Symmetry properties of n-doped (Cd,Mn)Te quantum well photoluminescence spectra: An exemplary evidence for anisotropy-induced valence-band mixing

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Symmetry properties of *n*-doped (Cd,Mn)Te quantum well photoluminescence spectra: An exemplary evidence for anisotropy-induced valence-band mixing

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The symmetry properties of photoluminescence spectra of an *n*-doped (001)-$Cd_{0.99}Mn_{0.01}Te$ quantum well provide a simple and intuitive demonstration of the valence-band mixing induced by spontaneous in-plane anisotropy of the sample. The spectra were taken with linear-polarization resolution in an in-plane external magnetic field. They consisted of several features which are characteristic for a two-dimensional electron gas. Because the quantum well layer contained "magnetic" ions of Mn, the spectra showed pronounced polarization-dependent modifications induced by the magnetic field. With the field applied, a 90° rotation of the sample about the surface normal axis resulted in a clearly different spectrum, meaning that the nominally equivalent [110] and $[1\bar{1}0]$ in-plane directions in the sample are not equivalent in fact. But, remarkably, the additional 90° rotations of both the polarizer and the analyzer restored the initial spectrum. This combined invariance regarding simultaneous 90° rotation of the sample and reversal of the polarization configuration was known earlier for spin-flip Raman spectra only. Our present observations are interpreted in terms of an intermixing of valence subbands leading to a pseudoisotropic *g*-factor of the ground-state holes. *Published by AIP Publishing.*
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Since the early 1990s, optical spectroscopy began to reveal the fact that epitaxial quantum well (QW)-and quantum dot (QD) heterostructures of diamond-like semiconductors frequently possess lower symmetry than it follows from simple geometric considerations involving ideal lattices.[1,2] As one of the principal consequences, there arises an intermixing of heavy- and light-hole subbands of the complex valence band Γ_{15}.[3,4] In particular, it results in the degeneracy removal of the doublet of "bright" exciton states $|\pm 1\rangle$, which was confirmed by direct measurements of the optical spectra at a single-particle level,[5,6] in addition to numerous indirect experimental evidences. Apart from the electron–hole interaction within the exciton, the heavy-light-hole mixing manifests itself in the interaction of the holes with an external magnetic field *B*. This latter consequence shows up even in the absence of the fine structure of the spectrum, e.g., in case of the optical emission from trion states.[7]

In an ideal structure grown along the [001] direction, the *B*-linear (Zeeman) spin splitting of the hole states should be practically absent when the field direction is in-plane; in reality, however, such a splitting shows up in quantum wells (QWs),[8,9] in single quantum dots (QDs) belonging to various heterostructure families (e.g., in CdSe,[7,10,11] CdTe,[12,13] InGaAs,[14] InAs,[15,16] and GaAs.[17]) and in QD ensembles.[18,19] This splitting, as well as specific features of the relevant optical transitions involving the split levels, originates from the anisotropic mixing perturbation in the lateral plane. The role and the nature of such perturbation have been analyzed in detail at a phenomenological and at a microscopic level,[9,13,20–22] and also experimentally. For example, the authors of Ref. 13 came to the conclusion that for CdSe quantum dots, the main source of the anisotropy comes from the in-plane strain through the Bir-Pikus Hamiltonian. Contrary to that, the authors of Refs.17 and 23 concluded a dominating role of the QD shape. Whatever the case, the symmetry of the light-emitting state (ideally, D_{2d}) turns out to be reduced down to C_2 or C_{2v} at least, leading to the similar effect on optical transitions.

The physics of valence-band mixing and pseudoisotropic *g*-factor of holes is widely recognized in the field of the optics of single QDs, where optical transitions involving separate spin sublevels can be resolved in the spectral domain and analyzed individually. For QWs, where the distinct transitions are buried within the broadened spectral lines, only indirect experimental evidences for the same physical mechanism were obtained (e.g., rather delicate measurements of the linear polarization degree of the photoluminescence (PL)).[8,9] However, a favorable model system can allow direct access to the impact of the mixing on the PL spectra. The details of this impact can convincingly demonstrate the symmetry of the optical transitions driven by the mixing, and can show that this physics applies to quite ordinary (regarding symmetry and technology) QWs.

In the present paper, we give an example of such a favorable model system. We describe the transformation properties of the PL spectra of a (Cd,Mn)Te QW subject to in-plane magnetic fields. We show how the polarization configuration of the PL experiment in combination with the orientation of the in-plane field **B** results in a particular shape of the PL spectrum. At moderate *B*-field values ($B \leq 1.5$ T),

the spectra demonstrate the property of *combined invariance* (CI), i.e., invariance regarding simultaneous 90° in-plane rotation of the sample and reversal of the polarization configuration. The CI was observed earlier for the spectra of spin-flip Raman scattering (SFRS) in undoped QWs[24] but was never reported for PL spectra. At higher values of B, the CI becomes violated. We discuss possible reasons for that.

The sample was a single 12 nm (001)-(Cd,Mn)Te type-I QW sandwiched between two $Cd_{0.85}Mg_{0.15}Te$ barrier layers. The content of the magnetic Mn^{2+} ions in the QW layer was 0.79% of the cation sites. The QW contained a confined two-dimensional electron gas with a reported[25] concentration $n_e = 2.1 \times 10^{11}$ cm^{-2}, induced by modulation doping from the barrier layers by iodine from a ZnI_2 source. Further details and previous experimental data regarding our sample can be found in Ref. 26 (sample #405 A).

The PL spectra were taken in backward geometry, always in crossed linear polarization directions of the polarizer for the incident laser light and the analyzer for the photoluminescence radiation. The PL was dispersed in a Dilor XY triple spectrometer and recorded using a CCD detector. Excitation came from a dye laser and, for the spectra presented here, was tuned to the energy ~30 meV above the QW PL. The sample was immersed in liquid helium at $T = 1.5$ K. The magnetic field B up to 4.5 T was induced by a split-coil horizontal magnet and was perpendicular to the optical axis of the experiment (Voigt configuration).

Various configurations of optical polarization and B-field orientation with respect to the in-plane crystal axis directions were achieved by rotations of the sample about the surface normal axis. These rotations were performed through re-positioning on the sample holder after warming up to room temperature.

The PL spectra of QWs with a 2D electron gas at this (intermediate) level of electron concentrations typically include several spectral features.[25,26] These features, or contours, are close in energy and broadened, so that they noticeably overlap. In addition, the QW layer in our sample is formed by a diluted magnetic semiconductor (DMS), a material characterized by a giant spin-dependent modification of the band structure in an external magnetic field.[27] The combination of these two factors makes the shape of the PL spectra both field- and polarization-dependent and rather individual.[28]

Fig. 1 shows the B-field evolution of the PL collected in two opposite linear polarizations (panels (a) and (b), respectively). Here and below, the experimental configurations are specified by a bracketed two-symbol notation as follows. The first symbol specifies the orientation of the [110] in-plane axis of the sample against the applied magnetic field. This orientation was either parallel (//) or perpendicular (⊥) to the B-field direction. The second symbol specifies the orientation of the analyzer[29] for the polarization direction of the PL radiation which was also chosen either parallel (//) or perpendicular (⊥) to the B-field. All in all, four physically nonequivalent configurations' results are as follows: (//, //), (//, ⊥), (⊥, //), (⊥, ⊥). For one of them, the relative orientations of B-field, sample [110]-axis, and PL polarization direction are sketched in the inset in Fig. 1(b), and for two of them in Fig. 3.

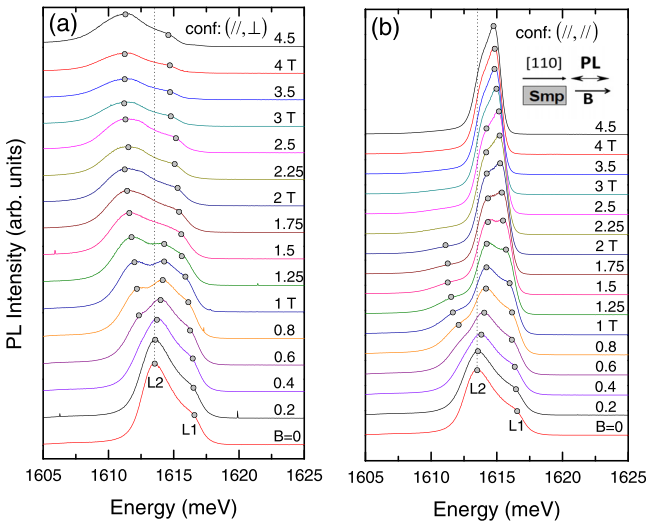

FIG. 1. Photoluminescence spectra of the n-type (001)-$Cd_{0.99}Mn_{0.01}$ QW taken at different values of the in-plane magnetic field **B**, directed along the [110] axis (specified at the curves), and at two different orientations of the PL analyzer: perpendicular to (a) and along (b) the magnetic field direction, as shown exemplarily by the inset in panel (b). The main features forming the spectra are marked by grey circles.

The PL spectra in both configurations presented in Fig. 1 are mainly formed by three features. The L1 feature is shifted down in energy as the field is applied, thus it is naturally perceived as a lower (downshifting) Zeeman branch of some optical transition. The two branches of the L2 feature represent the upper and the lower Zeeman partners of another transition,[26] and they can be traced back to a single line at $B = 0$ (Fig. 1). As shown in Ref. 26, the energetic separation of the split branches of the L2 line as a function of B follows the spin splitting of the conduction-band electron.

The interpretation of PL spectra like those in Fig. 1, either in terms of the Fermi Sea or in terms of multi-particle states, has been discussed in great detail elsewhere.[25,26,30] According to Ref. 26, the L1 line and the two L2 branches can be explained consistently by transitions, starting from a four-particle state called Suris tetron, which consists of a negatively charged exciton and a hole in the Fermi sea (Fermi hole, FH). The L1-transition leads to the ground state of the Fermi sea, while the L2-branches have as final state an excited electron–FH pair in a triplet state, which causes the B-induced splitting.[26] Alternative interpretations operate in terms of screened Coulomb interactions[25] or other versions of the multi-particle approach.[30] Note, however, that the detailed interpretation of the PL spectrum has no further relevance to our present subject. Our present findings rely on symmetry considerations and concern the valence-band ground state which equally well manifests itself in any type of the emitting band-edge transitions, no matter what arrangements of the conduction-band states in the initial and final states are assumed. This makes our conclusions independent of a particular model of the band-edge emission of the QW under study.

Here, we just note that in Fig. 1, the spectra recorded in opposite polarizations are obviously different. This is very clearly visible in Fig. 2, which shows PL spectra taken at a constant B-field with four different orientation configurations: the use of opposite orientation of the analyzer leads to

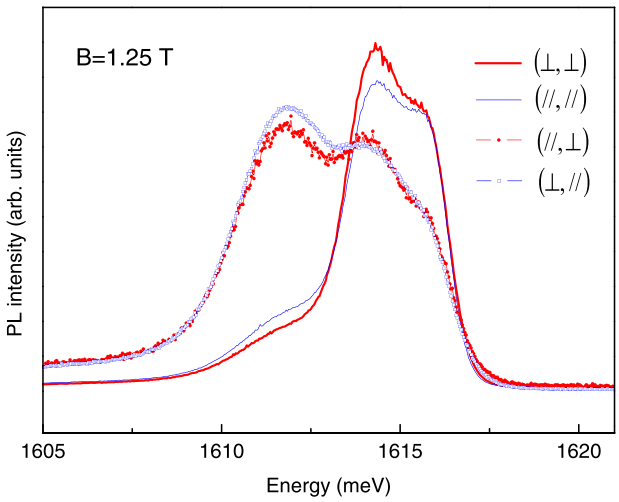

FIG. 2. The CI property demonstrated by the PL spectra taken at $B = 1.25$ T with four different orientation configurations of the in-plane [110] axis and the PL analyzer with respect to the B-field. Those spectra which are differing only by the sample-axis orientation (within the pairs $(\perp,\perp)$, $(//,\perp)$, and $(\perp,//)$, $(//,//)$) are clearly different, proving the in-plane anisotropy of the sample. Those spectra which are differing only in the orientation of the analyzer (pairs $(\perp,\perp)$, $(\perp,//)$ and $(//,\perp)$, $(//,//)$) are different too. However, combined rotations of the sample and the polarization analyzer result in remarkably similar spectra (pairs $(\perp,\perp)$, $(//,//)$, and $(//,\perp)$, $(\perp,//)$).

a clearly different spectral shape. Moreover, Fig. 2 shows that a sole 90° rotation of the sample leads to a strikingly different spectrum as well. The latter fact confirms the in-plane anisotropy of the sample and non-equivalence of its (nominally equivalent) [110] and [1$\bar{1}$0] directions.

Remarkably, if one combines the 90° rotation of the sample, i.e., the reversal of the [110] and [1$\bar{1}$0] axes, with the reversal of the laser polarization and detected polarization (i.e., performs a *combined reversal* operation, CR), the resulting PL spectrum turns out to be very similar to the initial one: see the pairwise coincidence of spectra in Fig. 2. This is the CI property.[24]

As established in Ref. 24, for the case of spin-flip Raman spectra, the CI originates from the extremely anisotropic (*pseudoisotropic*[7,9]) spin structure of the valence-band ground state. Summarized, the g-factor of the ground state holes is induced by an in-plane uniaxial perturbation (e.g., deformation), which mixes the heavy-hole and the light-hole subbands. The physics of the pseudoisotropic in-plain g-factor is well understood.[7–11,13,14,20–22] In brief, the in-plane g-factor of the pure heavy hole state is a 2×2 block of zeroes. An in-plane perturbation admixes the light-hole states and induces a non-zero g-factor, whose symmetry fully reproduces that of the perturbation. As a result, the Zeeman splitting of the mixed hole states is linear in B and does not depend on the orientation of the field, but the polarization selection rules are different as compared to the case of the isotropic g-factor. The (linear) polarization of every single optical transition between the Zeeman-split states follows the orientation of the crystal rather than that of the magnetic field. In particular, these selection rules result in the CI.

The QW sample studied here presents a convenient model system for demonstration of the CI: The spectra are individual enough and undergo pronounced polarization-dependent changes in the magnetic field. This has allowed the demonstration of the CI by means of PL spectra. In addition, no examples of the pseudoisotropic behavior of the valence-band g-factor were presented earlier for QWs containing electron gases. Our results show that this explanation applies to doped QWs too.

The CI persists as long as the electron spin splitting is controlled by the isotropic g-factor, while the hole spin splitting is governed by the pseudoisotropic in-plane g-factor (both being linear in the applied field B). In stronger magnetic fields, the CI may become violated because the pseudoisotropic B-linear term eventually gives way to a superlinear (and truly isotropic) contribution to the hole spin splitting. Fig. 3 shows the B-field dependence of the L1 and the upper L2 features. One can see that for the two experimental configurations $(\perp,\perp)$ and $(//,//)$, which are related to each other by the CR operation, the PL features reveal similar behavior in weak fields but deviate from each other in the fields above $B \approx 1.5$ T, where apparently the $\propto B^3$ contribution to the hole spin splitting[31–33] becomes comparable to the linear term.

Although the existence of both the $\propto B$ (pseudoisotropic) and the $\propto B^3$(isotropic) contribution in the hole spin was established very well in previous studies, we avoid to present any theoretical curve in Fig. 3, since such a calculation would require a specific weighting of the linear and superlinear contributions, which would give no deeper insight into the universal character of the observed phenomenon. Obviously, the increase of the B value will result, within any reasonable model, in a crossover from the anisotropic behavior (the polarization direction is bound to the crystal axes) to the truly isotropic behavior (the polarization direction is bound to the magnetic field direction), just as observed in our experiments.

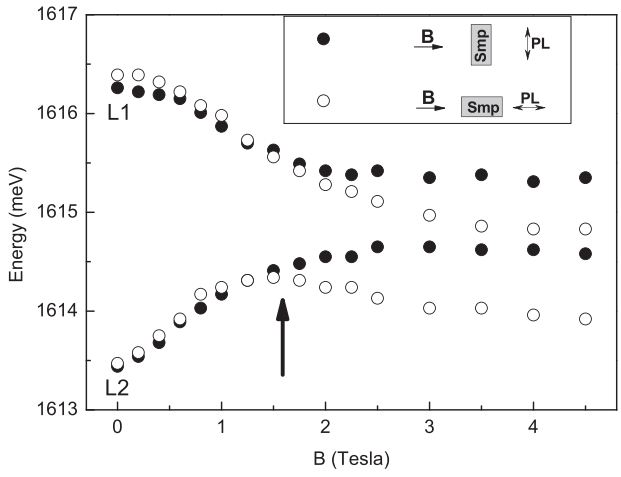

FIG. 3. Violation of the CI at strong magnetic fields. The circles show the peak positions of the lower Zeeman branch of the L1 line and of the upper Zeeman branch of the L2 line (see Fig. 1) versus the magnetic field strength. (The lower L2 branch is left out because its intensity vanishes for $B > 2$ T, see Fig. 1(b).) Open and closed circles correspond to two different experimental configurations which are related to each other by the CR operation. The peak positions coincide at weak fields (in compliance with the CI) but deviate above ~1.5 T, where the B-nonlinear contributions may get control over the hole spin splitting. Inset: experimental configurations as labeled by a two-symbol notation and further detailed by more intuitive pictograms of the elements layout.

Finally, a remark on the expected generality of our findings is appropriate. While the used QW was especially convenient for demonstration purposes, we do not see any reason why specifically (Cd,Mn)Te, or specifically DMS-based QWs, should show the valence-band mixing and relevant polarization selection rules. It is worth recalling that in the QD optics, the similar mechanism was found efficient for numerous material families and, equally well, for QDs containing many magnetic ions,[11,34] a single magnetic ion,[12,35] or no magnetic ions[36,37] inside. Similar to that, we expect the present mechanism to be relevant for other magnetic and non-magnetic QWs, while in the latter, it may not be easily proved by so straightforward experimental observations.

By means of the PL spectra of a DMS quantum well doped by electrons, we demonstrated the characteristic behavior of polarization selection rules for optical transitions. Typically for epitaxial QWs grown of cubic semiconductors along the [001] direction, the true symmetry turns out to be lower than the nominal symmetry (D_{2d}), with [110] and $[1\bar{1}0]$ directions being non-equivalent to each other. This leads to a sensitivity of the PL spectra regarding the respective orientation of the sample and the magnetic field. Remarkably, the CI behavior observed in this sample below $\sim$1.5 T confirms that the hole spin splitting (which cannot be spectrally resolved) is linear in the magnetic field B and originates from an in-plane distortion of the QW layer, rather than by properties of an ideal QW. As the field was further increased, a violation of the CI showed up, manifesting the onset of the superlinear contributions to the spin splitting of hole states.

The conclusions of the present report do not rely on any microscopic interpretation of the PL spectra of the system under study. They were derived on the basis of simple symmetry operations over the sample and the polarization optics. The quite distinctive PL spectrum of our system and its pronounced polarization-dependent behavior in the external magnetic field allowed the observation of the CI behavior in the photoluminescence, and also for a quantum well system containing a 2D electron gas.

This mechanism is expected to apply also for other magnetic and non-magnetic QWs, even though not always experimentally unambiguously provable. Still, if one evaluates a Zeeman splitting of a hole ground state in a real epitaxial QW with a similar band structure, the pseudoisotropic contribution induced by in-plane imperfections should never be ignored *a priori*.

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