# Observation of a Ferromagnetic Transition Induced by Two-Dimensional Hole Gas in Modulation-Doped CdMnTe Quantum Wells

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The presence of a ferromagnetic transition in single, modulation-doped, 8 nm quantum well of $Cd_{0.976}Mn_{0.024}Te/Cd_{0.66}Mg_{0.27}Zn_{0.07}Te$:N is evidenced by photoluminescence magnetospectroscopy. The transition is driven by long range Ruderman-Kittel-Kasuya-Yosida interactions between Mn spins, mediated by $2 \times 10^{11}$ holes per $cm^2$. It occurs at 1.8 K, in agreement with a mean-field model. [S0031-9007(97)03602-8]

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Because of complementary properties of semiconductor and ferromagnetic material systems, a growing effort is directed toward studies of hybrid semiconductor-magnetic nanostructures. Such devices, in which both electric and magnetic fields are spatially modulated, have usually been fabricated by patterning a ferromagnetic metal on the top of a modulation-doped GaAs/AlGaAs heterostructure [1] or by incorporation of magnetic clusters directly into a semiconductor matrice [2].

In this Letter, we show that the two-dimensional hole gas confined in modulation-doped quantum wells of $Cd_{1-x}Mn_xTe$ produces, *via* the Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism, a ferromagnetic coupling between the Mn spins. Actually, by the direct observation of a ferromagnetic phase transition, we demonstrate that this coupling can overcompensate antiferromagnetic interactions specific to II-VI diluted magnetic semiconductors (DMS) [3]. Our results mean, therefore, that the well-established methods of modulation of the carrier concentration in semiconductor quantum structures can be applied for tailoring of the magnetic properties. The transition to the ferromagnetic phase is put into the evidence by observing colossal Zeeman splittings of interband optical transitions, probed here by means of photoluminescence (PL) and its excitation spectra (PLE), a technique equivalent to absorption spectroscopy but which can be used with a strongly absorbing substrate. A quantitative description of our findings confirms predictions of a recent model [4] on the free carrier-induced ferromagnetism in structures of doped DMS. It makes it also possible to evaluate the strength of many body effects for the case of two-dimensional hole gas. Moreover, the data provide important information on critical phenomena in the disordered magnetic systems of reduced dimensionality, illustrating, in particular, how long-range spin-spin interactions stabilize an ordered phase and make fluctuations of magnetization irrelevant.

Our studies have been carried out on samples grown in a molecular beam epitaxy (MBE) chamber equipped with a home-designed electron cyclotron resonance (ECR) plasma cell as a nitrogen source. Prior to fabrication of the proper structures, doping characteristics of the barrier material $Cd_{1-y-z}Mg_yZn_zTe$ have been determined by means of the Hall effect, capacitance-voltage profiles, cathodoluminescence, and x-ray diffraction. It has been found [5] that by lowering the growth temperature down to 220–240° C it becomes possible to reduce the nitrogen-induced diffusion of Mg atoms and to obtain hole concentrations up to $5 \times 10^{17}$ $cm^{-3}$ in $Cd_{1-y-z}Mg_yZn_zTe$ with $z = 0.07$ and $y$ up to 27%.

The studied modulation-doped structures consist of a single 8 nm quantum well (QW) of nominal $Cd_{0.975}Mn_{0.025}Te$ embedded in $Cd_{0.66}Mn_{0.27}Zn_{0.07}Te$ barriers grown coherently onto a (100) $Cd_{0.88}Zn_{0.12}Te$ substrate. Such a layout ensures large confinement energies for the holes in the QW, minimizing at the same time the effects of lattice mismatch. Nitrogen-doped region in the front barrier is at the distance of 20 nm (samples No. 1 and No. 2) and of 10 nm (sample No. 3) to the QW. Furthermore, in order to reduce depleting effects, two additional nitrogen-doped layers reside at the distance of 100 nm from the QW on both sides. For control purposes, an undoped structure (sample No. 4) was also grown and examined. The nominal hole concentrations in the doped structures, evaluated from a self-consistent solution of the Poisson and Schrödinger equations, are $2 \times 10^{11}$ and $3 \times 10^{11}$ $cm^{-2}$ for the two employed values of the spacer width.

The actual samples have been characterized at 2 K and in the magnetic fields up to 110 kOe by PL, PLE, and reflectivity. The latter was taken in the presence of additional white light, so that under stationary conditions the holes are neutralized by electron diffusion [6]. In such a case, we observe free excitons with a well known [7,8] giant Zeeman splitting in the $Cd_{1-x}Mn_xTe$ QW's, whose quantitative description gives $x = 0.024 \pm 0.002$ for the studied samples, in agreement with results for the reference sample No. 4. It is worth recalling at this point that the splitting of the effective mass states in DMS is directly proportional to the magnetization of the localized spins, which allows for a rather sensitive magnetometry by means of magneto-optical studies [3].

The presence of the delocalized hole gas in the modulation-doped QW's is assessed by a step-like shape of PLE spectra in the $\sigma^+$ Faraday polarization, for which optical transitions involving the majority-spin hole band are probed in DMS [3,7]. This is illustrated in Fig. 1, which also shows how the field-induced redistribution of the holes between $\pm|3/2\rangle$ heavy hole subbands, and the corresponding phase space emptying, leads to the appearance of the *quasi*free $\sigma^-$ exciton in the minority-spin band. At the same time, the Moss-Burstein shift $E_{\mathrm{MB}}$ between the energies of the PLE step and the PL maximum in the $\sigma^+$ polarization makes it possible to evaluate the hole concentration $p$. In the case of the full spin polarization, we observe $E_{\mathrm{MB}} = 11 \pm 2$ meV for the wider spacer, $L_s = 20$ nm (samples No. 1 and No. 2), while for $L_s = 10$ nm (sample No. 3) $E_{\mathrm{MB}} = 23 \pm 2$ meV. For the bare values of the electron and the heavy hole in-plane effective masses, $m_e^* = 0.1m_0$ and $m_h^* = 0.25m_0$ [9], we obtain $p = (1.6 \pm 0.2) \times 10^{11}$ cm$^{-2}$ and $p = (3.2 \pm 0.2) \times 10^{11}$ cm$^{-2}$, respectively, the values which compare favorably with those expected from the design of the structures. This may point to a little importance of the renormalization of the Moss-Burstein shift by localization and hole-hole interactions in the case of their full spin polarization. It is important to note that the formation of magnetic polarons is possible in DMS provided that disorder is sufficiently strong [10]. The absence of the hole localization indicates, therefore, the high quality of the structures.

A thorough examination of the PL as a function of both temperature and the weak magnetic field has been carried out in a system equipped with copper Helmholtz coils and a rotary pump that allows one to reduce the temperature of the helium bath down to 1.3 K. Furthermore, in order to minimize heating of the hole and spin subsystems, the incoming photon energy was smaller than the barrier gap (direct excitation of the QW) and the laser power was kept below 50 $\mu$W per mm$^2$ of the illuminated surface. As shown in Figs. 2 and 3, the PL splitting $\Delta$ under such conditions is not only exceptionally large, but increases in a dramatic way on lowering temperature. Actually, in all doped samples, we observe an unusually large value of $\partial\Delta/\partial H$ at $T \geq 2$ K, and a zero field splitting below $T_c = 1.8$ K. The latter is shown in the right part of Fig. 2 for samples No. 1 and No. 2; in the case of sample No. 3, the low energy PL maximum is obscured by substrate lines. The large value of $\partial\Delta/\partial H$ explains also a strong polarization of the hole liquid in very small magnetic fields witnessed by the PLE spectra of Fig. 1. No such effects are visible either in the undoped structure or in the presence of illumination by white light that depletes the QW from the carriers. The appearance of zero-field splitting is preceded by a critical increase of $\chi$,

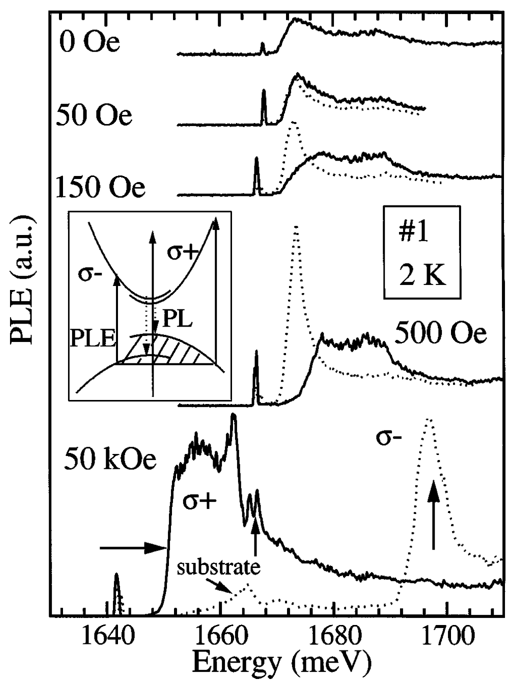


FIG. 1. PLE spectra for selected values of the magnetic fields in a modulation-doped $p$-type QW of $Cd_{0.976}Mn_{0.024}Te$ (sample No. 1) at 2 K for $\sigma^+$ (solid lines) and $\sigma^-$ (dotted lines) circular polarizations. The PL was collected in $\sigma^+$ polarization at energies marked by the narrowest features. The sharp maximum and step-like form (denoted by large arrows in 50 kOe data) correspond to quasifree exciton and transitions starting at the Fermi level, respectively. Substrate lines (denoted by small arrows) are superimposed on data for 50 kOe. The band arrangement at 150 Oe is sketched in the inset.

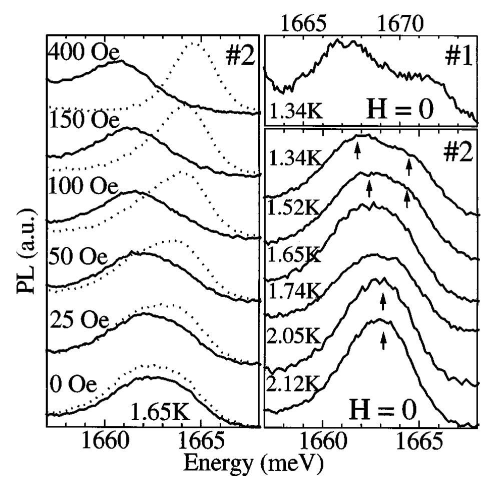


FIG. 2. Photoluminescence intensity in modulation-doped $p$-type QW's of $Cd_{0.976}Mn_{0.024}Te$ for selected values of the magnetic field $H$ at 1.65 K (left panel, sample No. 2) or temperature at $H = 0$ (right panel, samples No. 1 and No. 2). Solid and dotted lines in left panel correspond to $\sigma^+$ and $\sigma^-$ circular polarizations, respectively. Arrows denote values resulting from extrapolation to $H \to 0$ of data in the magnetic field, presented in Fig. 3(a).

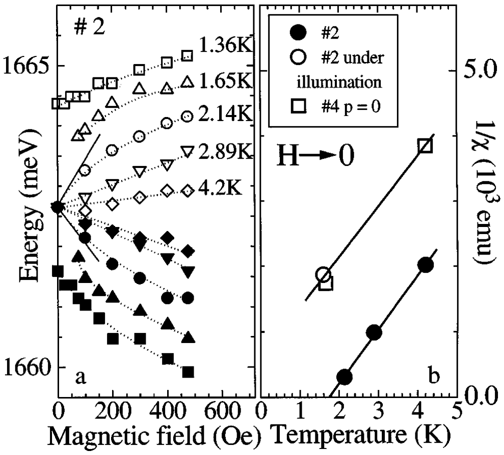


FIG. 3. (a) Energy $E_{\pm}$ of photoluminescence maxima at $\sigma^+$ and $\sigma^-$ circular polarizations (full and empty symbols, respectively) as a function of the magnetic field $H$ for selected temperatures in a modulation-doped $p$-type QW of $Cd_{0.976}Mn_{0.024}Te$ (sample No. 2). Dotted lines are guides for the eye; solid lines denote the assumed initial slope of $E_{\pm}$ at 2.14 K. (b) Inverse magnetic susceptibility calculated from $d(E_- - E_+)/dH$ at $H \to 0$, as given by data in (a) for $p$-type QW of $Cd_{0.976}Mn_{0.024}Te$ (full circles). Note the presence of a ferromagnetic transition. Results for empty $Cd_{0.976}Mn_{0.024}Te$ QW's, where antiferromagnetic interactions dominate, are shown by empty symbols. Lines are guides for the eye.

as shown in Fig. 3(b) for sample No. 2. We determine the susceptibility in the limit of vanishing fields as

$$\chi(T) = (\partial M/\partial H) = (g\mu_B/|\alpha - \beta|)(\partial\Delta/\partial H), \quad (1)$$

where $g = 2.0$ is the Landé factor of the Mn spins; $\alpha$ and $\beta$ are $s$-$d$ and $p$-$d$ exchange integrals, respectively, whose values are well known for $Cd_{1-x}Mn_xTe$ [3,7]. The critical behavior of $\chi(T)$ is in contrast to gradual changes of $\chi(T)$ associated with the formation of magnetic polarons [11,12], for which the finite volume involved precludes the existence of any second-order phase transition [10,11].

We interpret our findings as a ferromagnetic phase transition driven by the free holes, which by means of the RKKY mechanism mediate ferromagnetic exchange interactions between the Mn spins. This conclusion is strongly supported by the recent theoretical model [4]. According to this model we may write $\chi(T) = C/(T + T_o - \Theta)$, where $C$ is the Curie constant while $T_0 > 0$ takes short range antiferromagnetic interactions and a demagnetization correction into account, both $C$ and $T_0$ determined previously for $Cd_{1-x}Mn_xTe$ [7]. The influence of delocalized carriers is described by $\Theta$ which for the case of a two-dimensional hole gas residing in a QW of an effective thickness $\tilde{L}_W$ assumes the form [4]

$$\Theta = (C/g^2\mu_B^2)\beta^2 A_F(m_h^*/4\pi\hbar^2\tilde{L}_W). \quad (2)$$

Here, $A_F \geq 1$ is a Fermi-liquid parameter describing the combined effect of disorder and hole-hole correlations upon the RKKY interaction [4,13] and $\tilde{L}_W = 6.3$ nm is an effective width of the hole layer, which is related to the hole ground state envelope function $\varphi(z)$ by $\tilde{L}_W = 1/\int dz|\varphi(z)|^4$.

According to the data of Fig. 3(b), and in agreement with the model, the same values of $C$ are observed in the presence and the absence of the holes. Additionally, by fitting the data for the doped sample, we determine $\Theta = T_c - T_0$, from which we obtain $A_F = 2.5 \pm 0.5$, a reasonable value to say the least [4,13]. It is worth noting that all material parameters conspire to make $\Theta$ by a factor of about 50 greater for the holes than for the electrons in a given DMS QW.

According to the experimental results of Figs. 2 and 3, spin polarization of the holes is seen to increase rather rapidly below $T_c$. Neglecting renormalization effects to be discussed below, the splitting of the heavy hole subband at their full spin polarization is twice the Fermi energy in the absence of polarization, which gives $\Delta \approx 5.7$ meV for $p = 2 \times 10^{11}$ cm$^{-2}$ (samples No. 1 and No. 2), a value consistent with the experimental results of Figs. 2 and 3(a). The magnitude of Mn magnetization in the corresponding molecular field [4] $H^* = \beta p/2g\mu_B\tilde{L}_W \approx 800$ Oe, attains only about 5% of its saturation value in our structures. Such an unsaturated ferromagnetic phase is predicted by the model [4], but the present experimental results do not rule out completely other types of spin arrangements, such as occurring in ferrimagnetic or spiral phases. Work is under way aimed at determining the domain structure and the coercive force in this novel ferromagnetic system.

An important question addressed by our results concerns mechanisms which could lead to the violation of the Mermin-Wagner theorem on the absence of the long-range order in two-dimensional systems with Heisenberg spin-spin interactions [14]. We note, first of all, that a combination of the spin-orbit coupling with the confinement and strain effects make the exchange interaction between the ground state holes and the Mn spins to be Ising-like in DMS QW's, the axis of easy hole spin polarization being perpendicular to the interface [15]. Moreover, the de Broglie wavelength of the carriers, $\lambda_F = 2\pi/k_F \approx 50$ nm, is much longer than the average distance between the localized spins in DMS, $\bar{r} = (4\pi x N_0/3)^{-1/3} \approx 1$ nm. Accordingly, the first zero of the RKKY function occurs at a distance much greater than $\bar{r}$, so that the ferromagnetic coupling $J(r)$ mediated by the holes is effectively long range. This indicates, in particular, that thermodynamic fluctuations of magnetization will strongly be suppressed. Critical exponents for such a case were determined by Fisher, Ma, and Nickel [16], who considered the $d$ dimensional space and showed that as long as $\sigma < d/2$ in the dependence $J(r) \sim 1/r^{d+\sigma}$, the mean-field approach to the long wavelength susceptibility $\chi(T)$ is valid, a conclusion not affected presumably by disorder in the spin distribution. In contrast, the critical exponents [16] $\eta = 2 - \sigma$ and $\nu = 1/\sigma$ point to much faster decay of $\chi(q)$ with $q$ than that expected from the classical Ornstein-Zernike theory [17]. This means that the mean-field model [4] of the ferromagnetic transition driven by the RKKY interactions should remain valid down to at least $|T - T_c|/T_c \approx (\bar{r}k_F)^2$. At the same time, unlike the case of short range interactions, the length scale of magnetic correlations is set by $\lambda_F$, not by $\bar{r}$. This may explain the lack of critical broadening of the PL and PLE lines in our experiments. The same mechanism may account for the virtual absence of critical scattering of the carriers by the Mn spins in bulk $Pb_{1-x-y}Mn_xSn_yTe$ [18], where the free holes drive a ferromagnetic transition [19], and its presence in, *e.g.*, EuS:Ga and EuO:Gd [20], in which a short-range ferromagnetic interaction dominates [21].

Another interesting aspect of the data concerns the role of static disorder and carrier-carrier interactions. As already mentioned, those effects are thought to enhance $\Theta$. They account also for a band-gap renormalization [22] that amounts to 29 meV for the studied samples, as deduced from the energy difference between the free exciton reflectivity for the empty QW and the PL in the presence of the holes ($7 \pm 1$ meV at $H = 0$ for samples No. 1–3), and from an exciton binding energy of 22 meV, according to an estimate based on the $1s$-$2s$ excitonic energy separation [23] in the undoped sample No. 4. The Fermi-liquid effects are expected to vary strongly with the degree of carrier spin polarization. This invalidates attempts to describe $\Delta(H)$ and $\Delta(T)$ below $T_c$, neglecting disorder and within a one-electron model. It appears, however, that doped QW's of DMS are well suited for a quantitative examination of those effects, as the spin polarization sets in the fields, at which the Landau quantization is not yet relevant. This will be the subject of future studies.

In conclusion, our findings demonstrate that $p$-type doping constitutes the method for substantial enhancement of magnetic, and then magneto-optical effects in DMS, leading—in appropriately designed structures—to a ferromagnetic phase transformation. Since the interactions between localized spins mediated by the carriers are long range, a simple mean-field approach gives a correct quantitative description of magnetic properties, even in reduced dimensionality systems. While our results demonstrate the possibility of changing the magnetic phase by light, other means—such as gates—are expected to provide also a high degree of control over magnetic properties in modulation-doped structures. This opens new perspectives for further studies of coupled carrier liquids and localized spins in novel geometries and material systems.

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[1] P. Ye *et al.*, Phys. Rev. Lett. **74**, 3013 (1995); Semicond. Sci. Technol. **11**, 1613 (1996), and references therein; S. Izawa, S. Katsumoto, A. Endo, and Y. Iye, J. Phys. Soc. Jpn. **64**, 706 (1995); F. B. Mancoff *et al.*, Phys. Rev. B **51**, 13 269 (1995).

[2] Jing Shi, S. Gider, K. Babcock, and D. D. Awschalom, Science **271**, 937 (1996), and references therein.

[3] For 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).

[4] T. Dietl, A. Haury, and Y. Merle d'Aubigné, Phys. Rev. B **55**, R3347 (1997).

[5] S. Tatarenko *et al.*, in *Proceedings IX International Conference on Molecular Beam Epitaxy*, Malibu 1996, J. Cryst. Growth (to be published).

[6] A. J. Shields *et al.*, Semicond. Sci. Technol. **11**, 890 (1996), and references therein.

[7] J. A. Gaj *et al.*, in *Proceedings XXI International Conference on Physics of Semiconductors*, Bejing 1992, edited by P. Jiang and H.-Z. Zheng (World Scientific, Singapore, 1993), p. 923; Phys. Rev. B **50**, 5512 (1994).

[8] G. Mackh *et al.*, Phys. Rev. B **49**, 10 248 (1994).

[9] G. Fishman, Phys. Rev. B **52**, 11 132 (1995).

[10] C. Benoit à la Guillaume, Phys. Status Solidi (b) **175**, 369 (1993).

[11] T. Dietl and J. Spałek, Phys. Rev. Lett. **48**, 355 (1982).

[12] T. Wojtowicz, S. Koleśnik, I. Miotkowski, and J. K. Furdyna, Phys. Rev. Lett. **70**, 2317 (1993).

[13] B. L. Altshuler, A. G. Aronov, and A. Yu. Zyuzin, Pisma Zh. Eksp. Teor. Fiz. **38**, 128 (1983); see also B. L. Altshuler and A. G. Aronov, in *Electron-Electron Interaction in Disordered Systems*, edited by A. L. Efros and M. Polak (North-Holland, Amsterdam, 1985), p. 1; H. Fukuyama, *ibid.*, p. 155; P. A. Lee and T. V. Ramakrishman, Rev. Mod. Phys. **57**, 287 (1985).

[14] N. D. Mermin and H. Wagner, Phys. Rev. Lett. **17**, 1133 (1966).

[15] P. Peyla *et al.*, Phys. Rev. B **47**, 3783 (1993); B. Kuhn-Heinrich *et al.*, Solid State Commun. **91**, 413 (1994).

[16] M. E. Fisher, S.-k. Ma, and B. G. Nickel, Phys. Rev. Lett. **29**, 917 (1972).

[17] See, J. M. Yeomans, *Statistical Mechanics of Phase Transitions* (Oxford University Press, Oxford, 1993), p. 57.

[18] P. Łazarczyk, W. Dobrowolski, and T. Story, in *Extended Abstracts of International Conference on Electron Localization and Quantum Transport in Solids, Jaszowiec, Poland, 1996*, edited by T. Dietl (Institute of Physics, Warsaw, 1996), p. 129.

[19] T. Story, R. R. Gałązka, R. B. Frankel, and P. A. Wolff, Phys. Rev. Lett. **56**, 777 (1986); H. J. M. Swagten *et al.*, Phys. Rev. B **37**, 9907 (1988).

[20] S. von Molnar and T. Kasuya, Phys. Rev. Lett. **21**, 1757 (1968); S. von Molnar and M. W. Shafer, J. Appl. Phys. **41**, 1093 (1970).

[21] P. Wachter, CRC Crit. Rev. Solid State Sci. **3**, 189 (1972).

[22] S. Schmitt-Rink, D. S. Chemla, and D. A. B. Miller, Adv. Phys. **38**, 89 (1989), and references therein.

[23] H. Mathieu, P. Lefebvre, and P. Christol, Phys. Rev. B **46**, 4092 (1992).