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Influence of polarization of semiconductor lattice on its optical vibration spectrum

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Abstract. Considered in this paper are optical spectra of crystal lattice vibrations in semiconductor solid solutions, which have two vibration bands depending on solution composition. It has been shown that this dependence for the vibration spectrum is conditioned by polarization of this lattice, which is not taken into account by the traditional theory for the optical spectrum of lattice vibrations.

Keywords: polarization, vibration states of crystals, dielectric function, optical properties.

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1. Introduction

The experimental research of optical vibration spectra in the crystal lattice of semiconductor solid solutions $\text{GaAs}_x\text{P}_{1-x}$ and $\text{In}_x\text{Ga}_{1-x}\text{Sb}$ indicates the presence of two vibration bands depending on solution composition [1]. They are interpreted within the framework of two-oscillation model as oscillations of sublattices of the following two types [1], namely: vibrations of the GaAs sublattice and those of the GaP sublattice in the solid solution $\text{GaAs}_x\text{P}_{1-x}$. Besides, Ga vibrations take place in the InSb sublattice and vibrations of In in the sublattice of GaSb in the solid solution $\text{In}_x\text{Ga}_{1-x}\text{Sb}$. Experiments reveal the change in frequency of oscillator vibrations providing the oscillation mode of sublattice within the range of the low-frequency side of spectrum when increasing the concentration of oscillators related to this mode [1].

The same reconstruction of the vibration spectrum for crystal lattice of semiconductor solid solutions was observed in $\text{Zn}_{1-x}\text{Cd}_x\text{Se}$ [2, 3] and in $\text{Cu}_x\text{Ag}_{1-x}\text{GaS}_2$ [4] when changing compositions of solutions.

The physical cause of the above spectral reconstruction is not clear enough up to date. It has been shown in this work that this reconstruction is conditioned by polarization of the lattice, which is not taken into account by the traditional theory for optical spectra of lattice vibrations.

In the second section of the work, it has been shown that the traditional model of semiconductor

crystal lattice, which is used to examine its optical spectra, does not take into account the influence of polarization of this lattice on its vibrations.

In the third section of the work, it has been shown that the electrical conductivity that is used by this model is the external electrical conductivity. The internal electrical conductivity of the system of identical harmonic oscillators is derived in the third section of the work, too. This internal electrical conductivity can considerably differ from the external conductivity depending on the frequency of light. The indicated difference is conditioned by polarization of the system of harmonic oscillators, which is not taken into account by the traditional model of crystal lattice.

More exactly, the internal conductivity is included into the permittivity of matter and determines its optical properties [5, 6]. That difference appears when comparing the calculated optical properties of solids with experimental results [5, 6].

2. The traditional model of semiconductor crystal lattice

Let us consider the harmonic oscillator having the eigenfrequency ω_0 , the damping constant γ and the coordinate $\mathbf{x}(t)$ (where t is time). Let the variable electric field, which has the electric field strength $\mathbf{E}(t) = \mathbf{A} \exp(i\omega t)$, influences on it. Here, \mathbf{A} is the amplitude of this field, and ω is its frequency. The equation of motion for the oscillator in this field is [7, 8]

$$m_1 \frac{d^2 \mathbf{x}}{dt^2} + m_1 \gamma \frac{d\mathbf{x}}{dt} + m_1 \omega_0^2 \mathbf{x} = e_1 \mathbf{E}(t). \quad (1)$$

Here, m_1 is the effective mass of oscillator, e_1 is its effective charge. The solution of the equation (1) is

$$\mathbf{x}(t) = \frac{e_1}{m_1} \frac{1}{\omega_0^2 - \omega^2 + i\gamma\omega} \mathbf{E}(t). \quad (2)$$

Let us consider the model of semiconductor crystal lattice as a system of these harmonic oscillators. Let a unit of matter bulk has n oscillators, and the electric field $\mathbf{E}(t)$ acts on them in this matter. Then, the formula (2) gives the electric dipole moment of bulk unit for this matter $\mathbf{P}(t)$ (i.e. this matter polarization) in the following form:

$$\mathbf{P}(t) = \frac{e_1^2 n}{m_1} \frac{1}{\omega_0^2 - \omega^2 + i\gamma\omega} \mathbf{E}(t). \quad (3)$$

Let ε_0 is the frequency-independent component (background) of the semiconductor permittivity in the examined area of frequencies ω . If the external electric field that has the electric field induction $\mathbf{D}(t)$ acts on this matter, to ignore this matter polarization the electric field $\mathbf{E}(t)$ is expressed by the following formula:

$$\mathbf{E}(t) = \frac{\mathbf{D}(t)}{\varepsilon_0}. \quad (4)$$

In accordance with the formula (3), the electric current density $\mathbf{j}(t)$ in this matter, which is conditioned by oscillators and electric field $\mathbf{E}(t)$ (4), is given in this approximation by the following formula:

$$\mathbf{j}(t) = \frac{d}{dt} \mathbf{P}(t) = s(\omega) \mathbf{E}(t). \quad (5)$$

Here

$$s(\omega) = i\omega \varepsilon_0 \frac{\omega_p^2}{4\pi} \frac{1}{\omega_0^2 - \omega^2 + i\gamma\omega}, \quad (6)$$

$$\omega_p^2 = \frac{4\pi e_1^2 n}{\varepsilon_0 m_1}.$$

The physical quantity $s(\omega)$ is the electric conductivity of the considered system of harmonic oscillators.

The physical quantity $\varepsilon_S(\omega)$ expressed by the following formula is traditionally used for consideration of optical properties inherent to crystal lattice of semiconductor as this lattice permittivity [1, 7]:

$$\varepsilon_S(\omega) = \varepsilon_0 + \frac{4\pi}{i\omega} s(\omega). \quad (7)$$

The quantity $s(\omega)$ is given by the formula (6) here. The formula (7) does not take into account the polarization of semiconductor crystal lattice, and so it is approximate.

The substitution of the formula (6) in the expression (7) gives the following correlation:

$$\varepsilon_S(\omega) = \varepsilon_0 \left(1 + \omega_p^2 \frac{1}{\omega_0^2 - \omega^2 + i\gamma\omega} \right). \quad (8)$$

The following dimensionless oscillator strength is often used for the discussion of oscillator [1]:

$$f = \frac{e_1^2 n}{\varepsilon_0 m_1 \omega_0^2} = \frac{1}{4\pi} \frac{\omega_p^2}{\omega_0^2}. \quad (9)$$

3. Inserting the matter polarization to the permittivity

Optical properties of matter at the frequency of light ω are determined by its permittivity $\varepsilon(\omega)$. Applicability of the expression (7) for the permittivity of matter is based on the correlation (4) and does not take into account the shielding of the external field by charges of this matter (which are optically active at the examined frequency of light ω). These charges are under the action not of the external (in relation to them) field \mathbf{D}/ε_0 (4), but under the action of the internal field $\mathbf{E} \neq \mathbf{D}/\varepsilon_0$. The difference of the field \mathbf{E} from \mathbf{D}/ε_0 is given by the vector of matter polarization [8-14]:

$$\mathbf{P} = (\mathbf{D} - \varepsilon_0 \mathbf{E}) / (4\pi). \quad (10)$$

This difference is not taken into account at the derivation of the expressions (7), (8) for the permittivity $\varepsilon_S(\omega)$.

Indeed, the electric current density \mathbf{j} can be expressed both through the internal electric field \mathbf{E} and through the field \mathbf{D}/ε_0 [9, 10, 12-14]

$$\mathbf{j}(t) = \sigma(\omega) \mathbf{E}(t) = s(\omega) \mathbf{D}(t) / \varepsilon_0. \quad (11)$$

The electric conductivity $s(\omega)$ gives the response of electric current on the external field \mathbf{D}/ε_0 (in relation to charges of matter, which are optically active for the examined frequency of light ω), but the conductivity $\sigma(\omega)$ gives the response of electric current on the internal field \mathbf{E} . Therefore, $s(\omega)$ is named as the external electrical conductivity, while $\sigma(\omega)$ is named the internal one [12-14]. The difference between $\sigma(\omega)$ and $s(\omega)$ in the expression (11) is given by the difference of the polarization \mathbf{P} (10) from zero.

The charges of matter that do not take part in optical transitions within the studied interval of frequencies make the frequency-independent background contribution ε_0 to the permittivity of this matter [8, 11, 15]. Therefore, the expression for permittivity of this matter $\varepsilon(\omega)$ can be presented in the following form:

$$\varepsilon(\omega) = \varepsilon_0 + \frac{4\pi}{i\omega} \sigma(\omega). \quad (12)$$

The expression (12) follows from the correlation

$$\mathbf{D}(\omega) = \varepsilon(\omega)\mathbf{E}(\omega), \quad (13)$$

which is the definition of matter permittivity $\varepsilon(\omega)$, and follows from the interrelation (10) for the vector of matter polarization \mathbf{P} [8-14]. The relationship

$$\mathbf{j}(t) = \frac{d}{dt} \mathbf{P}(t)$$

is utilized in this case. This relationship arises from definition of the polarization vector \mathbf{P} as the density of dipole moments of charges of this matter [8, 11, 16]:

$$\mathbf{P}(t) = \frac{1}{V} \sum_{i=1}^N q_i \mathbf{x}_i(t). \quad (14)$$

Here V is the matter volume, N is the total number of charges of this matter, q_i is the charge of i -th charge carrier, $\mathbf{x}_i(t)$ is its coordinate.

The definition of matter permittivity requires σ being in it, but not s [9-14]. Further, for simplicity, let us suppose the quantities σ and s as the scalar ones, but not as the tensor ones. The formulas (11), (13) give the interrelation

$$\sigma(\omega) = s(\omega)\varepsilon(\omega) \frac{1}{\varepsilon_0}. \quad (15)$$

Furthermore, the formulas (12), (15) give the following

$$\sigma(\omega) = \frac{1}{1 - \frac{4\pi}{i\omega\varepsilon_0} s(\omega)} s(\omega). \quad (16)$$

Having substituted the expression (16) into the formula (12), we have

$$\varepsilon(\omega) = \varepsilon_0 \frac{1}{1 - \frac{4\pi}{i\omega\varepsilon_0} s(\omega)}. \quad (17)$$

The expression for the permittivity $\varepsilon(\omega)$ (17) that takes into account polarization of matter substantially differs from the approximate expression for the permittivity $\varepsilon_s(\omega)$ (7), in which matter polarization is not taken into account. Expansion of the right side of the expression (17) in series by powers of $s(\omega)$ gives that the permittivity $\varepsilon(\omega)$ coincides with $\varepsilon_s(\omega)$ only when $s(\omega)$ is sufficiently small. The formula (10) allows using the electrical conductivity s that is obtained without regard for polarization, under consideration of matter optical behavior.

4. Account of polarization in optical behavior of matter crystal lattice

The knowledge of the internal conductivity for a system of harmonic oscillators $\sigma(\omega)$ is necessary to investigate

optical behavior of this system. It can be derived by substitution of the external conductivity of this system $s(\omega)$ given by the formula (6) into the formula (17). The result of this substitution is expressed by the following expression:

$$\sigma(\omega) = i\varepsilon_0 \omega \frac{\omega_p^2}{4\pi} \frac{1}{\Omega^2 - \omega^2 + i\gamma\omega}, \quad (18)$$

where

$$\Omega^2 = \omega_0^2 \left(1 - \left(\frac{\omega_p}{\omega_0} \right)^2 \right). \quad (19)$$

The substitution of internal conductivity (18) into the formula (12) gives the permittivity of system of harmonic oscillators in the form of:

$$\varepsilon(\omega) = \varepsilon_0 \left(1 + \omega_p^2 \frac{1}{\Omega^2 - \omega^2 + i\gamma\omega} \right). \quad (20)$$

5. Discussion

The formula (20) for the permittivity $\varepsilon(\omega)$ of the system of harmonic oscillators, which takes into account the polarization of this system, differs from the approximate formula (8) for the permittivity $\varepsilon_s(\omega)$, which does not take into account this polarization, by the replacement of the frequency ω_0 by another frequency Ω in the denominator. This frequency is given by the formula (19) and diminishes with the growing concentration of harmonic oscillators n . This theoretical result coincides with the experimental ones for optical spectra of crystal lattice [1-4].

The formula (20) gives the change of frequency of a harmonic oscillator under the action of polarization of matter where it is. This polarization is created by all the oscillators in this matter. Thus, taking into consideration polarization of the system of non-interacting harmonic oscillators reduces to the interaction between these oscillators and changes their frequencies. It can be explained by the following simple microscopic consideration.

In compliance with the formula (10), the field \mathbf{E} in matter is given by the following expression:

$$\mathbf{E} = \frac{1}{\varepsilon_0} (\mathbf{D} + 4\pi\mathbf{P}). \quad (21)$$

Light and vibrations of the crystal lattice are examined by the method used above under the longwave approximation. Therefore, the vector of polarization for the system of identical oscillators in crystal lattice \mathbf{P} (14) can be examined approximately in the following form:

$$\mathbf{P}(t) = \frac{1}{V} \sum_{i=1}^N e_1 \mathbf{x}_i(t) \approx \frac{e_1}{V} N \mathbf{x}(t) = e_1 n \mathbf{x}(t). \quad (22)$$

Here, $\mathbf{x}(t)$ is the coordinate of one oscillator.

In compliance with the formulas (21), (22), the field in matter \mathbf{E} that acts on an oscillator has the form of

$$\mathbf{E}(t) = \frac{1}{\varepsilon_0} \mathbf{D}(t) + m_1 \omega_p^2 \mathbf{x}(t). \quad (23)$$

Therefore, the equation of motion of one oscillator in the field $\mathbf{E}(t)$ (23), where the polarization is taken into account, has the following form:

$$m_1 \frac{d^2 \mathbf{x}}{dt^2} + m_1 \gamma \frac{d\mathbf{x}}{dt} + m_1 \Omega^2 \mathbf{x} = e_1 \frac{1}{\varepsilon_0} \mathbf{D}(t). \quad (24)$$

The solution of equation (24) for the field $\mathbf{D}(t) \propto \exp(i\omega t)$ is

$$\mathbf{x}(t) = \frac{e_1}{m_1} \frac{1}{\Omega^2 - \omega^2 + i\gamma\omega} \frac{1}{\varepsilon_0} \mathbf{D}(t), \quad (25)$$

where the frequency $\Omega \neq \omega_0$ and is given by the formula (19).

The similar results of the influence of internal field on vibrations of crystal lattice are known from vast literature of the end of the 19th and the beginning of the 20th centuries [17]. However, vibrations of ions were here examined near lattice sites of crystal lattice. Moreover, these ions were examined being in the cavity of dielectric. The local electric field in such cavity \mathbf{E} differs from the external electric field \mathbf{D} on Lorentz-Lorenz correction [17] and depends on the form of this cavity. The following correlation takes place for the spherical form of cavity [17]:

$$\mathbf{E} = \frac{1}{\varepsilon_0} \left(\mathbf{D} + \frac{1}{3} 4\pi \mathbf{P} \right). \quad (26)$$

However, the distinction between formulas (21) and (26) is, which depends on the form of cavity. In addition, because of the transmission symmetry of crystal lattice, vibrations of ions near its lattice sites create the collective oscillation, which covers all the lattice [18]. Exactly, light interacts with such vibrations (phonons) under the longwave approximation that is valid for the infrared spectral range. Therefore, the approach expounded in this work seems to be more preferable.

The force of oscillator (9) is not utilized when deriving the formulas in this work, because the eigenfrequency of oscillator ω_0 is included in it. However, the frequency of harmonic oscillator changes under the action of polarization of matter, in which it is, and takes on the value $\Omega \neq \omega_0$. Therefore, for simplicity, the force of oscillator is not utilized.

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