Gyrotropy and magneto-spatial dispersion effects at intersubband transitions in quantum wells

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Abstract – Gyrotropic properties of multiple quantum well structures are studied theoretically. The symmetry analysis is performed yielding the gyrotropy tensor components for structures grown along the [001], [110] and [311] crystallographic directions. Angular dependences of circular dichroism and natural optical activity signals are established. The phenomenological model and microscopic theory based on spin-orbit splitting of size-quantized subbands are developed for photon energies close to the energy of the intersubband optical transition. Magneto-spatial dispersion effects arising from the diamagnetic shift of the intersubband energy gap linear in the electron momentum are also considered. It is demonstrated that the spectral dependence of the gyrotropy and magneto-spatial dispersion constants represents an asymmetrical peak with a degree of asymmetry governed by the mean electron energy. The estimates show that the considered effects are detectable in experiments.

Introduction. – While frequency dispersion of dielectric response of media is present in all wavelength ranges, spatial dispersion, i.e., dependence of the dielectric permittivity on light wave vector at a fixed frequency, manifests itself near resonant absorption lines. The brightest example are excitons known by their strong spatial dispersion effects [1]. New resonances present in the absorption of nanostructures owing to transitions between size-quantized levels should result in spatial dispersion effects in infrared and TeraHertz ranges.

A spatial dispersion of the first order called gyrotropy manifests itself in a variety of phenomena, absent in usual optical experiments [1–3]. First, linearly polarized light transmitted through gyrotropic media gets elliptically polarized, and the main axis of the polarization ellipse is rotated relative to the polarization plane of the incident light like in the Faraday effect. The polarization plane rotation is named natural optical activity because it takes place in the absence of an external magnetic field. Second, absorption in gyrotropic media is circularly dichroic, i.e., it is different for the right and left circularly polarized light. Finally, linearly polarized light becomes elliptically polarized after reflection from the gyrotropic system representing the Kerr effect. All these phenomena are usually studied in an external magnetic field, but the symmetry of gyrotropic media allows for such effects even at zero magnetic field. The role of the field is played by the light wave vector which has components transforming according to the same space group representations as a pseudovector. In order to be gyrotropic, the system should lack the space inversion center. Moreover, out of 21 non-centrosymmetric crystal classes only 18 allow gyrotropy while media with point group symmetry $T_d$, $C_{3h}$ and $D_{3h}$ are non-gyrotropic. In systems with $C_{3v}$, $C_{4v}$ and $C_{6v}$ symmetry, only the Kerr effect is allowed while the Faraday effect and helicity-dependent absorption are not present. These media are referred to as “weakly gyrotropic”.

Gyrotropy of bulk semiconductors has been studied in tellurium under interband excitation [4,5] and in weakly gyrotropic $A_2B_6$ semiconductors of wurtizte type as CdS near excitonic resonances [6]. Gyrotropy in bulk GaAs with $T_d$ point symmetry can be induced by application of deformation lowering symmetry to $D_{2d}$ [7], or at a surface of a semiconductor [8]. The situation is different for two-dimensional semiconductors because all quantum well (QW) structures are gyrotropic. Gyrotropy is present in QWs grown from any $A_2B_6$ or $A_3B_5$ material [9].

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However, the gyrotyropy of QWs was probed only in photogalvanic experiments [9] while the all-optical study of this phenomenon is absent up to date.

Gyrotyropy is described by a contribution to the dielectric permittivity linear in the light wave vector $q$ [1–3]:

$$\delta \varepsilon_{\lambda \mu} = i \gamma_{\lambda \mu \nu} q_{\nu}. \quad (1)$$

Here $\gamma(\omega)$ is a third-rank tensor antisymmetric with respect to the first two indices. Microscopically, gyrotyropy of multiple QW structures (MQWs) is caused by the spin-orbit splitting of the electron energy spectrum. The splitting is present due to the asymmetry of the heteropotential or the absence of inversion center in the bulk material. It is described by the Rashba and two-dimensional Dresselhaus contributions to the Hamiltonian of spin-orbit interaction given by $k$-linear terms

$$H_{so}(k) = \beta_{\mu \rho} \sigma_{\rho \kappa} k_{\nu}, \quad (2)$$

where $k$ is a 2D electron wave vector and $\sigma_{\mu}$ are the Pauli matrices. The gyrotyropy tensor $\gamma$ is related to the pseudotensor $\beta$ via

$$\gamma_{\lambda \mu \nu} \sim \epsilon_{\lambda \mu \rho} \beta_{\rho \nu},$$

where $\epsilon$ is the totally antisymmetric third-rank tensor.

Recently the optical activity of non-centrosymmetric metals caused by a spin-orbit interaction of such kind has been studied theoretically [10] motivated by experimental results [11]. In metals the spin-orbit splitting far exceeds the photon energy, and the effect is inversely proportional to the spin-orbit splitting: $\gamma \sim 1/\beta$ [10]. In semiconductor MQWs the situation is opposite, and the correction to the dielectric permittivity is linear in the small spin-orbit splitting.

In the presence of a magnetic field $B$, the dielectric permittivity acquires additional terms linear in both $B$ and $q$ which describe the magneto-spatial dispersion:

$$\delta \varepsilon^{(B)}_{\lambda \mu} = A_{\lambda \mu \nu} B_{\nu} q_{\nu}. \quad (3)$$

Magneto-spatial dispersion is allowed in all non-centrosymmetric media, therefore it is present in MQWs. Magneto-spatial dispersion results in magnetic-field–induced birefringence. In bulk semiconductors this effect has been investigated in both A$\text{gB}_{\text{b}}$ and A$\text{gB}_{\text{s}}$ materials [12,13]. In MQW structures, the magneto-spatial dispersion is caused by the $k$-linear diamagnetic shift of subband dispersions described by the Hamiltonian [14]

$$H_{\text{dia}}(k) = \alpha_{\mu \nu} B_{\mu} k_{\nu}. \quad (4)$$

We consider photon energies close to the peak of intersubband absorption in MQWs.

**Symmetry consideration.** – (001)-grown symmetrical QWs have $D_{2d}$ symmetry with reflection planes perpendicular to the in-plane axes $x || [110]$ and $y || [110]$.

The gyrotyropic corrections to the dielectric tensor of MQWs are described by one linearly independent constant:

$$\delta \varepsilon_{xx} = i \gamma_{xz}, \quad \delta \varepsilon_{yy} = -i \gamma_{y z}. \quad (5)$$

These terms result in circular dichroism: for light propagating in the structure plane, the absorbance has a contribution

$$\eta \propto \text{Im} \gamma P_{\text{circ}} q_{\nu} q_{\nu}, \quad (6)$$

where $P_{\text{circ}}$ is the circular polarization degree of light. The real part of $\gamma$ determines the natural optical activity signal. Both effects are maximal for light propagating along an in-plane cubic axis.

Asymmetrical (001)-grown QWs have $C_{2v}$ symmetry with the $C_2$ axis parallel to the growth direction $z$. Gyrotyropy in this case is described by two constants:

$$\delta \varepsilon_{xz} = i \gamma_{z}, \quad \delta \varepsilon_{yz} = -i \gamma'_{z}. \quad (7)$$

Change of light polarization in transmission experiments is described by the sum of these constants:

$$\eta \propto \text{Im} (\gamma + \gamma') P_{\text{circ}} q_{\nu} q_{\nu},$$

while their difference describing weak gyrotyropy can be probed in reflection experiments.

Symmetrical (110)-grown MQWs have $C_{2v}$ symmetry as well, but the $C_2$ axis lies in the QW plane: $C_2 || y$, where $y || [001]$. Therefore, the following corrections are present:

$$\delta \varepsilon_{xy} = -i \gamma_{1} q_{z}, \quad \delta \varepsilon_{yz} = -i \gamma_{2} q_{z}. \quad (8)$$

Here $z || [110]$ is the growth direction, and $x || [110]$. In transmission experiments on can measure the following angular dependence:

$$\eta \propto \text{Im} (\gamma_1 + \gamma_2) P_{\text{circ}} q_{\nu} q_{\nu}. \quad (9)$$

If the (110)-grown QW is asymmetric, then it belongs to the class $C_s$ with only one mirror reflection plane ($yz$). In this case two additional gyrotyropic contributions $\delta \varepsilon_{zz}$ are allowed by symmetry.

(113)-grown QWs also have $C_s$ symmetry therefore the above relations (7), (8) are valid for them if one chooses the axes as $x || [110]$, $y || [332]$, $z || [113]$. Magneto-spatial dispersion effects manifest themselves as corrections to $\varepsilon_{zz}$:

$$\delta \varepsilon^{(B)}_{zz} = A_{zz1\mu} B_{\mu} q_{\nu}. \quad (10)$$

Real and imaginary parts of $A_{zz1\mu}$ result in magnetic-field–induced birefringence and $B$-dependent absorption, respectively.

In (001) QWs of $C_{2v}$ symmetry the correction has the form

$$\delta \varepsilon^{(B)}_{zz} = A_1 B_z q_x + A_2 B_y q_x. \quad (11)$$

If the MQW structure is symmetric ($D_{2d}$ point group) then $A_1 = A_2$. 

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For symmetrical (110) QWs we have
\[ \delta \varepsilon_{zz}^{(B)} = A_3 B_z q_x + A_4 B_z q_z. \] (12)

In (311) MQWs all four constants \( A_{1-4} \) are nonzero and different from each other.

**Microscopic model.** – If we ignore the photon momentum \( q = 0 \), the intersubband absorption is due to direct optical transitions. Electron wave vectors in the initial state in the ground subband \( \mathbf{k} \) and in the final state in the excited subband \( \mathbf{k}' = \mathbf{k} \). If we also neglect spin-orbit splitting \( \langle \beta_{\mu\nu} = 0 \rangle \), only photons with energy \( \hbar \omega = E_{21} \) can be absorbed. Here \( E_{21} \) is the energy separation between the ground and the excited subbands, and we assume parabolic energy dispersions \( E_1(k) = E_2(k) = \hbar^2 k^2 / (2m) \) with equal effective masses in the subbands. Therefore the spectrum of intersubband absorbance is a sharp peak centered at \( E_{21} \):

\[ \eta = \eta_0(\hbar \omega - E_{21}), \]

where \( \eta_0(x) \) is a broadened \( \delta \)-function.

The account for a final photon momentum makes the optical transitions indirect: \( \mathbf{k}' = \mathbf{k} + \mathbf{q} \). Therefore the energy conservation reads

\[ \hbar \omega = E_{21} + \frac{\hbar^2 \mathbf{k} \cdot \mathbf{q}}{m} + \frac{\hbar^2 q_z^2}{2m}. \] (13)

The presence of the second term here leads to Doppler broadening of the absorption peak [15], and the third term in eq. (13) makes a shift of the peak to higher energies. As a result, the absorbance takes the following form:

\[ \eta(\hbar \omega - E_{21}) = \eta_0 - \frac{\hbar^2 q_z^2}{2m} \left[ \frac{\partial}{\partial (\hbar \omega)} - \frac{\partial^2}{\partial (\hbar \omega)^2} \right] \eta_0, \] (14)

where \( \mathcal{E} \) is the mean kinetic energy. Hereafter we assume filling of the ground subband only.

If we include the spin-orbit interaction (2), the energy dispersions in the subbands of size quantizations become splitted parabolas as is shown in fig. 1(a), (b). For (110) and (311) QWs the term \( \beta_{xz} \sigma_y k_z \) is present in the spin-orbit Hamiltonian (2), therefore the subbands for electrons with spin projection \( \pm 1/2 \) onto the \( z \)-axis are splitted. The selection rules for intersubband absorption of light propagating along the \( z \)-direction yield a higher transition rate from the \( | -1/2 \rangle_z \) state in the ground subband to the \( | +1/2 \rangle_z \) state in the excited subband for \( \sigma_+ \) radiation, while for left-handed polarization the transition between two other states is more probable [16]. This is illustrated in fig. 1(a). As a result, we have in the energy conservation law eq. (13) instead of the \( q_x \) component

\[ q_x \rightarrow q_x + \eta_0 k_{so}. \] (15)

Here the upper and lower signs correspond to \( \sigma_+ \) and \( \sigma_- \) polarizations, \( \mathbf{n} = \mathbf{q}/q \) is a unit vector in the light propagation direction, and \( k_{so} = (\beta_{yz}^{(1)} - \beta_{yz}^{(2)}) \mathbf{m} / \hbar^2 \), where \( \beta_{yz}^{(1,2)} \) are the corresponding spin-orbit constants for the first and the second subbands.

For (001) QWs the term \( \beta_{xy} \sigma_y k_x \) splits the states with a definite spin projection onto the \( y \)-axis. According to the selection rules, “spin-conserving” transitions between the states with the same spin projection are more probable for light with \( \mathbf{n} \parallel y \) as is shown in fig. 1(b). Therefore the replacement eq. (15) again takes place, but with \( k_{so} = (\beta_{yz}^{(1)} - \beta_{yz}^{(2)}) m / \hbar^2 \) and with \( n_y \) instead of \( n_z \).

Substituting eq. (15) into the \( q_z^2 \)-contribution in eq. (14), we obtain the circular dichroism of the intersubband absorption:

\[ \eta_{\sigma_+} - \eta_{\sigma_-} \propto k_{so} q_x n_{z,y} \left[ \frac{\partial}{\partial (\hbar \omega)} - \frac{\partial^2}{\partial (\hbar \omega)^2} \right] \eta_0(\hbar \omega - E_{21}), \] (16)

where the factor \( n_z \) should be taken for (110), (311) QWs, and \( n_y \) is for (001) QWs. The angular dependence of the circular dichroism (16) coincides with eqs. (6) and (9) obtained from symmetry arguments.

In the presence of a magnetic field, the diamagnetic interaction, eq. (4), results in the spin-independent shift of the second subband relative to the first one making the energy separation \( \mathbf{k} \)-dependent:

\[ E_{21}(\mathbf{k}) = E_{21} + \alpha_{\mu\nu} B_k k_{\mu\nu}. \] (17)
Therefore, similar to eq. (15) we have

\[ q_z \to q_z + k_{\text{dia}}, \]

where \( k_{\text{dia}} = \alpha_{\mu z} B_{\mu} m / \hbar^2 \), fig. 1(c). For (110) and (311) QWs \( \alpha_{xz} \neq 0 \) so that \( k_{\text{dia}} \propto B_z \), while for (001) QWs \( \alpha_{yz} \neq 0 \) and \( k_{\text{dia}} \propto B_y \). As a result, we obtain the contribution to the absorbance

\[ \Delta \eta(B) \propto q_z B_{z,y} \left[ \frac{\partial}{\partial (\hbar \omega)} - \frac{\partial^2}{\partial (\hbar \omega)^2} \right] \eta_0 (\hbar \omega - E_{21}). \]  

(18)

This expression corresponds to the phenomenological eqs. (11) and (12).

**Theory.** – Now we develop a rigorous expression for gyrotropy and magneto-spatial dispersion constants in MQW structures. The resonant contribution to the gyrotropy tensor has the form [4]

\[ \gamma_{\lambda \mu \nu} = \frac{4 \pi}{\omega^2 d} \lim_{q \to 0} \frac{\partial}{\partial q_\nu} \sum_{k,s,l} j_{1,s,l}^{1} j_{2,l,s}^{\mu} f_s(k) E_{21}(k + q) - E_{21}(k) - \hbar \omega - i \Gamma. \]  

(19)

Here \( d \) is a period of the MQW structure, the indices \( s,l \) enumerate eigenstates in the ground and the excited subbands found with account for the spin-orbit interaction, \( j \) is the electric current operator, \( \Gamma \) is the half-width of the absorption peak, and \( f_s(k) = f_0(E_{1s}(k)) \), where \( f_0(E) \) is the Fermi-Dirac distribution function. On the basis of these states

\[ E_{1s}(k) = E_k + H_0^{(1)}(k), \quad E_{21}(k) = E_{21} + E_k + H_0^{(2)}(k), \]

where \( \hat{H}^{(1,2)}(k) \) are operators of the spin-orbit interaction \( (2) \) for the first and the second subbands, and \( E_k = h^2 k^2 / 2m \). In eq. (19) we assume that the electric current operators for intersubband transitions are independent of the wave vector.

Expanding eq. (19) to the linear order in the spin-orbit interaction and performing summation over \( k \) we obtain

\[ \gamma_{\lambda \mu \nu} = - \frac{i}{2} \text{Tr} \left[ \frac{\partial \hat{H}^{(2)}}{\partial k_\nu} \hat{L}_{\mu} - \frac{\partial \hat{H}^{(1)}}{\partial k_\nu} \hat{L}_{\lambda} \right] F(\omega). \]  

(20)

Here \( \hat{L} \) is defined according to \( \hat{j} = \hat{L} \omega \nu \), where \( \nu^2 = -(i \hbar / m) \partial / \partial z \),

\[ F(\omega) = \left[ \frac{\partial}{\partial (\hbar \omega)} - \frac{\partial^2}{\partial (\hbar \omega)^2} \right] \epsilon_{21}(\hbar \omega), \]  

(21)

the resonant contribution to the dielectric permittivity is given by

\[ \epsilon_{21}(\hbar \omega) = \frac{4 \pi N}{\omega^2 d E_{21}} \frac{e^2 |\epsilon|^2}{\epsilon_{21}(\hbar \omega) - i \Gamma}. \]  

(22)

\( N \) is the 2D electron concentration, and the mean energy is defined as \( \bar{E} = 2 \sum_k E_k f_0(E_k) / N \).

The operators \( \hat{L} \) for intersubband transitions have the form [16]

\[ \hat{L} = (i \Lambda \hat{\sigma}_y - i \Lambda \hat{\sigma}_x \hat{I}). \]  

(23)

Here \( \hat{I} \) is the \( 2 \times 2 \) unit matrix, and

\[ \Lambda = \frac{E_{21} \Delta (2E_y + \Delta)}{2E_y (E_y + \Delta)(3E_y + 2\Delta)}, \]

where \( E_y \) and \( \Delta \) are the energy gap and the spin-orbit splitting of the valence band in the bulk semiconductor, respectively.

For (001) QWs eqs. (20) and (23) yield

\[ \gamma = \Lambda(\beta_{yy}^{(2)} - \beta_{yy}^{(1)}) F(\omega), \]

\[ \gamma' = \Lambda(\beta_{xx}^{(2)} - \beta_{xx}^{(1)}) F(\omega), \]  

(24)

and for (110) and (311) QWs we obtain

\[ \gamma_1 = \Lambda^2(\beta_{xx}^{(2)} + \beta_{xx}^{(1)}) F(\omega). \]  

(25)

The remaining constant \( \gamma_2 \ll \gamma_1 \) because the two-dimensional electrons do not feel the normal component of the photon momentum \( q_z \), which is much smaller than the inverse MQW structure period \( \frac{1}{d} \).

Equation (19) is also valid for calculation of the value \( -i \Lambda_{\mu \nu \rho \sigma} B_{\rho} \). Taking into account the diamagnetic interaction, eq. (4), we obtain

\[ A_{\mu \nu \rho} = \alpha_{\mu \rho} F(\omega). \]  

(26)

For (001) QWs this leads to eq. (11) with

\[ A_1 = \alpha_{yy} F(\omega), \quad A_2 = \alpha_{yz} F(\omega). \]

(27)

For (110) and (311) QWs we obtain eq. (12) where

\[ A_3 = \alpha_{xz} F(\omega), \]

(28)

and \( A_4 \ll A_3 \).

**Discussion.** – Equations (24), (25) and (27), (28) demonstrate that the spectral dependence of the gyrotropy constants \( \gamma_1 \), \( \gamma' \) and \( \gamma_1 \) as well as of the magneto-spatial dispersion constants \( A_{1,...,3} \) is given by the function \( F(\omega) \), eq. (21). However, \( \gamma \) and \( \gamma' \) are determined by a difference of corresponding spin-splitting constants in the excited and ground subbands, while \( \gamma_1 \) is proportional to the sum \( \beta_{xx}^{(2)} + \beta_{xx}^{(1)} \). All these findings agree with the microscopic model, cf. eqs. (21) and (16). The magneto-spatial dispersion in all cases is given by \( e_{\lambda \mu} \), which describes the shift of the second subband dispersion relative to the first one. The dependences \( \text{Im} F(\omega) \) and \( \text{Re} F(\omega) \) presented in fig. 2 demonstrate sharp spectral changes in the resonance region. The relation between the mean energy \( \bar{E} \) and the absorption peak half-width \( \Gamma \) governs the asymmetry of the resulting spectra of the gyrotropy and magneto-spatial dispersion constants.

Note that \( k \)-linear spin-dependent terms are also possible in the optical transition operators [17]. They yield an additional contribution to the gyrotropy tensor with the frequency dependence coinciding with \( \epsilon_{21}(\hbar \omega) \), eq. (22).
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In order to estimate a magnitude of the effect, we note that in real systems \( \Gamma \sim \overline{E} \sim 10 \text{ meV} \) \[16\], therefore for (001) MQWs we have

\[ \delta \varepsilon \sim \Lambda \frac{\beta q}{\Gamma} \epsilon_{21}, \]

and for GaAs-based QWs with \( \beta q \sim 10^{-3} \text{ meV}, \Lambda \sim 10^{-2}, N = 10^{12} \text{ cm}^{-2}, h\omega \sim E_{21} \sim 100 \text{ meV}, \) the experimentally measured quantity \( \delta \varepsilon d\omega n/c \sim 10^{-7} N \), where \( N \) is a number of QWs in the MQW structure. In multi-pass geometry this value may be increased by an order of magnitude. Such signals can be detected in experiments. The estimate for (110) and (311) QWs has an additional small factor \( \Lambda \) which is partially compensated by a larger spin-orbit splitting constant \( \beta_{zx} \). The situation is more favourable in \( p \)-type structures where the selection rules for circular polarization yield \( \Lambda \sim 1 \). However, the intersubband absorption peak is broadened in \( p \)-type QWs due to the complicated band structure of the valence band.

The magneto-spatial dispersion effects can be estimated as

\[ \delta \varepsilon^{(B)} \sim \frac{\alpha B q}{\Gamma} \epsilon_{21}. \]

The main contribution to the diamagnetic \( k \)-linear shift in (001) QWs is given by \( \alpha_{xy} = -\alpha_{yx} = (z_2 - z_1) \frac{\partial}{\partial z}, \) where \( z_n \) is the coordinate matrix element in the \( n \)-th subband. Estimation for \( z_2 - z_1 = 10 \AA, d = 100 \AA \) and \( B = 1 \text{T} \) yields \( \delta \varepsilon^{(B)} d\omega n/c \sim 10^{-5} N \).

**Conclusion.** In conclusion, we have studied the gyrotropic and magneto-spatial properties of MQW structures showing up at intersubband optical transitions. The phenomenological model and microscopic theory are developed for MQWs of various crystallographic orientations. Gyrotropy is shown to be caused by spin-orbit splitting of size-quantized subbands while magneto-spatial dispersion arises from the diamagnetic \( k \)-linear shift of the intersubband energy gap. It is demonstrated that the spectral dependence of the gyrotropy and magneto-spatial dispersion constants represents an asymmetrical peak with a degree of asymmetry governed by the mean electron energy. The estimates show that the considered effects are within the experimental sensitivity.

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