Hidden In-Plane Anisotropy of Interfaces in Zn(Mn)Se/BeTe Quantum Wells with a Type-II Band Alignment

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Circularly and linearly polarized radiation due to spatially indirect optical transitions is studied in semimagnetic (Zn,Mn)Se/BeTe and nonmagnetic ZnSe/BeTe quantum-well structures with a type-II band alignment. Because of the giant in-plane anisotropy of the optical matrix elements related to a particular interface, complete spin orientation of photocarriers induced by magnetic fields leads not to purely circular but instead to elliptical polarization of the luminescence. From comparison between theory and experiment the parameter of optical anisotropy of a ZnSe/BeTe interface is evaluated. The developed theoretical approach can be applied for the large class of nanostructures revealing optical anisotropy.

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A single heterojunction between two semiconductors with a zinc-blende lattice grown along the [001] crystallographic axis is anisotropic at the interface plane. The lateral anisotropy is related to the tetrahedral orientation of the chemical bonds along the $\langle 111 \rangle$ directions and can be described by the reduced point-group symmetry C_{2v}. On the other hand, an ideal quantum well (QW) structure with two equivalent interfaces has the higher symmetry group D_{2d}. It is uniaxially isotropic, because the chemical bonds at the opposite interfaces lie in mutually orthogonal planes [$(1\bar{1}0)$ and (110)] and their contributions to the anisotropy cancel. Thus, in ideal QW structures the *in-plane anisotropy of single interfaces is hidden*. However, under special conditions it can reveal itself, e.g., through a linear polarization of vertically emitted radiation. Such is the case in the presence of an electric field applied normal to the plane of the well (the quantum confined Pockels effect [1–3]) or in asymmetric QW's with different profiles of nonabrupt normal and inverted interfaces, and/or for heteropairs with no common atoms, e.g., InAs/AlSb and ZnSe/BeTe, with different kinds of chemical bonds at the abrupt interfaces [4–10].

In this paper we propose a method to uncover the hidden in-plane anisotropy of ideal QW's without breaking their invariance to the mirror-rotation operation S_4 and reducing the uniaxial symmetry. The method is based on the analysis of the magnetic-field-induced elliptical polarization of the photoluminescence (PL) and can be applied, in general, for a physical system consisting of optically anisotropic subsystems. Equilibrium spin polarization of the photoexcited subsystems in an external magnetic field applied in the Faraday configuration is an important ingredient of the method. It will be shown that the measured circular polarization of PL reveals the optical anisotropy of individual subsystems even in the extreme case where the observed luminescence has no linear polarization. We present both a theoretical basis and an experimental realization for the proposed method. In order to demonstrate its potentiality we chose Zn(Mn)Se/BeTe QW structures with a type-II band alignment (schema in Fig. 1), an object which is new and arouses itself a wide interest [8,11]. Its main specific feature is large conduction and valence band offsets, which result in a very small penetration of the carrier wave functions into the neigboring layers. In this case the band-edge optical transitions are in fact indirect in real space and involve conduction band electrons confined in ZnSe layers and valence band holes confined in neigboring BeTe layers (see Fig. 1). Because of the high barriers, the matrix elements for these transitions are extremely "interface sensitive." The role of two anisotropic subsystems mentioned above is played by electron-hole pairs attached either to normal (N, BeTe on ZnSe) or to inverted (I, ZnSe on BeTe) interfaces related by the symmetry operation S_4. Giant optical lateral anisotropy of ZnSe/BeTe structures has been reported recently [3,8], the PL linear polarization related to a particular interface amounts to 70%–80% and is weakly sensitive to photocarrier concentration and temperatures.

We discuss here results for two samples grown by molecular beam epitaxy on (100) GaAs substrates, however, the data were always verified on several different samples. The first sample is a 200 Å/100 Å $Zn_{0.97}Mn_{0.03}Se$/BeTe multiple quantum well (MQW) structure with 20 periods. A small fraction of magnetic Mn ions inserted in the $Zn_{0.97}Mn_{0.03}Se$ layers allows the achievement of complete spin polarization of carriers in weak magnetic fields ($B < 0.3$ T), due to the giant Zeeman splitting effect [11]. The second sample is a nonmagnetic 100 Å/50 Å ZnSe/BeTe MQW also with 20 periods. The Γ-point band gaps of ZnSe and BeTe at low temperatures are 2.8 and 4.5 eV, respectively. For both structures, growth conditions were chosen to form

Zn-Te (not Se-Be) chemical bonds at interfaces. UV lines of an Ar-ion laser (3.5 eV) were used for photoexcitation. This implies the laser light was absorbed only in ZnSe (or $Zn_{0.97}Mn_{0.03}Se$) layers. Linearly and circularly polarized PL spectra were recorded at a sample temperature of $T = 1.6$ K in magnetic fields up to 8.5 T applied in the Faraday configuration, parallel to the structure growth axis (z axis).

First we consider experimental results obtained on the semimagnetic $Zn_{0.97}Mn_{0.03}Se/BeTe$ structure. In Fig. 1a spatially direct (D) and indirect (ID) PL lines are shown. At zero magnetic field, the direct PL line located at 2.8 eV and caused by exciton recombination within the $Zn_{0.97}Mn_{0.03}Se$ layers is unpolarized. The indirect PL band results from electron-hole indirect radiative recombination at type-II heterointerfaces characterized by a very anisotropic oscillator strength. We distinguish two lines differing in the degree of linear polarization (see Fig. 1b), namely the B line located at 1.837 eV and the A line at 1.807 eV (two weak lines at 1.777 eV and 1.747 eV are LO-phonon replicas of the A line, the LO phonon energy in ZnSe is 31 meV). For the B line, the linear polarization degree, defined as $P_l = (I_{1\bar{1}0} - I_{110})/(I_{1\bar{1}0} + I_{110})$ with $I_{1\bar{1}0}, I_{110}$ being the intensities of PL components polarized along the corresponding directions, is equal to 0.72 (i.e., 72%). Within the limits 0.70–0.80 it is associated with a single type-II heterointerface [3,12]. Therefore, we assign this line to one particular kind of interface, either normal or inverted. The A line, with $P_l = 0.4$, is most likely caused by both interfaces.

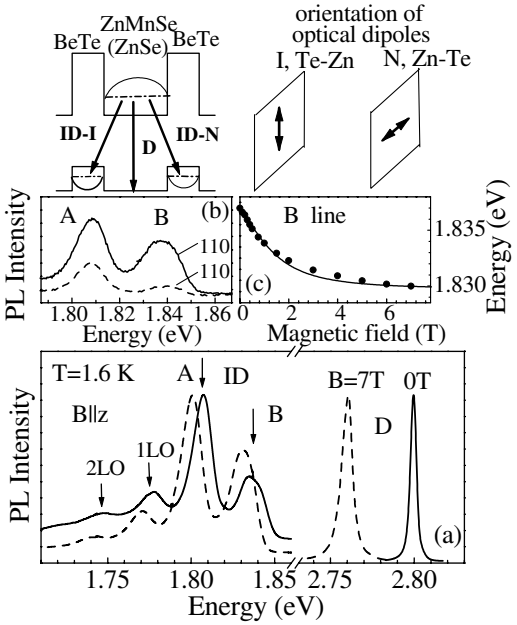

FIG. 1. A 200 Å/100 Å $Zn_{0.97}Mn_{0.03}Se/BeTe$ MQW. (a) PL spectra for direct (D) and indirect (ID) recombination at zero magnetic field (solid line) and $B = 7$ T (dashed, σ^+ polarization). (b) Linearly polarized PL along the $[1\bar{1}0]$ and [110] crystallographic axes for indirect PL band at $B = 0$ T. (c) Energy shift of B line in magnetic fields. Line is a fit with $S_{\text{eff}} = -1.8$, $T_0 = 1.2$ K [11].

In an external magnetic field, the Mn ions orient their spins along the field direction. The strong *sp-d* exchange interaction causes a giant Zeeman splitting of the conduction and valence band states in semimagnetic layers of $Zn_{0.97}Mn_{0.03}Se$. The large shift of 43 meV at $B = 7$ T shown in Fig. 1a for the σ^+-polarized direct PL line is composed of valence- and conduction-band contributions. The field-induced shift of the indirect B-line shown in Fig. 1c is only 7 meV at $B = 7$ T, since it results from the giant Zeeman effect for electrons in $Zn_{0.97}Mn_{0.03}Se$ only (holes are located in the nonmagnetic BeTe). More details concerning the semimagnetic properties of novel (Zn,Mn)Se/BeTe MQW's can be found in Ref. [11].

The direct PL band shows complete circular polarization $P_c = (I_+ - I_-)/(I_+ + I_-) = 0.98$ in magnetic fields above 0.5 T, see triangles in Fig. 2b (here I_+, I_- are the intensities of circularly polarized PL components). The polarization properties of the indirect band differ qualitatively. A fast increase of $P_c(B)$, which is similar to that for the direct PL, is followed by saturation at 0.65 (Figs. 2a and 2b, open circles). We stress here that an incomplete circular polarization for the recombination of spin polarized carriers (at least the photoelectrons are totally polarized) is very unusual, especially for semimagnetic materials, and requires an explanation. The linear polarization degree is insensitive to a magnetic field and keeps its value of 0.72.

The indirect radiative recombination at a type-II (001) interface between two zinc-blende-lattice semiconductors BeTe and ZnSe can be modeled by light-emitting two-dimensional dipole oscillators. This model is quite general and allows description of the polarized luminescence from systems of the C_{2v} symmetry (QW's, superlattices, quantum dots, and even anisotropic centers in solids). We assume that (a) each oscillator can equally oscillate along the planar axes, say $x' \parallel [1\bar{1}0]$ and $y' \parallel [110]$, and (b) the coupling of x' dipoles with the x'-polarized light differs from the $y' - y'$ coupling by a factor of Λ. The circular oscillations $x' \pm iy'$ effectively describe the electron-hole

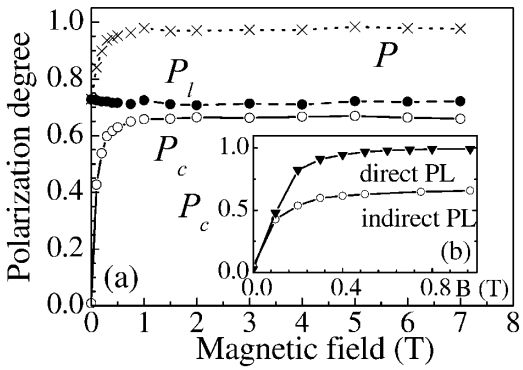

FIG. 2. Effect of magnetic fields on the indirect PL (B line) of a $Zn_{0.97}Mn_{0.03}Se/BeTe$ MQW. (a) Magnetic-field dependencies of the degrees of polarization: linear P_l, circular P_c, and total $P = \sqrt{P_l^2 + P_c^2}$. (b) Field dependence of circular polarization degree for direct and indirect PL. $T = 1.6$ K.

states $|-1/2,3/2\rangle$ and $|1/2,-3/2\rangle$, where $s = \pm 1/2$ and $j = \pm 3/2$ are the electron and heavy-hole spin components along z. For the C_{2v} point symmetry of an ideal (001) interface, two other states $|1/2,3/2\rangle$, $|-1/2,-3/2\rangle$ are optically inactive and can be excluded from the consideration. Then the nonzero matrix elements for emission of a photon can be written as

$$\begin{aligned} M_{1/2,-3/2}(\mathbf{e}) &= M_0(e^*_{x'} - i\Lambda e^*_{y'}), \\ M_{-1/2,3/2}(\mathbf{e}) &= M_0(e^*_{x'} + i\Lambda e^*_{y'}), \end{aligned} \tag{1}$$

where M_0 is a constant and $\mathbf{e}$ is the photon polarization unit vector. In fact, the model uses only one (real) parameter Λ which is the ratio, $p_{y'}/p_{x'}$, of the interband matrix elements of the momentum operator. This parameter can be calculated by applying a straightforward microscopic theory, e.g., the tight-binding theory outlined and developed in [3,8,13], or/and found from comparison with experiment. According to Eq. (1), a photon emitted by the oscillation $x' + iy'$ or $x' - iy'$ is elliptically polarized with the principal axes of the ellipse parallel to x', y' and the degrees of linear and circular polarization are given by

$$\begin{aligned} P_l^0 &= \frac{|M(\mathbf{e} \parallel x')|^2 - |M(\mathbf{e} \parallel y')|^2}{|M(\mathbf{e} \parallel x')|^2 + |M(\mathbf{e} \parallel y')|^2} = \frac{1 - \Lambda^2}{1 + \Lambda^2}, \\ P_c^0 &= \frac{|M(\sigma^+)|^2 - |M(\sigma^-)|^2}{|M(\sigma^+)|^2 + |M(\sigma^-)|^2} = \pm\frac{2\Lambda}{1 + \Lambda^2}, \end{aligned} \tag{2}$$

Here the sign $\pm$ coincides with the circularity of the oscillation. For *each oscillation* the emitted photon is *completely elliptically polarized*, i.e., the total degree of polarization P satisfies the condition

$$P^2 = (P_l^0)^2 + (P_c^0)^2 = 1. \tag{3}$$

At zero magnetic field, the states $|-1/2,3/2\rangle$ and $|1/2,-3/2\rangle$ are equally populated. Because of that the emission is linearly polarized with $P_l = P_l^0$, but lacks circular polarization, $P_c = 0$. In the presence of a longitudinal magnetic field the electron and hole spin states are split and, therefore, thermally populated. As a result, the photoluminescence acquires circular polarization

$$P_c(B) = \frac{\rho_h - \rho_e}{1 - \rho_e\rho_h}\sqrt{1 - (P_l^0)^2}, \tag{4}$$

where $\rho_e(B)$ and $\rho_h(B)$ are the degrees of electron and hole spin polarization. If at least for one kind of photocarriers the condition of equilibrium spin polarization $\tau_R \gg \tau_S$ (τ_R and τ_S are the lifetime and spin relaxation time), is satisfied [14], then at magnetic field where P_c is saturated these carriers are completely polarized (i.e., $|\rho_{e(h)}| \to 1$), the ratio $(\rho_h - \rho_e)/(1 - \rho_e\rho_h) = \pm 1$ and the circular polarization under saturation amounts to

$$P_c^{\text{sat}} = \pm\sqrt{1 - (P_l^0)^2} \equiv P_c^0. \tag{5}$$

At the same time the linear polarization is not affected by a longitudinal magnetic field, $P_l(B) = P_l^0 = \text{const}$. Thus, in a magnetic field $\mathbf{B} \parallel \mathbf{z}$, the photoluminescence arising from the electron-hole recombination in structures with the in-plane anisotropy becomes elliptically polarized and the degree of polarization $P(B) = \sqrt{P_l^2 + P_c^2(B)}$ tends to 1 with increasing field.

In the case where the PL signal is contributed by an equal number of optical dipoles characterized by the anisotropic parameters $\Lambda_1 = \Lambda$ and $\Lambda_2 = \Lambda^{-1}$, the linear polarization vanishes and the information on the optical anisotropy of the individual state is hidden. It can be, however, uncovered on the base of Eqs. (2)–(5) if a magnetic field dependence of the PL circular polarization is analyzed.

To be more specific we now consider, on the base of the developed approach, the experimental results for indirect radiative recombination at a type-II (001) interface in Zn(Mn)Se/BeTe QW's. For these type-II interfaces with high band offsets the anisotropy is determined by the alignment of the interface chemical bonds. For single interface the polarization P_l^0 reaches values of 0.70–0.80 [3,12]. According to Eq. (3), the expected degree of circular polarization is $P_c^0 = 0.71$–0.60. This expectation coincides well with the experimental value of $P_c^{\text{sat}} = 0.65$ shown in Fig. 2a. It is also confirmed by the total polarization degree approaching complete polarization with the growing magnetic field (crosses and dotted line). Microscopical calculations of anisotropic parameter Λ are given in [13]. In what follows we will concentrate on experimental evaluation of this parameter.

Let us turn to the situation where in QW's both normal and inverted type-II interfaces contribute to the PL at the same energy E. If the contributions to the PL intensity related to both kind of interfaces coincide, the linear polarization vanishes and the information on the optical anisotropy of the individual interface is hidden. We focus the attention here on a structure with identical termination of the interfaces which is the case for the samples studied here. In this case $\Lambda_n = \Lambda_i^{-1}$ and $P_{l,n} = -P_{l,i} \equiv P_l^0$, where the indices n, i refer to the normal and inverted interfaces. Then the degree of linear polarization is given by

$$P_l = \frac{P_{l,n} + \eta(E)P_{l,i}}{1 + \eta(E)} = \frac{1 - \eta(E)}{1 + \eta(E)}P_l^0. \tag{6}$$

Here $\eta(E) = I_i(E)/I_n(E)$ is the PL intensity related to the corresponding interface. As previously, P_l is independent of the B. The degree of circular polarization at saturating magnetic fields is determined by

$$P_c^{\text{sat}} = \pm\frac{\sqrt{1 - P_{l,n}^2} + \eta(E)\sqrt{1 - P_{l,i}^2}}{1 + \eta(E)} = \pm\sqrt{1 - (P_l^0)^2}, \tag{7}$$

which is identical with Eq. (5). Because of the function $\eta(E)$ the linear polarization P_l has a spectral dependence. In contrast, P_c^{sat} is spectrally independent and determined by only one parameter, P_l^0, of an individual interface. By measuring the values of $P_l(E)$ and P_c^{sat} and assuming P_l^0 to be positive, one can find P_l^0 and $\eta(E)$ as

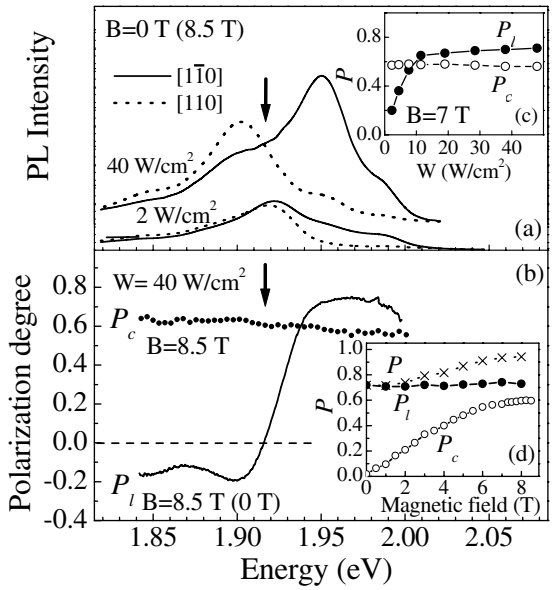

FIG. 3. (a) Linearly polarized PL spectra of a 100 Å/50 Å ZnSe/BeTe MQW. $B = 0$ T (magnetic field of 8.5 T does not modify the spectra). (b) Spectral variation of the PL linear and circular polarizations measured at $B = 8.5$ T. $P_l(E)$ is identical at $B = 0$ and 8.5 T. Insets: Polarization of the indirect PL detected at 1.95 eV vs excitation density (c) and magnetic field at $W = 40$ W/cm^2 (d). Lines are the guide to the eye. $T = 1.6$ K.

$$P_l^0 = \sqrt{1 - (P_c^{\mathrm{sat}})^2}, \qquad \eta(E) = 1 - \frac{2P_l(E)}{P_l(E) + \sqrt{1 - (P_c^{\mathrm{sat}})^2}}. \tag{8}$$

Let us now turn to nonmagnetic ZnSe/BeTe MQW, where under certain conditions the interface anisotropy is completely hidden. This allows us to approve the potential of the suggested method. Indirect emission in this structure is dominated by one line, which is only weakly polarized at low excitation densities $W < 2$ W/cm^2 (Fig. 3a). With increasing W it splits into two polarized components with maxima at 1.950 and 1.902 eV corresponding to the emission at normal and inverted interfaces [3]. The splitting results from the electric field induced by spatially separated carriers [15].

Magnetic field dependencies of P_l, P_c, and P are quite similar to those in magnetic structure (compare Figs. 3d and 2a), except the higher fields are required to orient the carrier spins in the nonmagnetic sample. Namely, P_l is independent of B, P_c approaches its saturation value of 0.60 at $B > 7.5$ T, and P reaches a value of 0.94 at high fields, tending to the complete polarization.

For $W > 10$ W/cm^2 linear polarization degree has a strong spectral dependence (Fig. 3b) varying from -0.20 on the low-energy wing to $+0.72$ on the high-energy wing. Arrows mark the "isotropic" point $E = 1.915$ eV where the anisotropy is completely hidden ($P_l = 0$). One can see from Fig. 3b that, in agreement with Eq. (7), the measured function $P_c^{\mathrm{sat}}(E)$ keeps a constant value of $\approx$0.60 within the whole energy range. It is also insensitive to variation of P_l with decreasing W (Fig. 3c). On the basis of Eq. (8) we conclude that the optical anisotropy of individual interfaces deduced from $P_c^{\mathrm{sat}}(E)$ is indeed giant, insensitive to E, and characterized by $|P_{l,n}| = |P_{l,i}| = 0.8$ in accordance with single-interface data [3]. Respectively, the experimentally determined values $\Lambda_n = \Lambda_i^{-1} = 0.33$.

It is worth mentioning that equations similar to Eqs. (6)–(8) can be used for characterization of a single non ideal type-II heterointerface. Real ZnSe-BeTe interfaces may have monoatomic-scale fluctuations and contain both Zn-Te and Se-Be chemical bonds which lie in the perpendicular (1$\bar{1}$0) and (110) planes. As a result the linear polarization P_l^0 of the photoluminescence will be reduced. By measuring P_c^{sat} one can obtain information about structural and compositional properties of interfaces. Let PL be determined by radiative recombination of localized excitons and let L_{loc} be the exciton localization length. If L_{loc} is comparable with the lateral size of monoatomic roughness r_{ma} then P_c^{sat} should be close to $\pm\sqrt{1 - (P_l^0)^2}$, where P_l^0 describes optical anisotropy of an ideal interface. On the other hand, if L_{loc} exceeds r_{ma} and averages interface roughness, a value of $|P_c^{\mathrm{sat}}|$ will exceed $\sqrt{1 - (P_l^0)^2}$ and tend to unity. This basis allows the development of quantitative approach to interface characterization.

To conclude, the in-plane anisotropy of interfaces hidden in QW structures can be revealed by polarized spectroscopy of spin-oriented carriers. The developed method is not limited to the interface studies and can be readily extended for the wide class of the physical systems consisting of optically anisotropic subsystems—e.g., to study the in-plane anisotropy of quantum dots even in the case of large ensembles of dots characterized by an arbitrary orientation of the optical dipole moments.

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