Light controlled and probed ferromagnetism of (Cd,Mn)Te quantum wells

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Abstract

Properties of carrier-induced ferromagnetism in modulation-doped quantum wells of *p*-type (Cd,Mn)Te are assessed by photoluminescence (PL) and PL excitation spectroscopy. Well chosen illumination makes it possible to deplete the quantum well and thus to control the strength of the ferromagnetic interactions. Spontaneous local magnetization is proportional to the hole concentration p but the Curie temperature T_C varies little over a wide p range, in agreement with the mean-field model for 2D systems. The spatial extension ξ of ferromagnetic correlation is probed by micro- and time-resolved PL and possible mechanisms controlling ξ below T_C are discussed.

1. Introduction

The possibility of creating a ferromagnetic order within the subsystem of the localized spins by either equilibrium [1] or photo-excited carriers [2] has been considered for diluted magnetic semiconductors (DMS) [3] for many years. Among early experimental achievements was the generation of magnetization by circularly polarized light [2] as well as the demonstration of carrier-controlled ferromagnetism in *p*-type (Pb,Sn,Mn)Te [4]. A number of more recent developments have stimulated the interest in ferromagnetic DMS even further. First, low-temperature epitaxy of (In,Mn)As [5], and then (Ga,Mn)As [6], has provided ferromagnetic III-V DMS, in which complementary properties of semiconductor quantum structures and ferromagnetic systems can be explored. Second, the theoretical prediction [7] that the band holes in II-VI DMS can induce a ferromagnetic order has been experimentally confirmed for both 2D [8,9] and bulk systems [10]. Third, there is a growing amount of findings demonstrating successfully the modulation of ferromagnetic properties isothermally by external means: photon flux [8,9,11] and electric field [12]. Finally, while in the case of (Ga,Mn)As the Curie temperature T_c attains a value up to 110 K [6], there are theoretical [13] and experimental [14] indications that room temperature ferromagnetic DMS can be synthesised as well as that the family of ferromagnetic DMS extends toward group IV elemental semiconductors [13,15].

Mn-based II-VI DMS are known for their peculiar magneto-optical properties such as giant Zeeman splitting of the band states and the related Faraday rotation [3]. These phenomena are caused by a strong exchange coupling between the carriers in the s,p bands and the electrons residing on the 3d shell of the magnetic ions. The interaction between these two subsystems leads to a ground state with long-range ferromagnetic correlation if the carrier-mediated ferromagnetic exchange is strong enough to overcome intrinsic short-range antiferromagnetic interactions. Such conditions can be achieved in quantum wells of $Cd_{1-x}Mn_xTe$ containing both a few percent of Mn and a heavy-hole liquid [7-9].

In this paper, we summarize results of our investigations aiming at the determination of the nature of the low-temperature phase in modulation-doped *p*-type quantum wells (QW) of $Cd_{1-x}Mn_xTe$, as well as at tracing the evolution of the system properties as a function of temperature T, hole concentration p, and Mn content x. Due to the excellent quality of MBE-grown II-VI structures, their magnetic properties can be assessed by sensitive and powerful magnetooptical methods, provided that the influence of carrier-carrier correlation on optical spectra is taken into consideration. Continuous-wave (cw), time-resolved, and micro-photoluminescence (PL) and PL excitation (PLE) spectroscopy are used to examine magnetic phases and, in particular, to evaluate the ferromagnetic correlation length. Importantly, light can be used not only to probe the ferromagnetism but also to control it. In particular, the photons *via* generation of electron-hole pairs in the barriers deplete the QW, which suppresses the ferromagnetic order in an isothermal and reversible way.

2. Samples and experiments

The samples containing a single 8 nm wide *p*-type $Cd_{1-x}Mn_xTe$ QW were grown [16] by molecular beam epitaxy on $Cd_{0.88}Zn_{0.12}Te$ (001) substrates. The barriers are made of $Cd_{1-y-z}Mg_yZn_zTe$, in which the Mg content (y = 0.25-0.28) determines the valence band offset, while the presence of Zn (z = 0.08-0.07) ensures a good lattice match to the substrate, so that the whole structure is coherent. The barriers are doped by nitrogen at three positions: at the distance L_s = 20 nm from the QW on the surface side and, in order to pin the Fermi level by neutralizing interface or surface charges, at 100 nm on each side of the QW. The doping is sufficiently high to achieve the limit, in which p is determined by the thickness L_s of the spacer layer. This reduces the influence of fluctuations in the nitrogen concentration.

The design of the heterostructure makes it possible to tune the hole concentration isothermally by varying the intensity of illumination by photons with energy greater than the band gap of the barrier material. The photo-holes generated in this way are partially trapped in the barriers while the photo-electrons migrate to the QW and recombine with the holes confined therein. The trapped holes tunnel more slowly to the QW. A competition between these two processes determines the steady-state hole concentration p.

The experiments are carried out in helium cryostats with samples mounted strain-free and immersed in pumped liquid helium. The magnetic field is generated by a superconducting magnet or, in the low field range, by a pair of Helmholtz coils. PL is excited by an Al_2O_3:Ti laser with power less than 9 mW/cm^2 tuned below the energy gap of the barriers, so that neither carrier concentration nor spin temperature are perturbed. An additional illumination, serving to change the hole concentration, is produced by a standard halogen lamp equipped with one blue and several gray filters. Such light is bright enough to evacuate the hole gas almost completely when any gray filter is inserted, but the intensity is low enough to avoid any spin heating.

In order to determine the hole concentration quantitatively the procedure developed previously is applied [9,17]. The Moss-Burstein shift between PLE and PL spectra provides the magnitude of p, evaluated assuming the standard values of the electron and hole masses,

m_e= 0.1m_0 and m_{hh}=0.22 m_0 [18], respectively, and neglecting the possible influence of disorder and carrier-carrier correlation on optical spectra. Since the shift is two times greater if the 2D carrier liquid is spin polarized, in order to improve the accuracy, the spectra are taken in a magnetic field of 1 T. In agreement with the previous studies [17], the relation between the light intensity I and the corresponding p value is well described by the formula expected for tunneling through a triangular potential barrier, $I = A(p - p_0)\exp(-B\sqrt{p})$, where p and p_0 are the hole concentrations with and without illumination, respectively. The parameters A and B, describing the rate of electron migration and hole tunneling for a given structure, are determined from the fit to the experimental data in the high hole concentration region. The above equation is then used to determine p at larger I, for which the Moss-Burstein shift is small.

In order to verify QW parameters magneto-transmission measurements are carried out under conditions of almost complete depletion of the QW from the holes. The Mn content x is obtained precisely by comparing the exciton Zeeman splitting measured in the Faraday configuration up to 5 T to the well know bulk values [19].

3. Phase transition and critical temperature

In the presence of the sp-d exchange interaction, the spin splitting of effective mass states is proportional to the magnetization of the Mn ions in the region visited by the corresponding quasi-particle [3]. Thus, optical transitions can be used to probe the magnetic state of the QW despite the extremely low amount of magnetic material. In particular, either spontaneous or field-induced magnetization can be determined from polarization-sensitive PL measurements.

Experimental results putting into evidence the existence of the hole-induced phase transformation in a QW of p-type $Cd_{1-x}Mn_xTe$ are summarized in Figs. 1-3. There are two ways of crossing the phase boundary in the present system: by varying T at constant p, or isothermally, by light-induced changes of p. PL spectra obtained for these two cases are collected in Fig. 1. As shown, a single line is observed above a critical temperature T_C. This line corresponds to optical

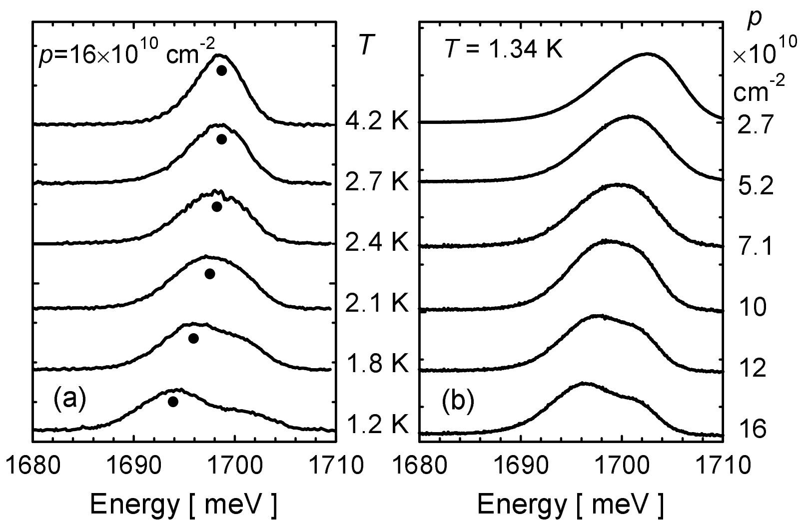

Fig. 1 PL spectra at various temperatures (a) and hole concentrations (b) in p-type $Cd_{0.96}Mn_{0.04}Te$ quantum well. Peak positions of the low energy line are marked by full circles in (a).

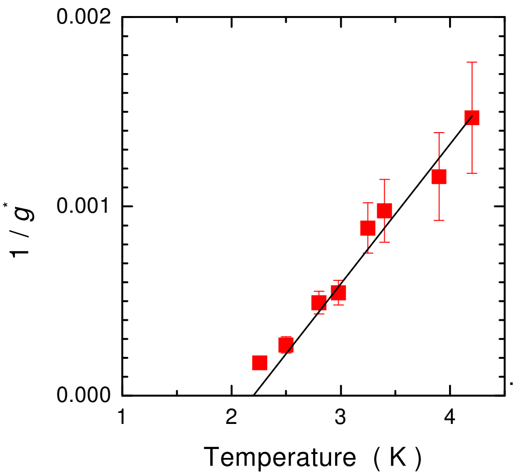

Fig.2. Inverse effective Landé factor g of the PL line in p-type $Cd_{0.96}Mn_{0.04}Te$ quantum well showing ferromagnetic transition at T_C = 2.2 K. The straight line represents the slope measured for bulk $Cd_{0.96}Mn_{0.04}Te$ [18], and demonstrates no effect of the holes on the Curie constant C.*

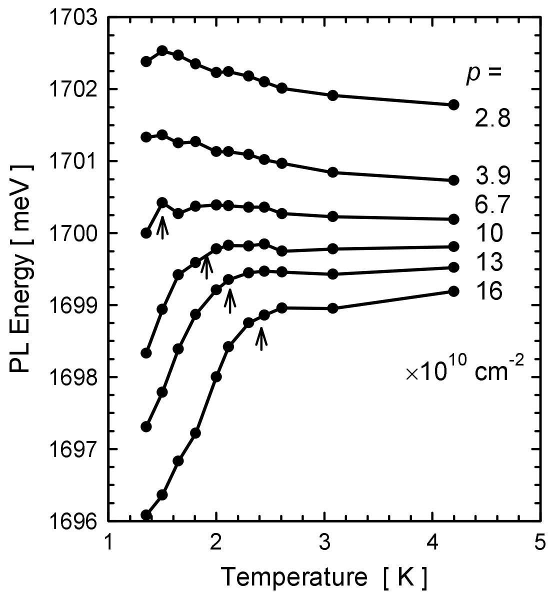

Fig. 3 Temperature dependencies of the low-energy peak positions for selected values of the hole concentrations changed by barrier illumination in p-type $Cd_{0.96}Mn_{0.04}Te$ quantum well. Arrows indicate temperature of ferromagnetic transition.

transitions involving the electron and hole ground-state subbands (e1-hh1). In a small magnetic field H, the line splits into two components, exhibiting opposite circular polarizations, separated by the energy ΔE. According to Fig. 2, the corresponding effective Landé factor defined by $g^*\mu_B = d\Delta E/dH$ at $H \to 0$, undergoes a critical divergence at $T \to T_C^+$, $g^* = C(T - T_C)^{-1}$. Below T_C, a zero-field splitting of the line becomes visible with a characteristic thermal shift of its lower component, as shown in Figs. 1 and 3.

None of the above effects have been detected in undoped DMS QW or in p-type non-magnetic QW. Furthermore, the critical behavior discussed here is in contrast to gradual changes of line splittings as a function of temperature associated with the formation of bound magnetic polarons [20,21], for which the finite volume involved precludes any second order phase transition [20,22]. Thus, the accumulated results lead one to the conclusion [8,9] that the mutual interaction between the 2D hole liquid and the localized spins results in a magnetic phase transition. According to the mean-field approach (MFA) [7], the ground state is ferromagnetic. The corresponding formula for T_C has been derived by some of the present authors [7-9] and others [23], the applicability of the MFA being justified by the long-range character of the ferromagnetic interactions. In the theory adopted here [7-9]: (i) the contribution of short range antiferromagnetic interactions is included in terms of an effective spin concentration and temperature; (ii) the hole-hole interactions are described within the Fermi liquid theory; (iii) the assumption that the hole liquid is in the highly degenerate limit is relaxed, and (iv) electrostatic disorder is taken into account by gaussian broadening of the hole density of states.

As shown in Fig. 4, there is a good agreement between experimental and theoretical values of T_C as a function of x and p. The experimental data are obtained by tracing the position of the lower PL line as a function of T at constant p. This procedure makes it possible to avoid

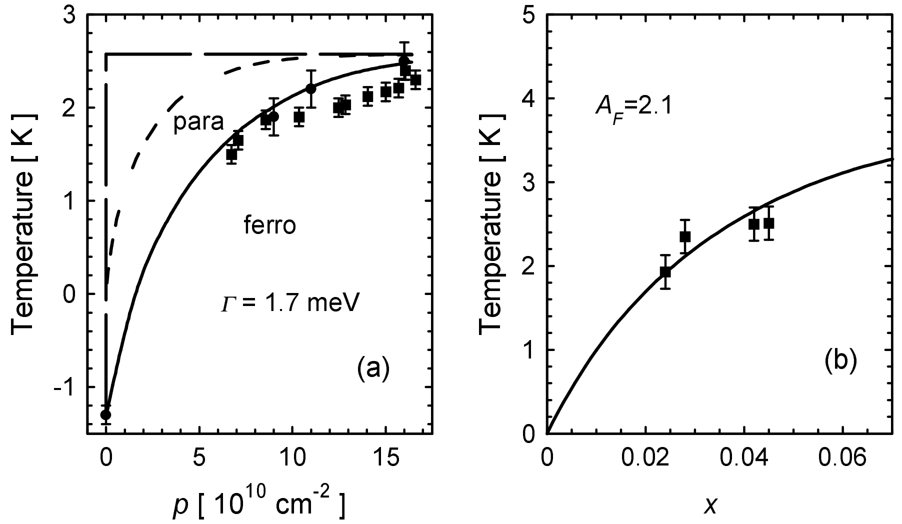

Fig. 4 Critical temperature T_C of the ferromagnetic transition, as a function of the hole (a) and Mn (b) concentrations in p-type $Cd_{0.96}Mn_{0.04}Te$ quantum well. Solid lines are theoretical taking enhancement of T_C by disorder (a) and hole-hole (b) correlation into account. The dashed line in (a) is theoretical neglecting disorder (Γ=0). The step function (dashed line) is calculated for 2D degenerate hole gas neglecting disorder.

uncertainties associated with renormalization of the PL energy by carrier-carrier correlation, which depends more on p than on T, an effect visible in Fig. 1(b). The theoretical curves are drawn with two adjustable parameters: the Fermi liquid parameter $A_F = 2.1$ and the full-width at half maximum of the density-of-states gaussian broadening Γ = 1.7 meV. Such a twofold enhancement of the Pauli susceptibility by the interactions, as implied by the magnitude of A_F, is to be expected for the hole concentration range in question [23]. In the absence of disorder, the MFA value of T_C is independent of p for a degenerate 2D carrier gas [7]. According to Fig. 4(a), the non-zero values of Γ and T account for the lowering of T_C observed in the regime of small hole concentrations. The onset of strong hole localization is expected in this regime, where the Fermi energy becomes comparable with Γ.

4. Low-temperature phase and magnetic domains

According to the MFA [7], the hole gas in the QW should be completely spin-polarized immediately below T_C. While disorder and carrier-carrier correlation perturb the development of the full polarization, it appears that below $T \approx 0.7T_C$ the half metallic situation is reached [9]. At the same time, the Mn spins remain far from the saturation as the molecular field produced by the polarized holes is rather small, of the order of 1 kOe. This field and, therefore, corresponding values of both the Mn magnetization M_f and the splitting of the PL line are expected to be proportional to the hole concentration. Experimental values of the zero-field splitting at 1.34 K, plotted versus carrier concentration in Fig. 5, confirm this prediction. Actually, the experimental splitting values are about 25% larger than the calculated ones, but a quantitative comparison should take into account the effects of carrier-carrier correlation upon optical spectra and their incidence on the determination of the hole concentrations.

Before discussing experiments aiming at elucidating the microscopic spin arrangement in the low-temperature phase, it is instructive to contemplate the unusual micromagnetic characteristics of the system. First, because of a strong interaction of the hole spin with strain and confinement potentials [24], the p-d exchange energy exhibits an appreciable uniaxial anisotropy with the easy axis oriented along the growth direction. Second, the static response function of the 2D degenerate fermion gas is known to be q-independent up to $q = 2k_F$. This means that the magnetic stiffness is small, *i.e,* that ground states corresponding to ferromagnetic order, $q = 0$, or to spin-density waves (SDW), $0< q \leq 2k_F$, have similar energies and, therefore, either is *a priori* possible [9].

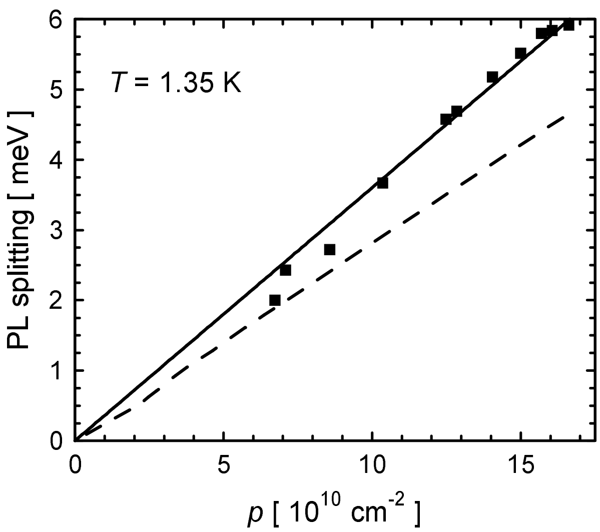

Fig. 5 Zero-field splitting of the PL line in p-type $Cd_{0.96}Mn_{0.04}Te$ quantum well as a function of the hole concentration. Solid line is drawn through data; dashed line is calculated neglecting effects of hole-hole correlation on optical spectra.

Since the easy axis is oriented along the growth direction, the intensity of PL lines for the two circular polarizations in the Faraday geometry provide information about the relative amount of spin packets with the two relevant orientations, as shown schematically in Fig. 6(a). The difference in the integrated intensities of the two components contributing to the PL line at given polarization, normalized to their values at the initial field $H = 0$, is depicted as a function of the magnetic field at 1.5 K in Fig. 6(b). The data represents an optically determined hysteresis loop, from which a number of conclusions emerge. First, these data point to values of remanent magnetization and coercive force below our experimental resolution, $M_r < 0.05M_f$ and $H_c < 4$ Oe, respectively. The virtual absence of remanence and hysteresis in the studied system is further supported by the observation that the spectra for the two polarizations, that is the amount of two kinds of the spin packets, is identical at $H = 0$, even after cooling the sample from above T_C down to 1.5 K in a magnetic field of 500 Oe. Second, while the in-plane field does not affect the spectra at all, the spin packets become aligned completely by the magnetic field $H_{al} \geq 100$ Oe applied along the growth axis. This confirms a strong anisotropy, and raises the question about mechanisms accounting for a relatively large magnitude of H_{al}.

In order to tell between the models invoking either SDW or ferromagnetic domains, micro- and time-resolved PL studies have been carried out. The micro-PL was performed with 1 μm spatial resolution in both excitation and detection channels. The spectra for both circular polarizations were identical to those obtained over a larger spot. In terms of the magnetic domain scenario there are two explanations to this finding: either the domains are smaller than 1 μm, or domain boundaries move faster than the time of spectra integration (several minutes). Within the SDW model, short length and time scales are, of course, expected. In particular, after the photoexcitation by a circularly-polarized pulse, the PL line should loose its polarization during

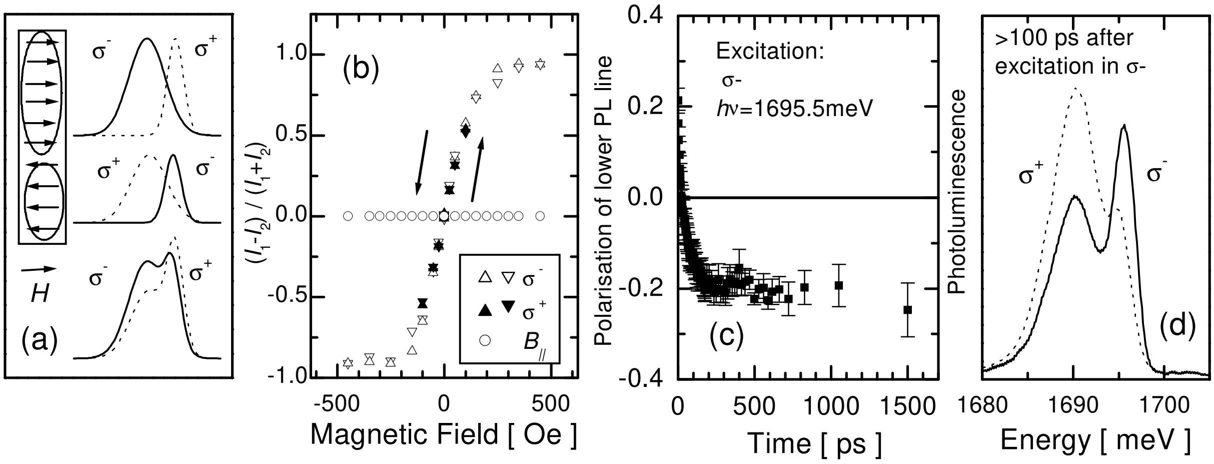

Fig. 6 (a) Schematic presentation of PL lines in the presence of magnetic domains; (b) magnetization loop determined from PL intensities in the two circular polarizations for the magnetic field perpendicular and parallel to the QW plane; up and down triangles stand for up and down field scans respectively; (c) time integrated PL spectra taken with the delay of 100 ps after resonant excitation by circularly polarized pulse; (d) time dependence of the polarization degree for the lower PL line.

the time corresponding to thermalization processes, as the SDW eigenstates at $H = 0$ consist of a coherent superposition with equal weights of the two spin orientations. To check this conjecture, the PL polarization degree P was measured as a function of time after resonant excitation within the upper line using circularly polarized pulses. As shown in Fig. 6c, during the first 100 ps, P decreases, changes sign, and finally stabilizes at a non-zero value of $P = -0.2$. This P value remains virtually unchanged as long as the PL can be detected, up to 1 ns. In this time domain the PL spectrum is similar to that observed in the cw regime and $H \neq 0$ (Fig.6d). Such a persistent polarization was not observed in the paramagnetic phase.

The data above imply that spin relaxation of photocarriers occurs during the initial 100 ps, in good agreement with spin relaxation times measured for electrons in samples with a lower Mn content [25]. Importantly, the non-zero value of P after thermalization contradicts the SDW model. Such an observation can be explained, in turn, assuming the presence of domains. Indeed, in the domain model, the time-independent value of P means that because of tunneling barriers and/or localization, the electron needs more than 1 ns to diffuse to a neighboring domain. The question then arises what is the mechanism accounting for the domain formation and for the magnitude of H_{al}. From the known magnitude of the zero-field spin-splitting, the value of the domain magnetization M_f, and its contribution to the magnetic induction can be evaluated, $B_f = 4\pi M_f = 14 \pm 1$ Gs at 1.5 K. Since $B_f << H_{al}$, the long-range dipole interactions do *not* constitute the dominant mechanism driving the domain formation. We suggest tentatively that the short-range antiferromagnetic interactions are involved and result in small values of the ferromagnetic correlation length.

5. Summary

Recent magneto-optical studies on properties of carrier-controlled ferromagnetism in p-type (Cd,Mn)Te-based quantum wells have been presented. It has been found that light control over ferromagnetic correlation is possible over a wide range. Both Curie temperature and zero-field splitting of the luminescence line are satisfactorily described by the mean-field approximation, provided that carrier-carrier correlation and static disorder are taken into account. In particular, the transition temperature is weakly concentration dependent, whereas the spin splitting is proportional to the concentration of the holes. Results of time-resolved spectroscopy support the existence of magnetic domains. It is conjectured that the small value of the remanent magnetization and the large magnitude of the magnetic field aligning the domains reflect the influence of short-range antiferromagnetic interactions.

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