PHYSICAL REVIEW B VOLUME 56, NUMBER 4 15 JULY 1997-II

Magnetic tuning in excitonic Bragg structures of $(Cd, Mn)Te/(Cd, Zn, Mg)Te$

J. Sadowski*

Laboratoire de Spectrome´trie Physique, Universite´ Joseph Fourier Grenoble 1, CNRS (UMR 55 88), Boıˆte Postale 87, F-38402 Saint Martin d'He`res Cedex, France and Institute of Physics, Polish Academy of Sciences, al. Lotniko´w 32/46, PL-02668 Warszawa, Poland

H. Mariette, A. Wasiela, R. André, and Y. Merle d'Aubigné

Laboratoire de Spectrome´trie Physique, Universite´ Joseph Fourier Grenoble 1, CNRS (UMR 55 88), Boıˆte Postale 87, F-38402 Saint Martin d'He`res Cedex, France

T. Dietl

Laboratoire de Spectrome´trie Physique, Universite´ Joseph Fourier Grenoble 1, CNRS (UMR 55 88), Boıˆte Postale 87, F-38402 Saint Martin d'He`res Cedex, France

and Institute of Physics, Polish Academy of Sciences, al. Lotniko´w 32/46, PL-02668 Warszawa, Poland

(Received 29 April 1997)

We demonstrate that giant spin splitting specific to diluted magnetic semiconductors offers a tool to tune the light wavelength (corresponding to the exciton energy) across the Bragg or anti-Bragg resonances in appropriately designed long-period multiple-quantum-well structures. In particular, an unusually strong influence of the magnetic field upon the shape, magnitude, and spectral position of polariton reflectivity is observed in the case of high-quality periodic structures, in which thick barriers of $Cd_{0.93}Mg_{0.04}Zn_{0.03}Te$ separate 20 thin equidistant quantum wells of nonmagnetic CdTe and magnetic $Cd_{0.96}Mn_{0.04}$ Te placed alternatively. The spectra obtained experimentally are well reproduced by theoretical calculations which incorporate effects of the *spd* exchange interaction into the formalism of transfer matrices. $\left[S0163 \text{--} 1829(97)50928 \text{--} 3 \right]$

Soon after the discovery¹ that the $sp-d$ exchange interaction between the mobile carriers and the localized Mn spins leads to strong magneto-optical effects in diluted magnetic semiconductors (DMS) (Ref. 2) a concept of *spin superlattices* was put forward. This notion, introduced by von Ortenberg³ and explored in more detail by Kossut and Furdyna, 4 has encompassed such phenomena which might result from the effect of the giant spin splitting of electron bands upon the confinement energies of *carriers* in quantum structures of DMS. Among the most important confirmations of those expectations is perhaps the demonstration by means of exciton spectroscopy that the magnetic field can drive the type I to type II transition in II-VI superlattices of suitable constituencies.⁵

It has recently been shown that rather efficient control over optical properties can be achieved by *photon* mode engineering,⁶ that is by modifying the coupling between light and electronic degrees of freedom. In particular, a strong modulation of polariton effects has been theoretically predicted^{7,8} and observed experimentally^{9–12} in long-period multiple-quantum-well (MQW) systems. These are Bragg structures, in which both the real and imaginary part of the refractive index *n* are relevant. In such structures, interference of light reflected by particular QW's leads to a large enhancement of the exciton reflectivity and its radiative recombination rate, provided that the period *P* of the MQW is equal to $\lambda/2$. Here, $\lambda = 2\pi c/\omega_0 n_b(\omega_0)$ is the light wavelength in the barrier material corresponding to the photon of the energy of the ground-state exciton in the QW, $E_x = \hbar \omega_0$. By contrast, destructive interference occurs if the anti-Bragg condition is fulfilled, $P = \lambda/4$. It is also known that exciton spectra are affected by the thickness *C* of the cladding layer.^{7,12,13} This is due to the interference of light reflected by the surface and by the exciton polariton in the MQW structure. In particular, when varying the thickness *C* from $\lambda/2$ to $\lambda/4$ the shape of the reflectivity changes from the emissionlike to the absorptionlike.

In this paper, we show that MQW structures containing DMS can be driven across the Bragg or anti-Bragg resonance by the magnetic field. Actually, we demonstrate that in appropriately designed structures the field tuning between these two extreme situations is possible in a single sample, resulting in a rather unusual sensitivity of reflectivity spectra to the external magnetic field and temperature. It is worth noting that such a field tuning has also been explored in order to determine the Rabi splitting in a microcavity containing a DMS component.¹⁴

The studied MQW structures were grown by molecular beam epitaxy (MBE) on $Cd_{0.95}Zn_{0.05}Te$ substrates in a II-VI Riber 32P system, equipped with CdTe, ZnTe, Cd, Te, Mg, and Mn effusion cells. The particular samples differ by the arrangement of $Cd_{0.96}Mn_{0.04}Te$ and CdTe QW's but in all of them both the barriers and the cladding layer are formed of $Cd_{0.93}Mg_{0.04}Zn_{0.03}Te$. This choice of the barrier material ensures a sizable confinement potential for both electrons and heavy holes.^{15–17} Being aware of a strong sensitivity of polariton effects to disorder,¹² an elaborated epitaxy procedure, described in detail elsewhere,¹⁸ was employed. In order to control precisely the composition and the thickness of particular layers, prior to the growth of proper MQW structures, the fluxes from all effusion cells were calibrated by monitoring the oscillations in the intensity of reflection high-energy

0163-1829/97/56(4)/1664(4)/\$10.00 56 R1664 © 1997 The American Physical Society

FIG. 1. Layout of the studied structures: Sample 1, upper panel; Sample 2, lower panel. Sample 1 consists of ten $Cd_{0.96}Mn_{0.04}Te$ quantum wells embodied by $Cd_{0.93}Mg_{0.04}Zn_{0.03}Te$ barriers. Sample 2 contains additionally ten quantum wells of CdTe. The samples are designed to have the periods *P* twice the thickness of the cladding layer *C*. According to x-ray diffraction $P=117.2$ nm, while $C_a = C_b = P/2 = 58.6$ nm.

electron diffraction (RHEED) during the deposition of test films. Afterwards, two buffer layers were grown, the first of which was a 250 nm thick layer of $Cd_{0.95}Zn_{0.05}Te$. The second served to minimize effects of both the interference in the buffer layer and 0.1% lattice mismatch between the substrate and the barrier material. This was achieved by gradual diminishing of the Zn content from 5% to 3% and by simultaneous increasing of the Mg concentration to 4% over the thickness of about 100 nm. The growth of the MQW structure was interrupted at each interface between the barrier and the QW in order to improve the morphology of the $Cd_{0.93}Mg_{0.04}Zn_{0.03}Te$ surface by its annealing in the Te flux. This procedure also revived oscillations in the RHEED intensity, and thus made it possible to control the epitaxy of the whole 1.2 μ m thick structures by the RHEED recording system. The QW's were grown thin enough to avoid the formation of misfit dislocations by strain relaxation. The obtained structures were characterized by x-ray diffraction, which provides the precise values of the period *P*. Satellite peaks up to seventh order were seen, which in the case of a MQW system with only ten periods points to high structural quality of the samples.

Two kinds of the MQW structures were examined by normal-incidence magnetoreflectivity at 1.6 K. The layout of these samples is depicted in Fig. 1, while the corresponding reflectivity and photoluminescence spectra are shown in Fig. 2. Sample 1 consists of ten 10 nm thick QW's of

FIG. 2. Reflectivity *R* in the region of the 1*s* e_1h_1 exciton for sample 1 (left panel) and sample 2 (right panel) in the magnetic field $B=0$, 1, 2, and 3 T (except for data at $B=3$ T, the subsequent ones are shifted by $R=0.25$). Photoluminescence in the absence of the magnetic field are shown by dashed lines. The arrows mark the exciton energy in the $Cd_{1-x}Mn_xTe$ quantum well. The layout of the structures is shown in Fig. 1.

 $Cd_{1-x}Mn_xTe$, where $x=0.40$ according to the value of the energy of the QW exciton. The distances between the QW centers *P* and between the front surface and the center of the adjacent QW *C* are designed to correspond to $\lambda/2$ and $\lambda/4$, respectively, where λ is the light wavelength in the barrier material corresponding to the exciton energy in the QW. The period $P=117.2$ nm is determined by fitting x-ray diffraction results.

Sample 2 contains additionally ten equidistant QW's of CdTe. Their thickness is 1.95 nm and they are inserted in the middle between QW's of $Cd_{0.956}Mn_{0.044}Te$, as showed in Fig. $1(b)$. Furthermore, the thickness of the barrier and the cladding layers is enlarged in order to make them commensurate with the wavelength of the exciton energy in the QW of CdTe. The actual value of *P* provided by x-ray diffraction is 120.3 nm.

We assign the two dominant reflectivity lines (at \sim 1655–1660 and 1670–1677 meV in the absence of the external magnetic field) in Fig. 2 to the ground state of the exciton polariton in the CdTe [Fig. 2(b)] and $Cd_{1-x}Mn_xTe$ [Figs. 2(a) and 2(b)] MQW's, respectively. Since $P = \lambda/2$ the exciton reflectivity is amplified by the interference effects. At the same time, they lead to an absorptionlike shape of the exciton line in the case of the $Cd_{1-x}Mn_xTe$ MQW's, for which $C = \lambda/4$ [Figs. 2(a) and 2(b)]. By contrast, an emission form of the signal is observed for the MQW's of CdTe in sample 2 [Fig. 2(b)] as for the latter $C = \lambda/2$. While polariton thermalization and localization preclude a detailed interpretation of the photoluminescence, 19 spectral positions of its maximas substantiate the above assignment of the reflectivity lines. This is consistent with theoretical calculations of 1*s* e_1h_1 exciton energies, which give 1659 meV for the QW of CdTe in sample 2 as well as 1671 meV (sample 1) and 1675 meV (sample 2) in the case of the QW's of $Cd_{1-x}Mn_xTe$. The strain^{17} was included in the computation, and the valence band offset was taken as 30% of the band-gap difference between $Cd_{1-x}Mn_xTe$ and $Cd_{1-y}Mg_yTe$,^{15,16} where *x* and *y* denote the Mn and Mg concentrations in the QW and in the barrier alloy, respec-

tively. Furthermore, a variational procedure was adopted to determine exciton binding energies. Guided by the results of this calculation we relate structures in the high-energy part of the spectra to the excited *e*1*l*1 exciton states. These weak lines will not be further analyzed.

While a moderately strong magnetic field has virtually no influence on the exciton polariton in the case of nonmagnetic MQW systems,¹² its striking effect on reflectivity spectra in the structures involving magnetic components is clearly seen in Fig. 2. Depending on the direction of circular light polarization, the magnetic field *B* perpendicular to the interfaces either diminishes (σ^+) or increases (σ^-) the exciton energy. The initial rate of the exciton splitting is 35 meV/T for Cd_{0.96}Mn_{0.04}Te at 1.6 K,²⁰ and the splitting reaches its saturation value of 75 meV when the Mn magnetic moments $g\mu_B S$ become totally polarized, $g\mu_B SB \ge k_B T$. Now, the shift in the exciton energy changes P/λ and C/λ , and thus alternates the shape of the exciton reflectivity in a way that depends on the architecture of the MQW structure.

In the case of sample 1, which contains only QW's of CdMnTe, the results of Fig. $2(a)$ demonstrate how the magnetic field, by changing the exciton energy, detunes the system of the Bragg resonance. The resonance condition $P = \lambda/2$ is seen to be met for σ^+ polarization at 0.2 T, where a rather perfectly symmetric reflectivity line is observed. As shown, not only the position but also the shape of the line varies strongly when the magnetic field is driven away from the resonance. It is worth noting that the tuning across the Bragg resonance makes it possible to determine rather precisely the value of the refractive coefficient of the barrier material. We obtain in this way $n_b = 3.18 \pm 0.01$ for our $Cd_{0.93}Mg_{0.04}Zn_{0.03}Te$ at 1670 ± 1 meV.

Particularly interesting are the results for sample 2, in which by increasing the magnetic field it makes it possible to tune the system from the Bragg to the anti-Bragg resonance. This is illustrated in Fig. $3(a)$, which presents reflectivity spectra in the region corresponding to the exciton energy in the CdTe QW's, for which the Bragg condition is met, so that both the amplitude and the width of the exciton reflectivity are very large. When increasing the magnetic field the exciton energy in the σ^+ polarization shifts down in $Cd_{1-x}Mn_xTe$, so that at 1.2 T it becomes equal to that of the excitons in CdTe QW's. This means that in this field the anti-Bragg resonance occurs, $P_{eff} = \lambda/4$. Accordingly, the exciton reflectivity becomes very small in this region. It is worth noting that most of the change of the spectrum occurs

FIG. 3. Reflectivity R in the region of the 1*s* e_1h_1 exciton of CdTe in various magnetic fields *B* from 0 to 1.5 T for sample 2 containing 10 quantum wells of CdTe and 10 quantum wells of $Cd_{0.956}Mn_{0.044}Te$: (a) experimental results; (b) results of the calculation. Except for the lowest curve, the subsequent ones are shifted by $R=0.13$. The layout of the structure is shown in Fig. 1.

over a field range as small as 0.2 T, demonstrating the efficiency of the modulation of optical properties by the magnetic field in such structures. By comparing the resonance energy to the period $P = 120.3$ nm as given by x-ray diffraction we obtain $n_b = 3.11 \pm 0.01$ for Cd_{0.93}Mg_{0.04}Zn_{0.03}Te at 1658 ± 1 meV.

The above qualitative considerations are strongly supported by a theoretical calculation presented in Fig. $3(b)$. A model discussed in detail elsewhere¹² together with the known²⁰ dependence of the exciton energy on the magnetic field in Cd $_{1-x}$ Mn *x*Te has been adopted for this calculation. In view that the radiative and inhomogeneous broadening energy, Γ_0 =0.08 meV and γ =0.7 meV, respectively, were the only adjustable parameters the agreement between the measured and calculated reflectivity spectra is to be regarded as very good.

In summary, we have proposed and demonstrated the feasibility of a method for the efficient modulation of optical properties. The method makes use of a strong dependence of the exciton energy on the magnetic field in diluted magnetic semiconductors. This dependence allows for the tuning of the exciton wavelength across the Bragg resonances in appropriately designed multiple-quantum-well structures, which is accompanied by strong changes in optical response.

The authors are grateful to J. Cibert for valuable suggestions.

* Electronic address: sadow@ifpan.edu.pl

- ¹ A.V. Komarov, S.M. Ryabchenko, O.V. Terleckii, I.I. Zheru, and R.D. Ivanchuk, Zh. Eksp. Teor. Fiz. 73, 608 (1977) [Sov. Phys. JETP 46, 318 (1977)]; G. Bastard, C. Rigaux, and A. Mycielski, Phys. Status Solidi B 79, 585 (1977); J.A. Gaj, R.R. Gałązka, and M. Nawrocki, Solid State Commun. 25, 193 (1978).
- 2For a review on DMS see, e.g., T. Dietl, in *Handbook on Semiconductors*, edited by T.S. Moss (North-Holland, Amsterdam, 1994), Vol. 3b, p. 1251; J.K. Furdyna, J. Appl. Phys. 64, R29 $(1988).$
- ³M. von Ortenberg, Phys. Rev. Lett. **49**, 1041 (1982).
- ⁴ J. Kossut and J.K. Furdyna, in *Diluted Magnetic Semiconductors*, Proceedings of the MRS Conference, edited by R.L. Aggarwal,

J.K. Furdyna, and S. von Molnar (MRS, Pittsburgh, 1987), p. 97.

- 5X. Liu, A. Petrou, J. Warnock, B.T. Jonker, G.A. Prinz, and J.J. Krebs, Phys. Rev. Lett. **63**, 2280 (1989); E. Deleporte, J.M. Berroir, G. Bastard, C. Delalande, J.M. Hong, and L.L. Chang, Phys. Rev. B 42, 5891 (1990); N. Dai, H. Luo, F.C. Zhang, N. Samarth, M. Dobrowolska, and J.K. Furdyna, Phys. Rev. Lett. **67**, 3824 (1991).
- 6See, e.g., *Confined Electrons and Photons: New Physics and Devices*, edited by C. Weisbuch and E. Burstein (Plenum, New York, 1995).
- ⁷E.L. Ivchenko, Sov. Phys. Solid State 33, 1344 (1991) ; see also, V.A. Kosobukin, *ibid.* 34, 1662 (1992); E.L. Ivchenko, A.I. Nes-

vizhskii, and S. Jorda, Phys. Solid State 36, 1156 (1994).

- ⁸ L.C. Andreani, Phys. Lett. A 192, 99 (1994); Phys. Status Solidi B 188, 29 (1995); D.S. Citrin, Solid State Commun. 89, 139 (1994); Phys. Rev. B **49**, 1943 (1994).
- $9Y$. Merle d'Aubigné, A. Wasiela, H. Mariette, and A. Shen, in *Proceeding of the 22nd International Conference of the Physics of Semiconductors, Vancouver, 1994*, edited by D.J. Lockwood (World Scientific, Singapore, 1995), p. 1201.
- 10V.P. Kochereshko, G.R. Pozina, E.L. Ivchenko, D.R. Yakovlev, A. Waag, W. Ossau, G. Landwehr, R. Hellmann, and E.O. Göbel, in Ref. 9, p. 1372.
- 11 M. Hübner, J. Kuhl, T. Stroucken, A. Knorr, S.W. Koch, R. Hey, and K. Ploog, Phys. Rev. Lett. **76**, 4199 (1996).
- ¹²Y. Merle d'Aubigné, A. Wasiela, H. Mariette, and T. Dietl, Phys. Rev. B 54, 14003 (1996).
- ¹³X.L. Zheng, D. Heiman, B. Lax, and F.A. Chambers, Appl. Phys. Lett. 52, 287 (1988).
- ¹⁴H. Ulmer-Tuffigo, R. André, J. Bleuse, F. Kany, and Le Si Dang, (unpublished).
- 15A. Waag, F. Fisher, T. Litz, B. Kuhn, U. Zehnder, W. Ossau, W.

Spahn, H. Heinke, and G. Landwehr, J. Cryst. Growth **138**, 155 $(1994).$

- ¹⁶W. Grieshaber, A. Haury, J. Cibert, Y. Merle d'Aubigné, A. Wasiela, and J.A. Gaj, Phys. Rev. B 53, 4891 (1996).
- ¹⁷P. Peyla, Y. Merle d'Aubigné, A. Wasiela, R. Romestain, H. Mariette, M.D. Sturge, N. Magnea, and H. Tuffigo, Phys. Rev. B 46, 1557 (1992).
- 18 J. Sadowski, H. Mariette, A. Wasiela, and Y. Merle d'Aubigné, Proceedings of Second International Workshop on MBE Growth Physics and Technology, Warsaw, 1996, edited by M. A. Herman [Thin Solid Films (to be published)].
- ¹⁹T. Dietl, Y. Merle d'Aubigné, A. Wasiela, and H. Mariette, in *Proceedings of the Fourth International Meeting on Optics of Excitons in Confined Systems*, Cortona, 1995 [Nuovo Cimento D **17**, 1441 (1995)].
- 20 J.A. Gaj, Y. Merle d'Aubigné, A. Wasiela, and H. Mariette, in *Proceedings of the XXI International Conference on Physics of Semiconductors, Beijing, 1992*, edited by P. Jiang and H.-Z. Zheng (World Scientific, Singapore, 1993), p. 923.