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# Optically induced instability of spin precession in magnetic quantum wells

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Dynamic phase separation in CdMnTe quantum wells under femtosecond pulse excitation leads to formation of hot and cold spin domains. Using Fourier spectroscopy of the time-resolved magneto-optical Kerr effect we determine the temperatures and the total areas of each kind of domain. The instability is shown to be triggered by the magnetic field above a threshold value.

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In a great variety of natural phenomena the nonlinearity in stochastic systems far from equilibrium is responsible for a complex behavior and auto-organization.[1] Among such systems one can mention chemical reactions, where local-density fluctuations are amplified by autocatalytic mechanism, or biological systems, where the feedback loop is controlled by generation of ferments. In magneto-optics, a complex behavior of magnetization vector can be expected in diluted magnetic semiconductors (DMS's),[2] that is, semiconductors, alloyed with transition metals, e.g., manganese. Indeed, under optical excitation the spins of magnetic ions are heated via spin flips with electron spins due to $s$-$d$ exchange interaction, and this heating is more efficient in hotter spin regions.[3] Thus in such a system any local fluctuation of ions spin temperature is amplified by the positive feedback loop. Therefore one expects the inhomogeneity of spin temperature that eventually results in dynamic phase separation. In this paper, we show that such phase separation does occur in CdMnTe quantum wells (QW's) subjected to the magnetic field in Voigt geometry under femtosecond pulse excitation, resonant with the QW fundamental optical transition. Using the time-resolved magneto-optical Kerr technique[4] we provide a direct evidence of hot and cold domains formation and determine both temperatures and total areas of these domains, as well as the magnetic-field intensity threshold for domain formation. Finally, the effect of optical excitation intensity and photon energy is discussed.

Our experimental technique exploits the well-known magneto-optical Kerr effect in a pump-probe geometry to detect the spin polarization in the sample excited by 100-fs circularly polarized laser pulses. The Kerr rotation is then monitored by a weak probe pulse. The repetition rate of the laser pulses is set to 82 MHz and the pump to probe intensity ratio 2:1 is chosen to optimize the signal. We use a triple modulation scheme, where the helicity of the pump beam is modulated at $f=50$ kHz, and the intensities of pump and probe are modulated at low frequencies $f_{pump}=147$ Hz and $f_{probe}=176$ Hz, respectively. The signal is detected at frequencies $f\pm(f_{pump}+f_{probe})$ where it is less affected by the low-frequency noise. To minimize the inhomogeneous excitation effects the laser beams are weakly focused onto a spot of $\sim$200-$\mu$m diameter. The sample is immersed in a bath of superfluid helium at 1.8 K.

The sample under scrutiny is an iodine modulation-doped CdMnTe/CdMgTe QW grown on GaAs substrate with a thick CdTe buffer layer. The details on the structure and optical properties of similar samples may be found elsewhere.[5] The QW electron-density modulation in the range of $10^{10}$–$10^{11}$ cm$^{-2}$ is obtained by changing the thickness of the iodine doping layer along a fixed direction in the plane of the sample. The residual electron density $10^{10}$ cm$^{-2}$ was estimated from photoluminescence experiments using the method proposed in Ref. 6. The QW width is 80 Å and the effective Mn concentration $x_{eff}=0.42\%$ was deduced from the electron-spin splitting of the conduction band. Spin domains are observed in the QW irrespective of the doping level and follow the same general trends. Most of the results reported hereafter are obtained on the nominally undoped piece of the sample.

Figure 1 shows a series of spectra, obtained by Fourier

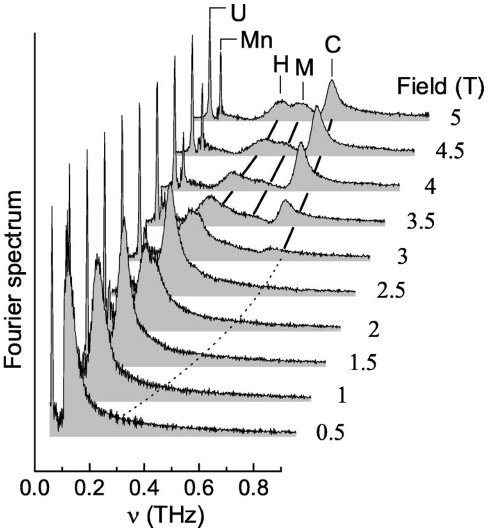

FIG. 1. Fast Fourier transform spectra of the time-resolved magneto-optical Kerr signal at 1.8 K. The intensity of line U is divided by a factor 5. The lines H, M, and C are assigned to electron spins precessing in spatial regions of the QW with different Mn spin temperatures (see text). The lines visualizing their evolution are guides to the eye.

transformation of time-resolved Kerr rotation measurements. The spectra reveal the precession frequencies of the spins optically created by the pump pulse. The photon energy and excitation density are set to 1.644 eV and 15 W/cm$^{-2}$, respectively,[7] and the in-plane magnetic field ranges from 0.5 to 5 T. At low magnetic field intensity the spectra exhibit only two lines denoted as U and H in Fig. 1. As the field increases a third line labeled Mn grows progressively in between. Finally, above 3 T two new lines labeled as M and C appear.

First we discuss the lowest frequency lines U and Mn. They shift linearly as the field increases, with $g$-factor absolute values 1.45 and 2, respectively. The line labeled Mn corresponds to the free precession of the Mn spins in the external field. It results from coherent rotation in the exchange field induced by the spin polarized photocreated holes.[8] The intensity of this line is expected to grow with the magnetization as far as the magnetic-field intensity is lower than 5 T.[9] This behavior is, indeed, observed (cf. Fig. 1). The origin of the line U is less obvious. At the excitation wavelength used, the major part of the pump beam not absorbed in the QW is absorbed in the thick CdTe buffer layer and eventually creates spin polarized electrons. However, the measured $g$ factor does not match the $g$-factor value for bulk CdTe equal to $-1.65$.[10] Therefore we attribute this line to the two-dimensional electron gas at the CdTe/CdMgTe interface. These interface electrons are expected to have a slightly reduced $g$ factor compared to the bulk CdTe due to the penetration of their wave function in the barrier material with a lower $g$ factor.[11]

Unlike the features discussed above, the high-frequency lines H, M, and C shift nonlinearly with magnetic field. This is illustrated in Fig. 2(a), where the frequencies of these lines are plotted as a function of the magnetic field. The nonlinear field dependence of lines C and H is typical for electron-spin splittings in the exchange field originating from Mn magnetization.[12] These splittings are proportional to the magnetization and usually described for the nearly paramagnetic Mn spin system by the modified Brillouin function $h\nu(B,T)=h\nu_s B_S[g\mu_b SB/k_b(T+T_0)]$. Here $\nu(B,T)$ is the field- and temperature-dependent Larmor frequency of the electrons, $S=5/2$ is Mn spin, $\nu_s=N_0\alpha x_{eff}S/h=0.550$ THz is the Larmor frequency at saturation of the magnetization, $N_0\alpha=0.22$ eV is the $s$-$d$ exchange integral for CdMnTe, and $h$ is the Planck constant. $T$ is the Mn spin temperature and $T_0=0.2$ K accounts for antiferromagnetic interactions between Mn spins. The fit of the experimental data with the modified Brillouin function gives the Mn spin temperature of $T=8.8$ K for the line H. Line C resides at $\nu_s$ value. Therefore it arises from spatial regions with Mn spin temperature very close to the helium bath ($\sim 1.8$ K). Moreover, its frequency is very close to the electron frequencies obtained at low excitation density where no Mn spin heating is expected [open triangles in Fig. 2(a)] so that both data sets are well fitted by the Brillouin function at 2 K. The line M seems to have a common origin with the lines C and H, since the total intensity of these three lines does not vary with magnetic field. However, the interpretation of this line goes beyond the simple Brillouin function description, suggesting that it corresponds to spatial areas with field-dependent spin temperature. This line was not systematically observed, so that, hereafter, we mainly concentrate on the lines C and H.

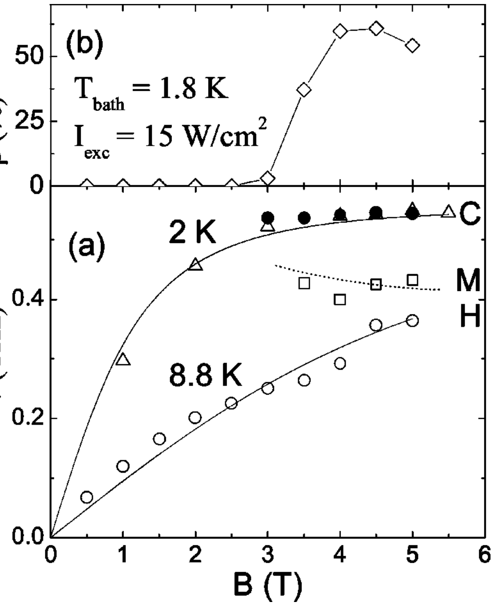

FIG. 2. (a) Frequencies of the lines H (open circles), M (open squares), and C (filled circles), extracted from Fig. 1, as a function of the field intensity. Open triangles show the electron-spin precession frequency under low excitation density when no spin instability occurs. Solid lines are fits of the data with a modified Brillouin function, and the dotted line is a guide for the evolution of the line M. (b) The spectral weight of the line C normalized to the total spectral weight of lines H, M, and C. It is proportional to the total area occupied by the cold domains.

The coexistence of two kinds of Mn ions with well defined temperatures is an essential physical result. It suggests the formation of Mn temperature spatial patterns with characteristic size exceeding the electron wave-function extension. Thus we conclude on the formation of hot and cold Mn spin domains characterized by two distinct temperatures. The formation of cold domains is triggered by the field above the threshold value of 3 T [see Fig. 2(a)], revealing the pitchforklike bifurcation point.[1] In other words we are dealing with the dynamic phase separation controlled by the external magnetic field. In this regard we speculate that the line M may be related to normally metastable domains stabilized by the nonmagnetic static disorder, e.g., interface roughness. The possible instable behavior of the spin temperature was pointed out by Tyazhlov *et al.*[3] In spite of essentially different experimental conditions, the origin of the instability is similar. Namely, it comes from the positive feedback loop, in which Mn spins are heated more efficiently in hotter regions, via mutual spin flips with electron spins.[13] This mechanism is indeed operative in our system, because the diffusion of the electrons in the excited spin state towards hot regions is energetically favored due to the reduced exchange splitting in hotter regions. The magnetic field threshold cannot be deduced from this simple reasoning, and was not addressed in Ref. 3. However, such critical behavior is typical for dynamic systems undergoing a bifurcation.

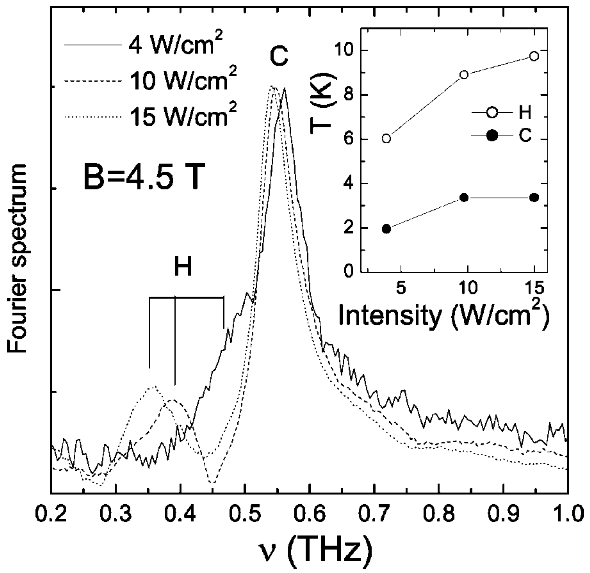

FIG. 3. Fourier spectra of the time-resolved magneto-optical Kerr effect under excitation densities 4 W/cm$^2$ (solid line), 10 W/cm$^2$ (dashed line), and 15 W/cm$^2$ (dotted line). The inset gives the temperatures of hot (H) and cold (C) spin domains, as estimated from the fits of the electron Larmor frequencies with the modified Brillouin function.

The next issue is the determination of the total area of each type of domain. It is based on the comparison between the weights of spectral features arising from cold and hot domains. Indeed, the weight of the line in the Fourier spectra is proportional to the number of precessing electron spins in the corresponding domain type. As a result, supposing that electrons are uniformly distributed in the QW, the weight of the spectral line appears to be a reliable measure of the total domain area.[14] Figure 2(b) shows the ratio $\rho$ of the spectral weight of the high-frequency line over the total spectral weight of lines H, M, and C. One can see that above the threshold for domain formation, at about 3 T, the cold domains grow rapidly until they reach the area comparable to that of hot domains at about 4-T field intensity.

Finally, we discuss the effect of the optical excitation density and the photon energy on the domain formation. These parameters control the excited carrier concentration and kinetic energy in the QW and thus are expected to play a crucial role in the distribution of Mn spin temperature. Figure 3 shows the Fourier spectra obtained at fixed photon energy 1.644 eV for three different excitation densities. The corresponding measurements were done on a piece of the sample with a doping level about $7\times10^{10}$ cm$^{-2}$ at a field of 4.5 T, that is slightly above the threshold field of 4 T for this doping level, photon energy, and the excitation density 15 W/cm$^2$.[15] While at the highest excitation density the two electron precession frequencies are well defined, at 4 W/cm$^2$ the low-frequency precession appears only as a shoulder of the high-frequency line. Therefore the excitation density threshold for domain formation slightly below 4 W/cm$^2$ seems to exist under the present experimental conditions. Thus the instability evolves even under rather low excitation density. This is essentially due to very slow Mn spin-lattice relaxation at low temperature and low concentration[16] and probably implies a cumulative effect of many pulses. It is interesting to mention that $\rho$ does not change substantially with excitation density. It means that hot domains do not grow, while their temperature increases to accommodate the power increase (see inset of Fig. 3). We recall that all the experiments discussed were done at photon energy of 1.644 eV, that is at 5 and 8 meV above the neutral and charged exciton resonances at zero field, respectively. A reduction of the photon energy by only 6 meV is shown to be enough to restore an homogeneous phase with an intermediate temperature. Thus the excess kinetic energy of the photocreated excitons does favor the domain formation and the thresholds discussed above depend on this excess energy.

In conclusion, our time-resolved experiments reveal a different example of pattern forming system, that is magnetic ion spins in DMS QW's under ultrafast excitation in magnetic field. We believe that the formation of hot and cold spin domains is an instability, originating from the kinetics of electron-spin relaxation and diffusion in the exchange field created by magnetic ions. The three main parameters controlling the instability are identified to be the magnetic field, the excitation density, and the photon energy. Upon the variation of these control parameters the spin system exhibits a bifurcation, so that the homogeneous spin temperature of magnetic ions is replaced by the domains at two distinct temperatures occupying well defined spatial areas. The determination of the domain sizes, as well as their eventual correlation with the static potential fluctuations due to the interface roughness, remain a challenge to be tackled by resonant Rayleigh scattering experiments.[17] In longer run a systematic study of the phase separation in the vicinity of the bifurcation point should allow the determination of the universality class for this instability.

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[14] The nonuniformities in the electron distribution may result from localization in potential fluctuations. The fluctuations of the exchange potential due to the domain formation do not provoke accumulation of precessing electrons in hot domains, because as long as the electron spin precesses it does not experience these fluctuations. However, the proposed method for domain area determination does not apply in the particular case where exchange potential variations and potential modulation due to the QW interfaces roughness are correlated.

[15] We note that in this sample the line M was not observed.

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