Dynamic Spin Organization in Dilute Magnetic Systems

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Low-temperature time-resolved Faraday-rotation measurements in $Cd_{1-x}Mn_xTe$ allow direct observation of the time evolution of the interaction between an electron spin and the magnetic moments of dilute impurity ions. The experiment displays real-time formation and evolution of magnetically organized states suggestive of the bound magnetic polaron.

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One of the many interesting problems in elementary magnetism concerns the fundamental exchange interaction of an electron with the spins of localized magnetic neighbors. Such a system can be made available for study by carefully diluting a normal semiconductor with magnetic ions, producing the so-called "dilute magnetic semiconductor." Recent studies of these materials[1] have stimulated interest in local magnetic phenomena which have been explained with the concept of the bound magnetic polaron,[2] where electrons or holes localized at impurity sites interact with the dilute magnetic ions. It has been suggested that a local ordering of the magnetic moments of these ions would arise from the Heisenberg exchange between the spin of the charged carrier and the paramagnetic ions within its Bohr orbit. This exchange interaction creates the dramatic spin-dependent effects observed experimentally, such as very large Zeeman splittings and giant dc Faraday rotations. Moreover, this "spin-ball" has a diameter which varies as a function of binding energy, and provides a unique opportunity to study microscopic spin organization.

The dynamics of polaron binding were first studied by Harris and Nurmikko[3] through transmission-modulated spectroscopy. However, in order to witness dynamically the formation and evolution of bound magnetic polarons, it is useful to employ a magnetically sensitive probe along with a high-speed time-resolved detection scheme. Consequently, we have recently developed new techniques to measure time-dependent magnetization on picosecond time scales. These involve the exploiting of the Faraday effect as a probe of the magnetic state of the system and use of optical fibers to conduct tunable laser pulses into high magnetic fields at low temperatures. We have chosen to study $Cd_{1-x}Mn_xTe$ because its ease of crystal fabrication, extensive characterization, direct optical gap, and cubic structure result in an isotropic sample with favorable optical properties. Studies of this material have shown that the dominant subgap transitions are due to acceptor bound excitons.[1] A complete description of the experimental technique will be given elsewhere,[4] but the key features are described below.

The experimental configuration for time-resolved Faraday rotation relies on ultrashort optical pulses from a synchronously pumped, wavelength-tunable dye laser. Pumped by an argon ion laser, the dye laser produces pulses of 5 psec duration, widely tunable within the visible region of the spectrum. The pulse train of the dye laser is split into a strong pump beam and a much weaker probe beam. The pump pulses promote approximately 10^{14}-cm^{-3} bound excitons in the sample and the probe pulses, when suitably delayed, monitor the time evolution of the Faraday rotation in this crystal after the initial excitation.

The $Cd_{1-x}Mn_xTe$ samples are cooled and kept at a temperature adjustable between 1 and 100 K in the field of a superconducting magnet providing fields of up to 6 T. Both the pump and probe light are guided into the cryostat and delivered to the sample by a single-mode polarization-preserving optical fiber. Their polarization is collinear and aligned with one of the principal axes of the fiber, and no substantial broadening of the optical pulses was measured at the sample. The sample is sandwiched between two polarizers whose axes are aligned at 45° with respect to one another to insure maximum detection sensitivity. The time evolution of this Faraday gate is monitored by our collecting the transmitted probe beam in a larger-core optical fiber and recording it as a function of delay between pump and probe pulses. A phase-sensitive detection scheme allows us to detect only the influence of the strong pump pulse on the weaker probe pulse in the transmitted light, thus selecting only the transient behavior of the Faraday rotation.

The samples were grown by the Bridgman method and have been characterized in several ways. Clear crystals were selected by examining the samples with both visible and infrared microscopes. The structure of the material was confirmed by a Gandolfi camera and an x-ray diffraction measurement. Finally, low-field SQUID magnetic susceptibility measurements did not detect any magnetically ordered phases within our temperature range and served as an additional check of the manganese concentration. The samples were typically 0.25 mm^2 and 100 μm thick.

The spectral position of the bound exciton was determined for each of our samples by cw luminescence, and the pump energy was adjusted to coincide with the high-energy side of this luminescence band.

A characteristic set of curves representing the magnitude of the Faraday rotation versus time for several different fields is shown in Fig. 1. Each curve represents the average of 100 scans where an individual point is the average of approximately 10^8 laser pulses. We verified experimentally that the observed signal vanishes for photon energies less than that of the bound exciton, and becomes weaker for higher pump energies. This confirms that we are indeed witnessing the time evolution of bound excitons in these samples.

The sharp feature at $t=0$ arises from a coherent coupling between the copolarized pump and probe pulses, and has been studied in detail by other authors.[5] Although very useful as a timing mark, the coherent artifact simply maps the autocorrelation function of the optical pulses and does not contain much physical information about the system response. Our primary interest focuses on the fast (~ 200 psec) rise and subsequent slower decay of the signal that is clearly seen to vary as a function of field (Fig. 1) and manganese concentration (Fig. 2).

Several characteristic features should be pointed out. This signal vanishes in the absence of an applied field, and thus we conclude that we are indeed monitoring an induced rotation of the plane of polarization by Faraday rotation and not either a partial bleaching of the absorption or an optically induced birefringence. This is further emphasized by the overall shape of the response which starts at almost zero amplitude, evolves towards a maximum, and subsequently dies away. We would expect an induced bleaching response to be maximum initially and then decay. These data support the concept of a bound magnetic polaron which evolves from the exciton-manganese spin system after the instantaneous creation of an exciton, in contrast with a response from the excitons themselves. The decrease of the initial rise time with lower manganese concentration and the total absence of this behavior in pure CdTe both underline the crucial role of the manganese atomic spins in this process. As a final experimental check, the observed effects disappear completely at higher temperatures ($T > 10$ K).

In the following discussion we describe a qualitative model for the transient Faraday rotation which accounts for our observations. A giant, strongly temperature-dependent, static Faraday rotation has been previously observed[6] whose spectral dependence is of the form $\theta(\omega) \simeq \omega^2/(\omega_0^2-\omega^2)^2$, where ω_0 is the band-gap energy. This can be derived from the well-established band-splitting model[7] with the experimentally determined exchange parameters[7,8] for $Cd_{1-x}Mn_xTe$. Following Heiman,[9] the dielectric constant is expressed as a sum of oscillators,[10] and given $n^2=\epsilon$, one obtains for the Faraday rotation per unit length

$$\theta=\frac{\omega}{2c}[\Delta n_+-\Delta n_-]=\frac{\pi}{2n\lambda}[\Delta\epsilon_+-\Delta\epsilon_-]$$
$$=\frac{-\pi\omega_p^2}{n\lambda}\left\{\sum_{j+}\frac{f_{j+}\omega_j\Delta\omega_j}{[\omega_j^2-\omega^2]^2}-\sum_{j-}\frac{f_{j-}\omega_j\Delta\omega_j}{[\omega_j^2-\omega^2]^2}\right\}, \quad (1)$$

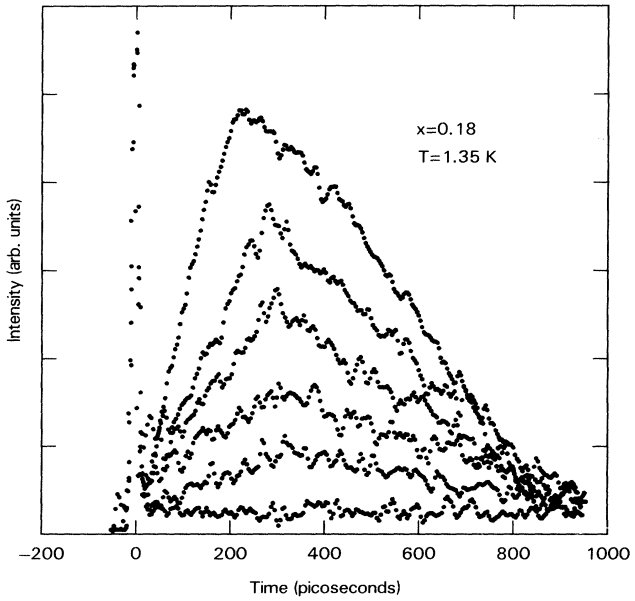

FIG. 1. Time-resolved Faraday rotation with $E_{pump}=1.839$ eV, for $B=0, 1, 2, 3, 4, 5$ T shown in ascending order.

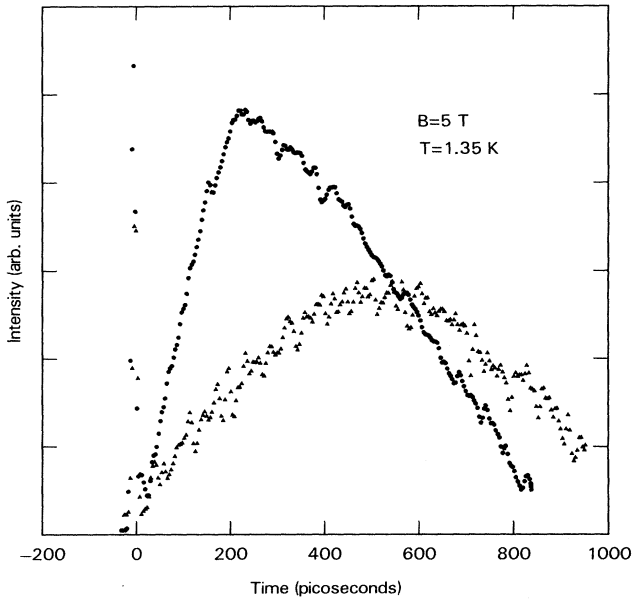

FIG. 2. Time-resolved Faraday rotation for $x=0.18$ (circles) and $x=0.115$ (triangles). $E_{pump}=1.839$ and 1.754 eV, respectively.

where + and − indicate right and left circular polarizations, n is the refractive index, ω_p the plasma frequency, f the oscillator strength, and λ the excitation wavelength. $\Delta\omega_{j\pm}$ represents the energy splittings with applied magnetic field for the respective polarizations from the oscillator resonance ω_j. In order for the probe pulse to sample the changes in magnetization from the pump, it is assumed that a substantial overlap exists between the two wave functions.

Gaj *et al.*[7,8] have calculated the field splittings of the exciton absorption which give rise to the dominant magneto-optic effects, and have evaluated the splitting parameters for $Cd_{1-x}Mn_xTe$ using polarized reflectivity. These results show that in the presence of an applied field, four transitions (two for each circular polarization) arise between the valence-band–derived fourfold-degenerate ground state ($J=\frac{3}{2}$) and the conduction-band-derived twofold-degenerate excited state ($J=\frac{1}{2}$). These energies are

$$E_1^{\pm} = E_0 \pm \tfrac{1}{2}(\alpha-\beta)N_0x\langle S_z\rangle, \tag{2a}$$

$$E_2^{\pm} = E_0 \mp \tfrac{1}{2}(\alpha+\beta/3)N_0x\langle S_z\rangle, \tag{2b}$$

where E_0 is the unperturbed exciton energy, α and β are the exchange integrals for conduction and valence electrons ($\alpha/\beta=-0.25$), N_0 is the number of cations per unit volume, x is the Mn^{2+} concentration, and $\langle S_z\rangle$ is the thermal average of Mn^{2+} spins along the applied field. Substitution of Eqs. (2a) and (2b) in Eq. (1) then yields the important Faraday result that

$$\theta = \frac{-19}{18}\frac{\pi}{n\lambda}\frac{E_p^2E_0}{(E_0^2-E^2)^2}f\beta N_0x\langle S_z\rangle, \tag{3}$$

where $E_p=\hbar\omega_p$.

In light of Eq. (3), clearly all first-order time dependences of θ are contained in the time dependence of $\langle S_z\rangle$. However, in order to describe the local character of the evolution of the Mn^{2+} polarization about the acceptor bound exciton, one must consider the time-dependent deviations from the local background after the excitation pulse. The present experiments were performed in magnetic fields between 1 and 5 T. Therefore, an estimate of the background magnetization $\langle S_z\rangle$ due to the (paramagnetic) Mn^{2+} spins may be obtained from the data of Gaj *et al.*[8] This yields a background polarization $\langle S_z\rangle/S_{sat}=0.28$ and 0.70 for 1 and 5 T, respectively, for $x=0.18$. In order to estimate the final polarization after the pump pulse near the bound exciton, we use the recent results of spin-flip Raman scattering[11] together with polarized luminescence from the donor-acceptor pair recombination.[12,13] In Ref. 11 the electron polarization is found to be 100% in fields larger than 0.2 T. On recombination with unpolarized holes this would lead to 50% luminescence polarization, p. The fact that $p\simeq 0.6$[12,13] implies that the hole polarization is extremely small.

From the perspective of our experiment, the significance of these results is twofold: (1) The hole and therefore local Mn^{2+} polarization effected by the hole-ion exchange is small compared to the background, and (2) the effect of the pump pulse is to create polarons which reduce the local magnetization from the background value. Furthermore, this difference becomes more pronounced the higher the magnetic field. The net time-dependent change in magnetization is then observed by the probe pulse through the relation for θ of Eq. (3).

Of the time scales in the problem, the most rudimentary are the creation of acceptor bound excitons, the organization of the Mn^{2+} spins within the Bohr radius of the exciton (τ_s), and the recombination time of the exciton (τ_r), either radiatively or otherwise. The exciton creation time is very fast, less than 20 psec,[14] and may be ignored. τ_s is reflected in the rise of the signal (Fig. 1) and in $Cd_{1-x}Mn_xSe$ has been estimated to be $\simeq 400$ psec,[3] whereas τ_r determines the signal decay rate. The latter has been measured for the present $Cd_{1-x}Mn_xTe$ samples by time-resolved luminescence to be of order 1000 psec.[4] The gross features of Fig. 1, i.e., the initial rise peaking at 200–400 psec and loss of signal after approximately 10^{-9} sec, are understood in terms of τ_s and τ_r.

The rise of the curves in Fig. 1 can be described by an initial slope

$$\langle S(t)\rangle = [\langle S_z\rangle - \langle S_z\rangle_p](1/\tau_s), \tag{4}$$

where $\langle S_z\rangle$ is the unperturbed average magnetization

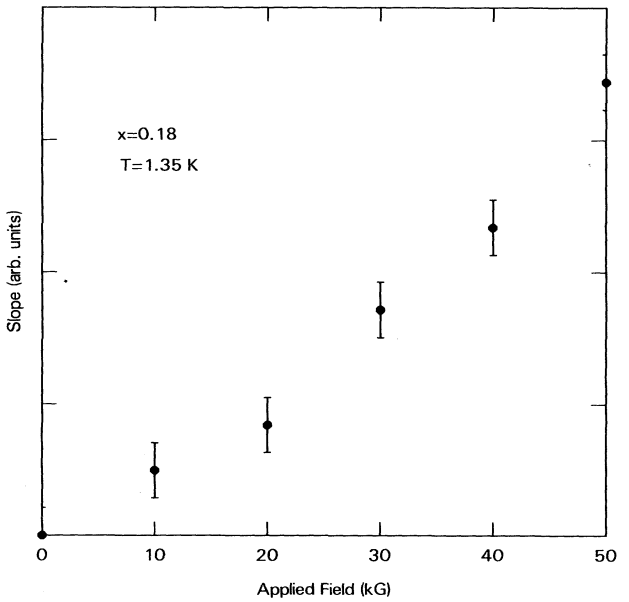

FIG. 3. Field-dependent slopes of the rise of the Faraday data.

and $\langle S_z \rangle_p$ the equilibrium magnetization in the presence of infinite-lived bound excitons. We assume that $\langle S_z \rangle_p$ is less than $\langle S_z \rangle$ because the luminescence polarization saturates at $p \simeq 0.6$,[12,13] most of which can be ascribed solely to the electron polarization. Hence the hole is not well polarized, and its presence therefore decreases $\langle S_z \rangle$ in its neighborhood. It is clear that Eq. (4) then explains the major features of the data. For $B = 0$, no net magnetization is created by the pump. In finite fields greater than 0.5 T, $\langle S_z \rangle$ increases, whereas $\langle S_z \rangle_p$ remains approximately constant. Consequently, the slope increases proportionally to the difference. The observed weak superlinear dependence of this slope on applied field (Fig. 3), however, suggests that τ_s is also a function of B. This conclusion is supported by the fact that τ_s is longer for less concentrated samples (Fig. 2). It is, furthermore, clear why the effect vanishes for $T > 10$ K. Thermal fluctuations frustrate the magnetic interactions[15] and no substantial difference between the magnetization terms in Eq. (4) remains.

In summary, we have studied the dynamics of spin organization in the dilute magnetic semiconductor $Cd_{1-x}Mn_xTe$. The new technique of time-resolved Faraday rotation provides a unique probe of the local magnetic interactions within this system. We observe a characteristic time for spin organization of $\simeq 300$ psec, with a clear dependence on magnetic dilution. A phenomenological model based upon local deviations from the average magnetization within a sphere containing an excited carrier and its surrounding Mn^{2+} spins is successful in explaining the salient features of the observed effects.

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