Strong linear polarization induced by a longitudinal magnetic field in II-VI semimagnetic semiconductor layers

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(Cd,Mn)Te-based semimagnetic epilayers are studied by polarized reflection spectroscopy under a longitudinal magnetic field. We observe a huge *linear* polarization induced by the magnetic field: the degree of linear polarization reaches almost 100% when the heavy hole and light hole bands are brought to a resonance, driven by the giant Zeeman effect. A clear anticrossing between light and heavy hole exciton lines is also observed at the resonance. These features are characteristic of a light-heavy hole mixing by an anisotropic potential with C_{2v} symmetry. We suggest that the latter is due to in-plane uniaxial strain associated with anisotropic relaxation of the lattice mismatch.

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I. INTRODUCTION

III-V and II-VI semiconductor structures grown epitaxially on (001) oriented substrates often exhibit in-plane optical polarization anisotropy, which is not expected from the symmetry of bulk constituents. Several mechanisms have been discussed, such as surface-reconstruction and related effect,[1] alloy ordering,[2] anisotropy of interface bond configuration,[3] or the presence of oriented defects generated during the growth process.[4] Another possibility is represented by in-plane uniaxial strain. The latter can be of particular importance in the case of self-assembled quantum dot systems, where "intrinsic" strain anisotropy can result from dot shape anisotropy. In the case of bulklike epitaxial layers, "extrinsic" in-plane uniaxial strain can result from partial, anisotropic plastic relaxation of the lattice mismatch.

Except for the effects arising on the deep level defects, anisotropy is due to a mixing of the heavy and light holes at the Brillouin zone center, and the effect is simply governed by some coupling matrix element and the energy separation between heavy and light holes.[4] In this work we examine in-plane anisotropy of strained, bulklike CdZnMnTe semimagnetic epilayers where the giant Zeeman effect allows a convenient tuning of the hole band splitting.

II. SAMPLES

We have examined two samples. Sample S1 contains an asymmetric, 20 monolayer- (ml) thick CdMnTe quantum well (QW) embedded between CdMgMnTe and CdZnMnTe thick barriers. This heterostructure is grown on a thick CdTe buffer layer deposited on a GaAs substrate. Sample S2 has the same structure, but without the quantum well. The sample structure is shown in Fig. 1. Using published lattice constants of CdTe,[5] MnTe,[6] MgTe,[7] and ZnTe[8] and assuming Vegard's law and perfectly pseudomorphical growth we calculated the lattice mismatch values given in Fig. 1. They correspond to positive (tensile) biaxial strain.

Nominal values are given for the sample parameters, in particular significantly different Zn mole fractions in the outer barrier for the two samples. However, from post-growth characterization (see Fig. 2 and discussion below), it is clear that real compositions of the two barriers are very close. It can be safely assumed that a several μm-thick CdTe buffer is fully relaxed with respect to the GaAs substrate, but the significant lattice parameter difference between the well, barriers, and CdTe buffer can give rise to partial plastic relaxation within the epilayer when individual layer thicknesses exceed a critical thickness. The lattice mismatch between the first (inner) barrier and CdTe is 0.2% only, which corresponds to a large critical thickness for the onset of plastic relaxation (nucleation of first misfit dislocation). Therefore we naturally assume that this inner barrier is biaxially strained to accommodate to the CdTe buffer, in both samples. Biaxial strain is also obviously the case for the narrow CdMnTe well (lattice mismatch of 0.1%). Conversely, the lattice mismatch of the second (outer) barrier made of (Cd,Zn,Mn)Te with at least 10% of Zn is much

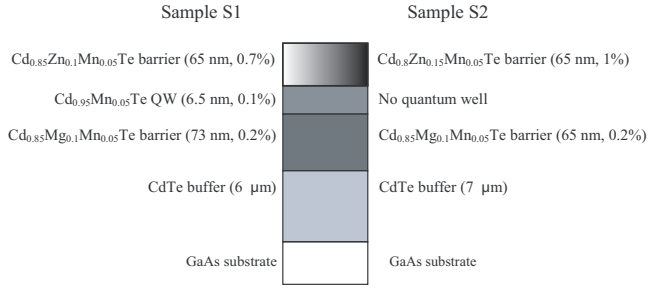

FIG. 1. (Color online) Structure of the samples. Nominal values of thickness and composition are given. Lattice mismatch values relative to CdTe are given in %.

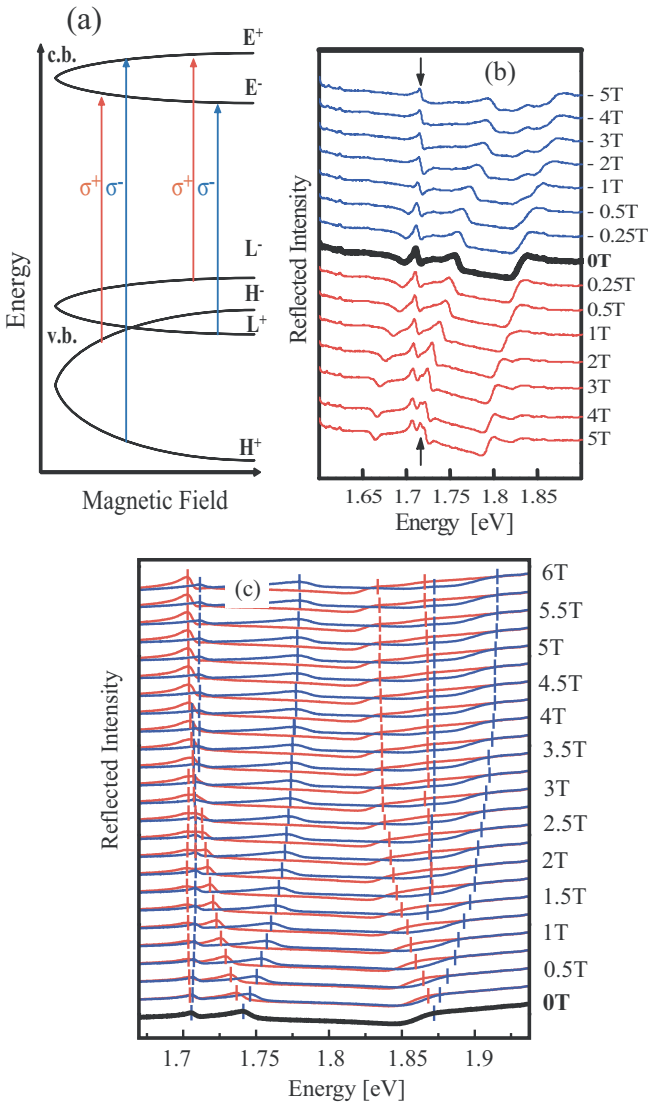

FIG. 2. (Color online) Scheme of the energy levels and optical transitions in the strained semimagnetic layer (a). Reflectivity spectra measured at 1.6 K in magnetic field in $\sigma+$ and $\sigma-$ circular polarizations for samples S1 (b) and S2 (c). The sign of the magnetic field used in (b) fixes the circular polarization.

larger (nominal values of 0.7% and 1.0% for S1 and S2, respectively). The outer barrier thickness is comparable with the critical thickness[9] and a partial plastic relaxation is possible.

III. IDENTIFICATION OF OPTICAL TRANSITIONS

Figure 2 shows reflectivity spectra of samples S1 and S2 in a longitudinal magnetic field, measured for both circular polarizations. Note that in these experiments, changing the circular polarization at a fixed field direction is equivalent to changing the field direction at a fixed circular polarization. It is clear from the relative thicknesses and related absorption strengths that bulk features associated with the barriers must dominate the reflectivity spectrum in the corresponding energy gap range. Due to the presence of the manganese in the samples, it is easy to distinguish between light and heavy hole transitions. Both exhibit giant Zeeman effect, but the heavy hole exciton splitting is more than 4 times larger than that of the light hole exciton. Besides, a light hole transition intensity is about three times smaller than a heavy hole one of the same origin. We use the standard notation L±, H±, and E± for the corresponding valence and conduction levels. The scheme of energy levels and allowed optical transitions (ignoring possible anticrossings) is shown in Fig. 2(a).

Three transitions are well visible at zero field (bold traces) for both samples: a light hole transition at about 1.7 eV and two heavy hole ones at about 1.75 eV and 1.85 eV, respectively. These energy values allow us to assign the transitions to three-dimensional (3D) excitons in the (Cd,Zn,Mn)Te barrier (1.7 eV and 1.75 eV) and in the (Cd,Mg,Mn)Te barrier (1.85 eV), respectively. As mentioned before, close values of the (Cd,Zn,Mn)Te barrier transitions in both samples indicate close values of the barrier compositions. Combining known energy gap composition dependence of (Cd,Mn)Te[6] and (Cd,Zn)Te[10] ternary alloys we estimate that the measured transition energy of 1.75 eV corresponds to a common value of zinc mole fraction in the outer barrier of both samples of about 13%.

Application of the magnetic field reveals two additional transitions: a light hole and a heavy hole one, masked at zero field by the transitions mentioned previously. We assign them to a light hole exciton in the (Cd,Mg,Mn)Te barrier (visible in both samples) and to a heavy hole transition in the quantum well (in sample S1 only). Different barriers make the quantum well asymmetric and very shallow. Due to the strain in the (Cd,Zn,Mn)Te barrier, the light hole potential does not provide any confinement in the (Cd,Mn)Te quantum well. Therefore no light hole transition is observed in the quantum well, whereas the QW heavy hole transition is very close in energy to the light hole transition in the (Cd,Zn,Mn)Te barrier. The light-heavy hole transition separation provides information on the strain present in the barriers. This separation is negligible for the (Cd,Mg,Mn)Te barrier, confirming the estimated small strain value. In the (Cd,Zn,Mn)Te barrier the light-heavy hole separation $\delta E_{L-H}=-40$ meV in both samples, with the light hole transition at a lower energy, as expected for a tensile strain. Knowing deformation potential b (−1.15 eV for CdTe[11]) and elastic constants[12] $S_{11}=0.417$ and $S_{12}=-0.172$ [$\times 10^{-10}$ m^2/N], we estimate the biaxial strain of the barrier as $\delta E_{L-H}=2be(S_{11}-S_{12})/(S_{11}+S_{12})$ where e denotes biaxial strain of the layer. We obtain $e=0.007$, corresponding to a Zn composition of 12%. This value of e is in close agreement with the x-ray diffraction measurement of sample S2 that gives $e_z=2eS_{12}/(S_{11}+S_{12})=-1\%$.

IV. ANTICROSSING BEHAVIOR

We can see from Fig. 2 that at a sufficient magnetic field the lower component of the heavy hole transition in the (Cd,Zn,Mn)Te barrier (H_-E_-) is brought to a resonance with the L_+-E_- transition from the same barrier. An anticrossing occurs at the resonance, resulting in mixing of the

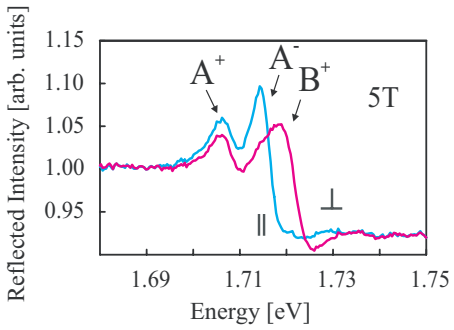

FIG. 3. (Color online) Reflectivity spectra of sample S1 at 1.6 K and 5 T in the anticrossing region, measured in two perpendicular linear polarizations. A^+, A^-, and B^- denote optical transitions discussed in the text. ∥ and ⊥ refer to the ⟨110⟩ orthogonal directions.

wave function, witnessed by a relaxation of polarization selection rules. This resonance is observed at about 5 T in Sample S1 and near 2.5 T in Sample S2. For example, the transition pointed with arrows in Fig. 2 is well visible in both polarizations at $|B|=5$ T. It is interesting to note here that a magnetic field applied in Faraday configuration splits usually optical spectra in circularly polarized components [see Fig. 2(a)]. Here, as we show in the following it produces a strong enhancement of the *linear* polarization. The paradox results from the resonance between the light and heavy hole bands due to giant Zeeman effect in the diluted magnetic semiconductor. At zero field, any perturbation of C_{2v} symmetry (its physical nature will be discussed later) introduces an optical anisotropy by mixing of light and heavy hole states, H_- and L_+ (and respectively H_+ and L_-). This mixing, and therefore the linear polarization rate, is controlled by a matrix element of the C_{2v} potential and the energy separation of light and heavy holes, which in turn can be controlled by the magnetic field. The anisotropy, being small at zero field, becomes giant when the light and heavy hole bands are brought to a resonance and an anticrossing appears.

In Fig. 3, we switch to linear polarization parallel to ⟨110⟩ directions and examine a blowup of the anticrossing spectral range in sample S1. The line denoted A^+ in Fig. 3 corresponds to the $L_- - E_+$ transition, which has a pure circular σ^+ polarization, visible in Fig. 2. Accordingly, in Fig. 3 it is equally intense in both linear polarizations. Besides the line A^+, two lines labeled A^- and B^+ are visible in Fig. 3. Line A^- is predominantly linearly polarized in one of the ⟨110⟩ directions, whereas the spectrum in perpendicular polarization shows a completely polarized line B^+, and a trace of line A^-, showing that its polarization is not complete. This asymmetric behavior of both lines can be traced back to the difference of their intensities, expected between the $L_+ - E_-$ (A^-) and $H_- - E_-$ (B^+) transition. This intensity difference is directly visible in Fig. 2 at fields far from resonance. The energetic distance between lines A^- and B^+ (about 5 meV) represents a rough estimate of the anticrossing energy. Strictly speaking, its value would be observed when both lines become polarized symmetrically (orthogonal elliptical polarizations). At 5 T we are slightly off resonance in sample S1. A simple empirical anticrossing model (diagonalization of a 2×2 matrix) of two lines with a 3:1 intensity ratio predicts in our situation an increase of the energy distance by about 15% relative to the anticrossing energy, reducing the estimate to about 4.3 meV.

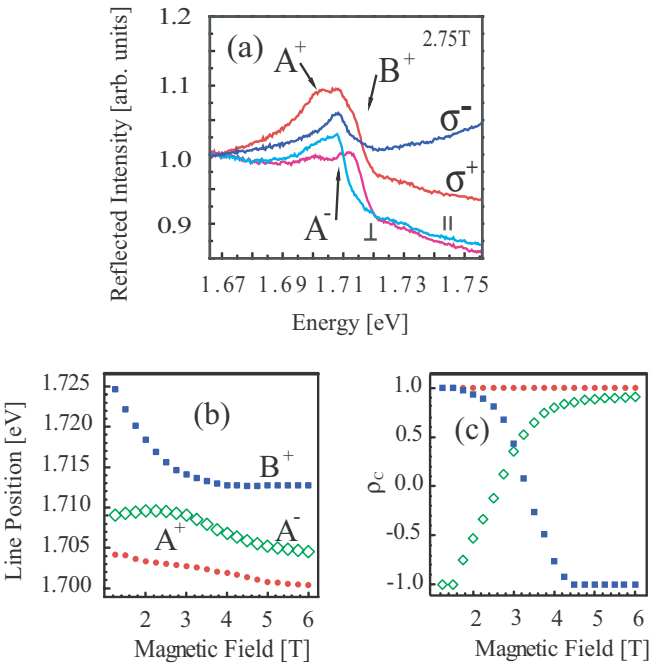

FIG. 4. (Color online) (a) Reflected intensity spectra of sample S2 taken at 1.6 K and field of 2.75 T, measured in circular and linear polarizations; (b) line position, and (c) degree of circular polarization, obtained from Lorentzian fits of the magnetoreflectivity spectra of sample S2, as a function of magnetic field.

More complete data are available for sample S2, where systematic fits of spectral structures were performed. Far from anticrossing, we have found that the line shape can be described by the complex Lorentzian

$$L(E) = t\left(\frac{a_{Im}}{1+t^2(E-E_0)^2} - \frac{a_{Re}t(E-E_0)}{1+t^2(E-E_0)^2}\right),$$

where E is the photon energy, E_0 the transition energy, t describes the line width, and the ratio of the imaginary and real part of the amplitude is $a_{Im}/a_{Re}=1.02$. In the anticrossing region, we combined the various transitions and determined the parameters from simultaneous fits of the spectra for two opposite circular polarizations. The results are illustrated in Fig. 4. The anticrossing behavior of the A^- and B^+ transitions is well visible in energy [Fig. 4(b)], and circular polarization [Fig. 4(c)] of the two lines. Figure 4(b) allows us to read directly the anticrossing energy, equal to 5 meV.

V. DISCUSSION

Our results show that the observed anisotropy is not related to the presence of the quantum well in the sample. It is therefore natural to assume that it has a volume origin. A possible mechanism generating the observed anisotropy is anisotropic relaxation of the mismatch strain. No bulk in-plane anisotropy is expected in case of a perfect, isomorphic (Cd,Zn,Mn) layer on a (001) substrate. The layer would

experience a biaxial in-plane tensile strain equal to the lattice mismatch relative to the CdTe buffer. This strain is accompanied by a compressive strain along the growth axis, resulting from vanishing stress component in that direction. As a result, a splitting between light and heavy hole bands appears, proportional to the lattice mismatch. At a sufficient layer thickness, strain relaxation starts. A (001) interface in a zinc blende structure possesses a C_{2v} symmetry, with inequivalent [110] and [1-10] directions. Therefore an anisotropic strain relaxation[13] can be expected. The extreme case would be, e.g., a complete relaxation in [1–10] direction with no relaxation along [110]. Such a case would correspond to a uniaxial stress along [110] direction, producing a tensile strain equal to the lattice mismatch. It is easy to see from the strain Hamiltonian that in such a situation, the light-heavy hole separation would correspond to the anisotropic splitting, occurring in this case at zero magnetic field. It is clear that we are far from this situation, since the anticrossing energy in our samples (5 meV) is much smaller than the light-heavy hole separation (40 meV). To the first approximation, we can therefore estimate that anisotropic relaxation is an order of magnitude smaller than the misfit strain, a result which is not unlikely. Another possibility to explain the observed anisotropy would be a spontaneous ordering of the (Cd,Zn,Mn)Te alloy. Such ordering along the [111] direction has indeed been reported[14] for ternary (Cd,Zn)Te alloys, and could be another source of the C_{2v} potential mixing light and heavy holes in a layer grown along [001]. We have examined oblique incidence x-ray diffraction and found no measurable evidence of alloy ordering. Also, this structural investigation reveals a small misorientation (0.8° towards [110]) of the CdTe buffer layer with respect to the GaAs substrate [001] axis. This phenomenon, already observed in this system and attributed to the screw component of misfit dislocations at the substrate/buffer interface,[15] implies that the strain field in the CdZnMnTe epilayer has a small in-plane uniaxial component. However, this effect is at least one order of magnitude too small to account for the observed anticrossing. We conclude that partial, anisotropic plastic relaxation (that unfortunately could not be evidenced directly from our x-ray diffraction data) is the most likely source of the observed C_{2v} perturbation.

VI. CONCLUSIONS

We observe a remarkable linear polarization induced by a longitudinal magnetic field in a strained semimagnetic epilayer. The effect is explained in terms of the interplay between a structural perturbation with C_{2v} symmetry and the tuning of the valence band structure with the giant Zeeman effect. We suggest that anisotropic strain relaxation is the most likely source of such structural perturbation. This study illustrates once more how polarization-resolved spectroscopy works as a powerful tool for the measurement of tiny coupling phenomena associated with symmetry-breaking perturbations.

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[1] D. Paget, C. Hogan, V. L. Berkovits, and O. E. Tereshchenko, Phys. Rev. B **67**, 245313(R) (2003); and references therein.
[2] E. Gregery, K. H. Guldeny, P. Riely, H. P. Schweizery, M. Mosery, T. Kippenbergz, G. Schmiedelz, P. Kieselz, and G. H. Döhler, Quantum Semiclassic. Opt. **10**, 271 (1998).
[3] O. Krebs and P. Voisin, Phys. Rev. Lett. **77**, 1829 (1996).
[4] K. Kowalik, A. Kudelski, J. A. Gaj, T. Wojtowicz, O. Krebs, and P. Voisin, Solid State Commun. **126**, 467 (2003).
[5] B. Segall, M. R. Lorenz, and R. E. Halsted, Phys. Rev. **129**, 2471 (1963).
[6] J. K. Furdyna, J. Appl. Phys. **64**, R29 (1988).
[7] J. M. Hartmann, J. Cibert, F. Kany, H. Mariette, M. Charleux, P. Alleysson, R. Langer, and G. Feuillet, J. Appl. Phys. **80**, 6257 (1996).
[8] W. L. Roth, in *Physics and Chemistry of II-V1 Compounds*, edited by M. Aven and J. S. Prener (North Holland Publishing, Amsterdam, 1967).
[9] J. Y. Marzin, J. M. Gerard, P. Voisin, and J. A. Brum, in *Semiconductors and Semimetals*, edited by T. P. Pearsall (Academic Press, Boston, 1990), Vol. 32, Chap. 3, p. 56.
[10] V. A. Tyagai, O. V. Snitko, V. N. Bondarenko, N. I. Vitrikhovskii, V. B. Popov, and A. N. Krasiko, Fiz. Tverd. Tela (S.-Peterburg) **16**, 1373 (1974).
[11] M. Zigone, H. Roux-Buisson, H. Tuffigo, N. Magnea, and H. Mariette, Semicond. Sci. Technol. **6**, 454 (1991).
[12] R. D. Greenough and S. B. Palmer, J. Phys. D **6**, 587 (1973).
[13] K. L. Kavanagh, M. A. Capano, L. W. Hobbs, J. C. Barbour, P. M. J. Maree, W. Schaff, J. W. Mayer, D. Pettit, J. M. Woodall, J. A. Stroscio, and R. M. Feenstra, J. Appl. Phys. **64**, 4843 (1988).
[14] A. Marbeuf, R. Druilhe, R. Triboulet, and G. Patriarche, J. Cryst. Growth **117**, 10 (1992).
[15] G. Patriarche, A. Tromson-Carli, J. P. Rivière, R. Triboulet, Y. Marfaing, and J. Castaing, Phys. Status Solidi A **138**, 437 (1993); A. Tromson-Carli, G. Patriarche, R. Druilhe, A. Lusson, Y. Marfaing, and R. Triboulet, Mater. Sci. Eng., B **16**, 145 (1993).