitions, with the same probability.

The magnitude of the induced dipole moment, $d_0 = |\vec{d}_i|$, located at the symmetry point defect, is given by $d_0 = e\Delta r$, where $\Delta r$ is the characteristic charge separation distance, and $e$ is an elementary charge. The reasonable estimate for the parameter $\Delta r$ is the displacement, $\Delta r$, of the electronic density for the 4p$^1$ electron of the Ga atom due to the hybridization with 4p$^3$ orbitals of the As atom. Since this parameter approximately coincides with the displacement of the Ga atom, the charge separation distance can be estimated as $\Delta r_{el} \approx 2.2 \AA$ is the length for the atomic bond [27], $R_{Ga} \approx 1.30 \AA$, and $R_{As} \approx 1.29 \AA$ are the atomic radii for the As and Ga atoms, respectively [28].

Note, that the Rabi frequency, $\Omega_R = d_0 E_0 / h \approx 1.2 \times 10^{14}$ rad s$^{-1}$ is much smaller than the following parameters: the transition frequency, $\nu = \Delta U / h = 2.2 \times 10^{15}$ rad s$^{-1}$($\Delta U = 1.43$ eV), the laser frequency, $\omega = 2\pi c / \lambda \approx 10^{15}$ rad s$^{-1}$, and their difference, $\nu - \omega \approx 1.2 \times 10^{15}$ rad s$^{-1}$.
Since the induced dipole moment $\vec{d}$ depends solely on both the initial unperturbed electronic state, and the state following the valence-conduction band transition, $\vec{d}$ has no field orientation $\theta$ dependence. Consequently, the orientation of the dipole moment $\vec{d}$ depends solely on the orientation, $i$, of an affected GaAs atomic bond. Since all four different orientations of the dipole moments, $\vec{d}_{i}$, cannot be simultaneously collinear with the single orientation of the electric field vector $\vec{E}$, the dipole moments, $\vec{d}_{i}$, are the sources for the birefringence. Note, that there is also a component of the induced dipole moment, linear with respect to the field, $\vec{d} = \alpha_{\theta} \vec{E} |\vec{E}|$, which is parallel to the electric field ($\alpha_{\theta}$ is a polarizability scalar). This component provides the isotropic optical response and is due to both the plasmonic mode of conduction electrons and the contribution from energetically higher transitions related to bound electrons.

Both $\vec{d}$ and $\vec{d}_{i}$ coexist in the crystal, which becomes a mixture of two components: optically isotropic component, due to $\vec{d}$, and optically anisotropic component, due to $\vec{d}_{i}$. Consequently, the averaged total dipole moment, per unit of crystal volume, $\vec{d}_{\text{tot}}$, is the sum of two terms:

$$\vec{d}_{\text{tot}} = \alpha \vec{E} \left( 1 - \sum_{i} \frac{\Delta N_{0}^{i}(t)}{N_{0}^{i}} \right) + \sum_{i} \frac{\Delta N_{0}^{i}(t)}{N_{0}^{i}} \vec{d}_{i}.$$  \hspace{1cm} (1)

In equation (1), the first term, is the optically isotropic contribution, $\vec{d}$, and the second term is the optically anisotropic part due to the localized dipole moments, $\vec{d}_{i}$. Here, $N_{0}$ and $\Delta N_{0}^{i}(t)$ are the total density of the GaAs bonds and the time-dependent densities of the affected bonds with the orientation $i$, respectively. Since the second term in parenthesis of equation (1) is much smaller than 1 (see below), it can be omitted. Also, in the calculation of the macroscopic reflection, $R$, one should consider the contributions from the first and the second terms of equation (1) separately, since the localized dipole moments, $\vec{d}_{i}$, are randomly distributed inside the crystal. Moreover, each orientation, $i$, separated in space from other orientations, provides its own contribution into the reflection.

3. Quantum dynamical solution

A consistent description of the interband electron transitions in large-gap semiconductors ($\hbar \omega < \Delta U_{g}$) should take into account, alongside of VB $\rightarrow$ CB transitions, the transitions of electrons from the conduction band back to the valence band, or CB $\rightarrow$ VB transitions. The interband transitions problem is formally analogous to the problem of the coupling of a two-level atomic system with a single mode of the electromagnetic field. The standard method to treat this problem in Quantum Optics is the so-called atom-field semiclassical theory, or the probability amplitude method [29–32]. This scheme considers the time-dependent Schrödinger equation for a two-level system to obtain the time-dependent populations of the atomic states.

The electron transitions are due to the non-diagonal matrix elements for the electric dipole, $\vec{d}_{i}$, between two states. Based on this analogy, since we deal with two energy levels and two VB $\rightarrow$ CB transitions, the probability amplitude method is utilized here.

The scheme [29], or the probability amplitude method [29] treats the kinetics of electrons involved in transitions, based on the solutions for the probability amplitudes for a valence electron to occupy either VB or CB. The approach allows to calculate all relevant, and principally time-dependent, measurable for the system. The measurable of our interest are the time-dependent populations of the symmetry defects, $\Delta N_{0}^{i}(t)$, averaged over the space coordinates, $\vec{r}$. Note, that the densities, $\Delta N_{0}^{i}$, of the symmetry defects are small when compared with the total density, $N_{0}$, of the atomic bonds, $\Delta N_{0}^{i} \ll N_{0}$ (see an estimate below). Consequently, the interband transitions due to GaAs atomic bonds with different orientations are considered here as independent processes.

The two-level quantum system, figure 5, consists of a ground state of GaAs atomic bond, $|b\rangle$, with a valence electron in VB, and an excited state, $|a\rangle$, (valence electron excited into CB). We will consider the Cartesian coordinate system defined by the axis lines, $X$ [001], $Y$ [100] in the (100) plane, and $Z$ [100]. Both states correspond to the specific orientation of GaAs atomic bond with an index $i = \{1, 2, 3, 4\}$. The non-diagonal dipole moments between the states $|a\rangle$ and $|b\rangle$ are expressed in $[X, Y, Z]$ Cartesian system in terms of their components, $\vec{d}_{i} = \{d_{i}^{x}, d_{i}^{y}, d_{i}^{z}\}$. Note, that the relaxation of the excited dipole moment, $\vec{d}_{i}$, due to an external reservoir, is not considered in the probability amplitude method [28]. Consequently, the components of the dipole moments are the real numbers, $\text{Im} d_{i}^{x,y,z} = 0$. The electric field, $\vec{E} = \vec{E}_{0} \cos(\omega t)$, has the amplitude components, $\vec{E}_{0} = \{ \cos \theta, \sin \theta, 0 \}$, (figure 1). The time-dependent wave function, $|\Psi(t)\rangle$, of the two-level system is written in the form:

$$|\Psi(t)\rangle = C_{a}(t)|a\rangle + C_{b}(t)|b\rangle,$$  \hspace{1cm} (2)

where $C_{a}(t)$ and $C_{b}(t)$ are the probability amplitudes of finding the system in states $|a\rangle$ and $|b\rangle$, respectively. The corresponding Schrödinger equation is given by:
\[ |\Psi(t)\rangle = -\frac{i}{\hbar}\hat{H}|\Psi(t)\rangle, \quad (3) \]

with Hamiltonian,
\[ \hat{H} = \hat{H}_0 + \hat{H}_I. \quad (4) \]

Here \( \hat{H}_0 \) and \( \hat{H}_I \) represent the unperturbed and interaction parts of the Hamiltonian, respectively \([28]\):
\[ \hat{H}_0 = \hbar \omega_a |a\rangle \langle a| + \hbar \omega_b |b\rangle \langle b|, \quad (5) \]
\[ \hat{H}_I[a] = \hbar \omega_a |a\rangle, \quad (6) \]
\[ \hat{H}_I[b] = \hbar \omega_b |b\rangle, \quad (7) \]
\[ \hat{H}_I = -\{ (d_x^a \cos \theta + d_y^a \sin \theta) |a\rangle \langle b| + |b\rangle \langle a| \} E_0 \cos(\omega t), \quad (8) \]
\[ d_{x,y}^a = e(a|x,y,|b). \quad (9) \]

One can see from equation (8), that, since the electric field vector has no component in \( z \) direction, the problem is two-dimensional.

The equations of motion for the amplitudes \( C_a(t) \) and \( C_b(t) \) are given by:
\[ \dot{C}_a = -i\omega_a C_a + i\Omega_{R}^a \cos \omega t C_b, \quad (10) \]
\[ \dot{C}_b = -i\omega_b C_b + i\Omega_{R}^b \cos \omega t C_a, \quad (11) \]

where the Rabi frequency \( \Omega_{R}^{a,b} \) is defined as
\[ \Omega_{R}^{a,b} = \frac{E_0}{\hbar} (d_x^a \cos \theta + d_y^a \sin \theta). \quad (12) \]

Next, to solve equations (10)–(11), we will utilize the rotating-wave approximation, which ignores fast counter-rotating terms proportional to \( \exp[\pm i(\nu + \omega)t] \) and keeps slowly varying terms \( \sim \exp[\pm i(\nu - \omega)t] \). This method is utilized in many atom-field interaction schemes and is generally a very good approximation [29]. Since, in experiment [16], the parameter \( (\nu + \omega)/(\nu - \omega) \approx 2.7 \gg 1 \) [16], the rotating-wave is a reasonable approximation.

The equations for the slowly varying amplitudes,
\[ c_a = C_a e^{i(\omega + \nu)t}, \quad (13) \]
\[ c_b = C_b e^{i(\omega - \nu)t}, \quad (14) \]

as follows from equations (10)–(11), are
\[ \dot{c}_b = \frac{i\Omega_{R}^b}{2} c_a e^{-i(\nu - \omega)t}, \quad (15) \]
\[ \dot{c}_a = \frac{i\Omega_{R}^a}{2} c_b e^{i(\nu - \omega)t}. \quad (16) \]

Here, the transition frequency, \( \nu = \omega_a - \omega_b \).

The exact solution of equations (15)–(16), with initial conditions,
\[ c_a(0) = 0, \ c_b(0) = 1, \] is given by:
\[
c_a(t) = i \frac{\Omega \hbar}{\Omega^2} \sin \left( \frac{\Omega t}{2} \right) e^{i \Delta t/2},
\]
\[
c_b(t) = \left\{ \cos \left( \frac{\Omega t}{2} \right) + i \frac{\Delta}{\Omega} \sin \left( \frac{\Omega t}{2} \right) \right\} e^{-i \Delta t/2},
\]
where \( \Omega = \sqrt{\left(\Omega_{\mathrm{R}}\right)^2 + \Delta^2}, \Delta = \nu - \omega. \)

The probabilities, \( |c_a(t)|^2 \) and \( |c_b(t)|^2 \) of a system being in states \( |a\rangle \) and \( |b\rangle \) at time \( t \) are:
\[
|c_a(t)|^2 = \frac{\Omega_{\mathrm{R}}^2}{\Omega^2} \sin^2 \left( \frac{\Omega t}{2} \right),
\]
\[
|c_b(t)|^2 = 1 - \frac{\Omega_{\mathrm{R}}^2}{\Omega^2} \sin^2 \left( \frac{\Omega t}{2} \right).
\]

As follows from equations (19)–(20) \( |c_b(t)|^2 \) oscillates in time with the frequency \( \Omega/2 \). Since the number of the symmetry defects, \( \Delta N(t) = N_0 |c_b(t)|^2 \), the total number of all symmetry defects, \( \Delta N \), averaged over the large measurement time \( \tau \gg (\Omega)^{-1} \), is the sum:
\[
\Delta N = N_0 \sum_{i=1}^{4} \left( \frac{E_0}{\hbar} \right)^2 (d_{i}^i \cos \theta + d_{i}^j \sin \theta)^2 \frac{1}{(\nu - \omega)^2}.
\]
Hence, one may estimate the fraction (density) of the symmetry point defects \( \beta = \Delta N / N_0 \), as:
\[
\beta = \frac{4}{\hbar} \left( \frac{E_0}{\hbar} \right)^2 \sum_{i=1}^{4} (d_{i}^i \cos \theta + d_{i}^j \sin \theta)^2 \frac{1}{(\nu - \omega)^2}.
\]

Since the Rabi frequency is smaller than \( \Delta, |\Omega_{\mathrm{R}}|/|(\nu - \omega)| \approx 0.1 \) (see above), we put \( \Omega = (\nu - \omega) \) in equations (21)–(22). The total induced dipole moment, \( \vec{d}_{\mathrm{tot}} \), is given by:
\[
\vec{d}_{\mathrm{tot}} = \alpha E_0 (\cos \theta \vec{i} + \sin \theta \vec{j}) + \frac{1}{2} \left( \frac{E_0}{\hbar} \right)^2 \sum_{i=1}^{4} (d_{i}^i \cos \theta + d_{i}^j \sin \theta)^2 \vec{d}_{i}^i.
\]
where \( d_{i}^i = d_{i}^i \vec{i} + d_{j}^j \vec{j} + d_{k}^k \vec{k}. \)

Here the first term is the linear polarization-induced dipole moment in an ideal cubic lattice which provides the optically isotropic response. The second, non-linear with respect to the electric field, term, is the \( \text{VB} \rightarrow \text{CB} \) transition-driven dipole moment responsible for the optically anisotropic.

Note, that in equation (23) the index \( i \) represents the dipole moment, \( \vec{d}^i \), as a sum of three mutually perpendicular dipole moments, \( d_{i}^i \vec{i}, \ d_{j}^j \vec{j}, \ d_{k}^k \vec{k} \) which includes the moment, \( d_{i}^i \vec{i}. \)

Since the \( d_{k}^k \vec{k} \) component of the dipole moment is perpendicular to the electric field vector, \( \vec{E} \), this component does not oscillate with the field frequency. Hence, \( d_{i}^i \vec{k} \) provides no contribution to the reflection, \( R \). Consequently, in calculation of an optical response the dipole moment is reduced to the sum of two terms:
\[
\vec{d}^i = d_{i}^i \vec{i} + d_{j}^j \vec{j}.
\]

**4. Optical anisotropy**

The scheme developed above allows to calculate the induced \( \theta \)-dependent anisotropic transmission, \( T(\theta) \). The transmission, if one neglects the absorption and scattering, is given by:
\[
T = 1 - R,
\]
where \( R \) is the reflection.

Here the reflection depends on the induced dipole moment, \( \vec{d}^i \), and \( R \) is proportional to the square of \( |\vec{d}| \):
\[
R = k |\vec{d}|^2.
\]

The equation (26) is derived from the expression for the time-averaged power \( P_{\text{av}} \) radiated by the oscillating dipole moment \( \vec{d} \) per unit solid angle [33]:
\[
\frac{dP_{te}}{d\Omega} = \frac{e}{8\pi} \left(\frac{2\pi}{\lambda}\right)^4 |\vec{d}|^2 \sin^2 \varphi,
\]

(27)

where the angle \( \varphi \) is measured from the direction of the \( \vec{d} \). The equation (27) yields the total reflected power, \( P_{th} \), per a unit surface, due to dipole moments within in a slab of a thickness \( L \):

\[
P_{th} = \frac{e}{8} \left(\frac{2\pi}{\lambda}\right)^4 \Delta N L_{\Delta} |\vec{d}|^2 \int_0^\pi \sin^2 \varphi d\varphi
\]

\[
= \frac{e}{6} \left(\frac{2\pi}{\lambda}\right)^4 \Delta N L_{\Delta} |\vec{d}|^2,
\]

(28)

Since the reflection by definition, is \( R = \frac{P_{th}}{I} \), then the equation (26) follows, with the coefficient, \( \kappa \), given by:

\[
\kappa = \frac{e}{6I} \left(\frac{2\pi}{\lambda}\right)^4.
\]

(29)

As follows from equation (23), the induced dipole moment \( \vec{d}_{ind} \) has two components: the isotropic part (the first term) and the sum of the non-isotropic induced components with different orientations, \( i \), (the second term).

Since the induced components of the dipole moments are located randomly on different lattice sites, the contributions to \( R \) from the anisotropic part should be calculated separately for each orientation of the dipole moments:

\[
R = R_{is} + R_{anis},
\]

(30)

\[
R_{anis} = \kappa \alpha^2 E_0^2,
\]

(31)

\[
R_{anis} = \xi \sum_{i=1}^{4} (d_i^x \cos \theta + d_i^y \sin \theta)^4 (d_i^{x^2} + d_i^{y^2}),
\]

(32)

where the coefficient \( \xi = \frac{\kappa}{4} \left(\frac{E_0}{(\hbar \omega - \epsilon_0)}\right)^4 \).

In equation (32), the sum describes the contributions from each of four differently oriented dipole moments.

Consequently, the transmission is given by:

\[
T(\theta) = (1 - \kappa \alpha^2 E_0^2) - \xi \sum_{i=1}^{4} (d_i^x \cos \theta + d_i^y \sin \theta)^4 (d_i^{x^2} + d_i^{y^2})
\]

(33)

Here, the first term of equation (33) is the optically- isotropic part of the transmission, \( T = \text{const} \). The second term is the optically-anisotropic part of \( T \) which depends on the orientation of the polarization vector, \( \theta \).

There are two kinds of symmetries in a regular atomic system: (1) the symmetry of the electrons density distributions and, (2), the symmetry of the crystal lattice. The induced dipole moments are due to the local violations for the symmetry of the electron density distribution. Since the density of the induced dipole moments (see below), is small, we will assume that they do not affect dramatically the symmetry of the crystal lattice. Hence, we will utilize the requirement that the orientations of four induced dipole moments, \( \vec{d}_i = \{d_i^x, d_i^y, d_i^z\} \), supports the crystal symmetry. Since the \( d_i^z \) components are irrelevant and do not affect the reflectivity (see above), they are dropped from the calculations. Consequently, four two-dimensional dipole moments, \( \vec{d}_i = \{d_i^x, d_i^y\} \), are considered in the (100) plane. In the plane (100) with the axis lines, \( X \ [001], \ Y \ [010], \) (figure 1), the symmetry requirements must be satisfied. These requirements are the four-fold symmetry of the \( d_i^x, d_i^y \) components and the symmetry with respect to both directions of the axis lines \( X \ [001], \ Y \ [010] \). There are only two solutions, (a,b), for the dipole moments orientations in the plane (100) which satisfy these symmetry constraints:

(a): \[
\{d_1^x, d_1^y\} = \{d_0, 0\},
\{d_2^x, d_2^y\} = \{-d_0, 0\},
\{d_3^x, d_3^y\} = \{0, d_0\},
\{d_4^x, d_4^y\} = \{0, -d_0\},
\]

(34)

(b): \[
\{d_1^x, d_1^y\} = \{d_0/\sqrt{2}, d_0/\sqrt{2}\},
\{d_2^x, d_2^y\} = \{d_0/\sqrt{2}, -d_0/\sqrt{2}\},
\{d_3^x, d_3^y\} = \{-d_0/\sqrt{2}, d_0/\sqrt{2}\},
\{d_4^x, d_4^y\} = \{-d_0/\sqrt{2}, -d_0/\sqrt{2}\}.
\]

(35)
The solution (a) when substituted in equation (33) taking into account the identity
\[
\cos^2 \theta + \sin^2 \theta = (2 - \sin^2(2\theta))/2,
\]
yields an angular dependence \( T(\theta) \) for the transmission:
\[
T(\theta) = 1 - R_0[1 + \gamma(2 - \sin^2(2\theta))]
\]
where \( \gamma \) is the coefficient, and the second term in brackets is the anisotropic correction. The predicted transmission, \( T(\theta) \), from equation (36), has a four-fold periodicity which matches the behavior of the experimental curve [16], \( T(\theta) = T_0 + A \sin^2(2\theta) \), with a positive coefficient \( A > 0 \).

The solution (b) yields a similar expression for the transmission:
\[
T(\theta) = 1 - R_0[1 + \gamma(2 + \sin^2(2\theta))].
\]

The solution (b) yields a negative sign for the amplitude, \( A < 0 \) of the four-fold oscillations. Since the experimental result [16] corresponds to the positive \( A > 0 \) (or solution (a)), one might suggest that the alternative dipole moments configuration (b) has a higher activation energy than the activation energy for the configuration (a). Consequently, in the experiment [16], only configuration (a) survives. Since the experimental result [16] corresponds to the positive \( A > 0 \) (or (a) solution), one might suggest that the alternative dipole moments configuration (b) has higher activation energy than the activation energy for the (a) configuration.

Additionally, we have compared the calculated amplitude for four-fold oscillations of transmission with the estimated experimental amplitude, \( A \sim 0.1 \). For this goal, we utilized the following parameters from literature: the bulk polarizability of GaAs, \( \alpha \approx 25 \times 10^{-25} \text{ cm}^3 \) [34, 35], \( E_0 = \sqrt{4\pi e^2/c} \approx 3.9 \times 10^4 \text{ eV} \text{ cm}^{-2} \), the incident beam intensity, \( I \approx 2 \times 10^4 \text{ W cm}^{-2} \) [16], the non-compensated dipole moment, \( d_0 \approx e\Delta r_{\text{eff}} \), the transition frequency, \( \nu = \Delta \nu / \hbar = 2.2 \times 10^{15} \text{ rad s}^{-1} \), the laser frequency \( \omega = 2\pi c/\lambda \approx 10^{15} \text{ rad s}^{-1} \), and the Rabi frequency, \( \Omega = d_0 E_0 / \hbar \approx 1.2 \times 10^4 \text{ rad s}^{-1} \). The substitution of these parameters into the equation (36), yields the amplitude of the oscillations, \( A \sim 0.13 \), which is close to the experimental \( A \sim 0.1 \) [16].

It would be instructive to estimate averaged over the time the fraction or the density of symmetry point defects or the dimensionless parameter \( \beta = \Delta N / N_0 \). On substituting the parameter \( N_0 = \rho/(M_{\text{Ga}} + M_{\text{As}}) \approx 2.2 \times 10^{22} \text{ cm}^{-3} \), with density, \( \rho \approx 5.3 \text{ g cm}^{-3} \), and the masses of atoms, \( M_{\text{Ga}} \approx 1.16 \times 10^{-22} \text{ g} \); \( M_{\text{As}} \approx 1.25 \times 10^{-22} \text{ g} \), into equation (22) we obtain \( \beta \approx 0.01 \).

5. Conclusions and discussion

We have shown that the two-level Quantum Optics model for an electron interacting with a laser field [29] adequately describes physics of the strong-field transitions in gallium arsenide semiconductor. This scheme is non-perturbative, i.e. valid for any coupling constant between the wave and the system. Consequently, we believe that the two-level Quantum Optics model can be useful for treatment non-linear effects in wave-crystal strong interactions.

In a consistent analysis of the problem, the transitions of electrons from the conduction band back to the valence band should be included into the scheme. As follow from our results, the inclusion of the time coordinate in the laser-crystal interaction picture provides key insights into the physics of interband transitions.

One should mention an early attempt to describe the process of \( VB \rightarrow CB \) transitions done by Keldysh [36]. In Keldysh’s model, the atomic physics approach for ionization of isolated atoms was utilized. In this scheme, the back transitions, \( CB \rightarrow VB \), are not taken into account and the time-dependent nature of the interaction of an electromag-netic wave with matter is ignored. We think that this oversimplified approach does not provide a consistent description for the interband transitions in the solid phase.

The time evolution of the electronic configuration in both affected bands was analyzed. Due to the strong-field-induced back and forth \( VB \rightarrow CB \) transitions, the electronic configurations, in both bands, experience dramatic changes. These changes include both the redistribution of the electrons between bands and opening of the breathing modes for their populations.

The mechanism of the optical anisotropy is explained as follows. For any polarization of the electric field vector \( \vec{E} \), at least 3 out of 4 different orientations of the non-compensated dipole moments, \( \vec{d} \), are non-collinear with the single electric field vector \( \vec{E} \). According to classical optical theory such a crystal should be optically anisotropic [1].

Based on the calculations, we have found that a single-crystal GaAs, in a strong IR field, experiences the first order transition from an optically isotropic phase to an optically anisotropic one.

The derived expression, (36), explicitly describes the anisotropic transmission in GaAs as a function of the laser beam polarization and radiation parameters. The amplitude of the transmission oscillations, \( A \), is proportional to the square of an amplitude of the electric field, \( A \sim E_0^2 \), and dramatically increases with
resonance, $A \sim (\Delta U_0 - \hbar \omega)^{-4}$, when the photon energy approaches the bandgap. Both the symmetry and the amplitude characteristic of the optical anisotropy are in agreement with the experimental data [16].

It is important to stress that the optical anisotropy is a strong-field induced second-order nonlinear effect with respect to the electric field $\vec{E}$. The symmetry defects of the electronic density, being included in consistent optical analysis, explain the experimental data on both qualitative and quantitative levels.

There are two alternative explanations for the strong-field induced optical anisotropy in cubic crystals [16, 37]. The approach developed in Ref. [37] provides an explanation for the small optical anisotropy of the single crystal Si, having Zinblende cubic structure. The mechanism of an optical anisotropy in this crystal is based on high-order optical effects which lead to more than one wave of the same frequency with different propagation properties. Since the optical anisotropy of Si, $\Delta n = n_{1(100)} - n_{1(101)} \sim 10^{-6}$, is $\sim 5$ orders of magnitude smaller than the measured optical anisotropy of GaAs, $\Delta n \sim 0.1$ [16], we believe that these high-order optical effects are irrelevant in our case.

An attempt to explain the optical anisotropy in GaAs based on an analysis of the curvature of a heavy-hole valence band structure, combined with Keldysh scheme [36], was made in [16]. Since the DFT calculations of the band curvature were done in [16] for an ideal lattice, this approach tacitly assumes that the optical anisotropy is an inherent property. Meantime, we know from both the experiment and the classical optical theory [1], that the ideal cubic crystal in a weak field is optically isotropic. Consequently, one might assume that the scheme [16] contradicts the optical principles. Additionally, this scheme totally ignores the time coordinate, the field-induced symmetry defects and $CB \rightarrow VB$ interband transitions.

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