Dynamics of Spin Organization in Diluted Magnetic Semiconductors

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It is shown that spin-spin interactions rather than spin-lattice coupling control the dynamics of the magnetization formation visible in time-resolved luminescence and SQUID magnetometry of bulk and layered diluted magnetic semiconductors. A numerical solution of a nonlinear Schrödinger equation is applied to find the relation between formation time of the exciton magnetic polaron and relaxation time of the spin subsystem.

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A rapid progress in the application of ultrashort light pulses for the monitoring of dynamical processes has made it possible to trace the emerging and vanishing of polarization of the localized spins after optical injection of carriers across the band gap in bulk [1–7] and layered [8–11] diluted magnetic semiconductors (DMS). These studies have been concerned with the important question of dynamics of spin organization and relaxation in mutually coupled systems of effective mass electrons and localized spins in different semiconductor matrices as well as for various electronic and magnetic confinement and magnetic dilution. Since the pioneering work of Harris and Nurmikko [1], in which the magnetic portion of the bound exciton energy was studied in $Cd_{0.9}Mn_{0.1}Se$ through time resolved transmission-modulated spectroscopy, two other methods have been successfully employed to witness spin organization in DMS in the absence of an external magnetic field. In the first one, the energetic position of the photoluminescence maximum [2–6,10,11] or the degree of its circular polarization [12] have been measured as a function of time. In the second method, developed and brought up to remarkable sensitivity by Awschalom and co-workers [7–9], a SQUID sensor has been employed in a pump-probe scheme to observe the temporal evolution of sample magnetization under the influence of spin-polarized carriers, instantaneously created by circularly polarized optical pulses. The above studies, together with their static counterpart [13–18], have led to a number of important findings. In particular, it is by now generally accepted that during the first few picoseconds of their life, the excitons created by the above-band-gap excitation are trapped by localized states associated with defects or with alloy and/or magnetic fluctuations. Simultaneously, the spin orientation of the photocarriers is being lost as a result of the spin exchange with magnetic ions [7,12,17,18], spin-orbit coupling [18], or electron-hole interaction [12]. The exciton history develops further in the time scale of hundreds of picoseconds and involves three competing processes [4–6,10,11,16]: (i) tunneling between localized states, (ii) radiative or nonradiative recombination, and (iii) formation, via sp-d exchange interaction, of a cloud of ferromagnetically aligned localized spins in the exciton vicinity. Such a complex, known as the exciton magnetic polaron (EMP), results in the lowering of the exciton recombination energy and shrinking of its localization volume [19]. Actually, the process of polaron formation can be made dominating in the time-resolved experiments by the so-called selective excitation, i.e., by tuning the pump to the region of localized exciton states, where the low density of states precludes migration of the excitons [4–6,10,11,16] and dephasing of their spins [7,14,18].

The above scenario leaves, however, a number of issues open. It is unclear, in particular, what is the dominant mechanism that breaks the conservation of spin momentum and makes the appearance of local or macroscopic spin alignment possible. Is the modulation of the magnetic cluster energy by lattice vibrations important in this context [7,9]? What is the role played by interactions among localized spins [1,18,20]? Is the spin transfer from the carriers [7,17] or spin diffusion from the polaron vicinity [3,21–23] relevant? Can the isothermal magnetic susceptibility be used for the description of this system far from thermal equilibrium [20]? Moreover, the existing theoretical works on the dynamics of polaron formation [21–23] have suggested the lack of any simple relation between the polaron formation time τ_f and the microscopic time characterizing the dynamics of the localized spins τ_S. For instance, according to Spałek [21] τ_f of 400 ps [1] implies τ_S of 50 ns, while in the model of Kavokin *et al.* [22] τ_f of 250 ps [9] corresponds to τ_S of 40 ps.

In this Letter, we discuss arguments suggesting that the formation of magnetic polarons by spin diffusion contributes little to the process. We then present a numerical solution of a nonlinear Schrödinger equation with a time-dependent magnetic potential well and employ it to determine quantitatively the relation between τ_f and τ_S. We also explain why the isothermal susceptibility characterizes the system despite that the spin-spin rather than spin-lattice coupling sets the relevant time scale for spin organization and relaxation. Finally, we show that results of our work allow a consistent description of time-resolved data for DMS quantum structures.

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Consider a carrier trapped at $t = 0$ in a localized state. Its adiabatic wave function is described by a Schrödinger equation which contains (i) kinetic energy, taken here in the one-band effective-mass approximation, (ii) potential energy that includes the localizing and confining potential, and (iii) a nonlinear term $\hat{H}_M = \hat{H}_M(\mathbf{r}, t)$, which describes the exchange interaction with the time-dependent polarization of the surrounding localized spins,

$$\hat{H}_M = \frac{J}{2g\mu_B} \int d\mathbf{r}' \int dt' \, G(\mathbf{r} - \mathbf{r}', t - t') M_0[H^*(\mathbf{r}', t')]. \quad (1)$$

Here $J = \alpha$ or β is the electron- or hole-magnetic ion exchange integral, while g and $M_0(H)$ denote the Landé factor and the equilibrium magnetization of the localized spins, respectively. The molecular field H^* produced by the carrier and experienced by the ionic spins appears at $t = 0$ and is then given by

$$H^*(\mathbf{r}, t) = -J|\psi(\mathbf{r}, t)|^2/2g\mu_B . \quad (2)$$

Finally, $G(\mathbf{r}, t)$ is the response function of the spins, whose Fourier transform—if the spin dynamics can be described by Bloch equations with the diffusion term included—assumes a familiar form,

$$G(\mathbf{q}, \omega) = \frac{Dq^2 + \tau_S^{-1}}{Dq^2 + \tau_S^{-1} - i\omega} . \quad (3)$$

We shall now present arguments suggesting that spin diffusion in the form described by Eq. (3) is of minor importance *vis à vis* spin relaxation in the dynamics of EMP.

For a while, we neglect the time dependence of the exciton localization radius a. We then find that at a given diffusion coefficient D, the diffusion operates for $a^2/4D < t < \tau_S$ and in this time domain leads to a power-law dependence of the EMP energy E_p on t, $E_p(t) - E_p(\infty) \sim t^{-3/2}$. Thus, the diffusion model is in conflict with the experimental findings [1–6,10,11], as they point to the exponential dependence of E_p on t.

Furthermore, following Awschalom, Warnock, and von Molnar [7] we note that the diffusion conserves the total magnetic moment m, and, therefore, changes of the local magnetization by such a process are not visible in the time-resolved magnetometry. Indeed, under assumption that the spin density of the photocarriers decays as $\exp(-t/\tau_e)$ and $M_0(H^*)$ is in the linear regime (no saturation), we obtain $m(t)$ to be proportional to

$$m(t) \sim [\exp(-t/\tau_S) - \exp(-t/\tau_e)]\tau_e/(\tau_S - \tau_e). \quad (4)$$

This formula has been fitted to the photomagnetization data [7–9], and the resulting values of τ_S will be presented later on (see Figs. 1 and 2). We only note here that they fall in the time domain of hundreds of picoseconds. Thus, they are of the same order as τ_f deduced from time-resolved luminescence [4–6,10,11], confirming a nondominant contribution from the diffusion in the latter measurements.

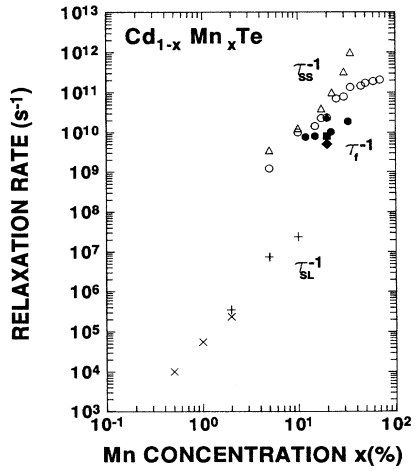

FIG. 1. Spin-spin relaxation rate (empty circles [27] and triangles [28]) and spin-lattice relaxation rate at 5 K (crosses [20], pluses [26]) compared to polaron formation rate (full triangles [4], squares [5], circles [6], and diamonds [7]).

In order to determine the influence the shrinking of the carrier wave function accompanying the polaron formation may have on its dynamics, we have solved the Schrödinger equation taking the nonlinear history-dependent term (1) into account with $D = 0$. The Kimball-Shortley algorithm [24] has been adopted with the 0.5 Å path. Our method gives access to the important time range $t \leq \tau_S$, inaccessible in the analytical model of Kavokin *et al.* [22], and therefore provides quantitative information on the relation between τ_S and τ_f, a time constant characterizing $E_p(t)$.

Since the exciton energetics is dominated by the hole, we take $m^* = 0.71m_0$. To simulate the effect caused by the unknown localizing potential, the Coulomb

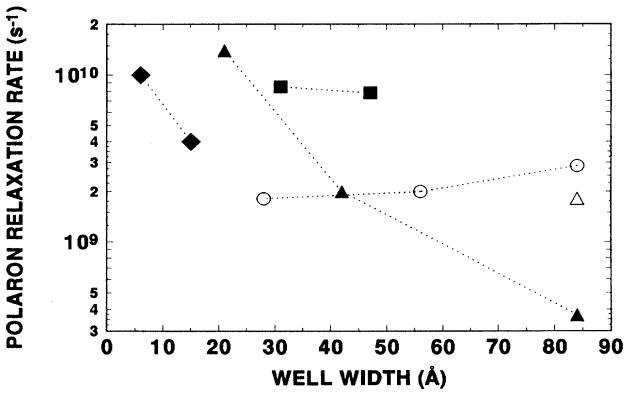

FIG. 2. Polaron rates as a function of the quantum well width for structures with nonmagnetic and magnetic barriers—empty and full symbols, respectively (empty circles $Cd_{0.77}Mn_{0.23}Se/ZnTe$ [9] and triangles $Cd_{0.87}Mn_{0.13}Se/ZnTe$ [9]); full diamonds $CdTe/Cd_{0.78}Mn_{0.22}Te$ [10], triangles $Cd_{0.93}Mn_{0.07}Te/Cd_{0.62}Mn_{0.38}Te$ [8], and squares $Cd_{0.86}Mn_{0.14}Te/Cd_{0.59}Mn_{0.41}Te$ [11]).

attraction by the electron, and the lattice polarization we have supplemented the Hamiltonian with a square well potential. This potential was assumed to have either spherical or cylindrical symmetry for bulk and layered structures, respectively, and its radius and depth were treated as adjustable parameters to fit the polaron energy at $t \rightarrow \infty$. Other parameters characterizing the most thoroughly studied system (Cd,Mn)Te are already known to a good accuracy [25].

The results of our computations of $E_p(t)$ compared to the time dependence of the luminescence Stokes shift, as measured by Mackh *et al.* [6] for $Cd_{0.85}Mn_{0.15}Te$ at 1.8 K, are presented in Fig. 3(a). As shown, τ_S = 70 ps leads to τ_f = 125 ps observed experimentally. A similar calculation has been extended for EMP in quantum structures. Figures 3(b) and 3(c) depict a comparison between experimental and theoretical results for two

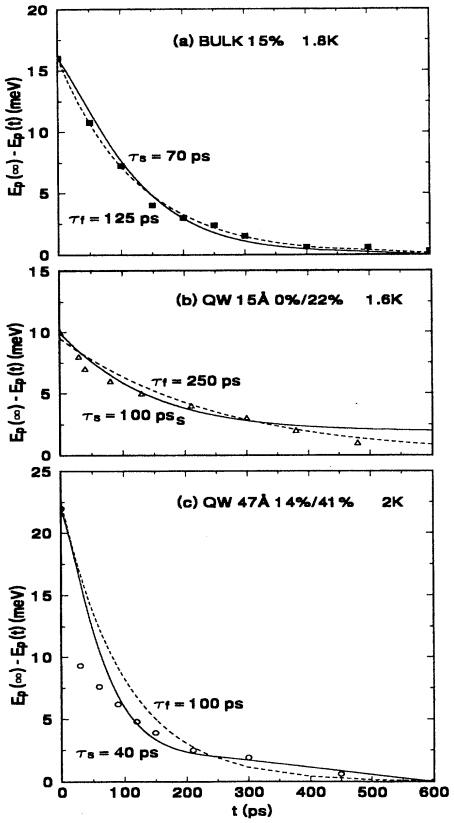

FIG. 3. Determination of polaron formation time τ_f and spin-relaxation time τ_S by fitting a simple exponential function (dashed lines) or solutions of the Schrödinger equation (solid lines) to the experimental time dependences of the Stokes shift of exciton luminescence: (a) $Cd_{0.85}Mn_{0.15}Te$ [6], (b) $CdTe/Cd_{0.78}Mn_{0.22}Te$ [10], and (c) $Cd_{0.86}Mn_{0.14}Te/Cd_{0.59}Mn_{0.41}Te$ [11]. Initial depth and radius of the three- or two-dimensional localizing well is 15 meV, 35 Å; 15 meV, 30 Å; 20 meV, 25 Å for solid lines in (a), (b), and (c), respectively.

extreme physical situations in the (Cd,Mn)Te system—for EMP residing in nonmagnetic [10] and magnetic [11] quantum wells, respectively. The results presented in Fig. 3(c) suggest that in the case of the structures which consist of the different magnetic layers, the description of the data by a single value of τ_S is an oversimplification. In particular, the evaluated magnitude of τ_S has a meaning of an average value over the relevant magnetic layers and interface regions with the weights determined by the hole envelope function.

In general, τ_f/τ_S is independent of τ_S and is greater the larger is the variation of the carrier wave function during the polaron formation. At the same time, as long as the dependence $M_0(H^*)$ is linear, the magnitude of the magnetic moment m is insensitive to the carrier wave function, and thus $\tau_S \approx \tau_f$ in the case of $m(t)$.

Having determined that in the studied systems τ_f is no more than two to three times greater than τ_S we turn to the identification of microscopic mechanisms of spin relaxation that control the dynamics of EMP formation. As shown in Fig. 1, the experimentally obtained [20,26] spin-lattice relaxation rate τ_{SL}^{-1} is by more than 2 orders of magnitude too slow to lead to a picosecond time scale of EMP formation in DMS. At the same time, τ_S^{-1} resulting from τ_f agrees very well with the spin-spin relaxation rate τ_{SS}^{-1} deduced from the EPR studies [27,28]. This appears to be rather striking as the EMP formation involves both spin and *energy* relaxation. On the other hand, such a conclusion is compatible with the previous findings [20,29], showing that the time response of the Faraday effect to the high-frequency magnetic field is described by τ_{SS}, not by τ_{SL}. In order to explain why it could be so we recall that there is no spin response to the external field for $t < \tau_{SL}$ if spin-spin interactions are of the Heisenberg form, so that the magnetic moment is a constant of motion and $\tau_{SS}^{-1} = 0$. If, however, the nonscalar part of the spin-spin interactions is strong enough, a nonzero magnetization can be formed adiabatically already at $\tau_{SL} > t \geq \tau_{SS}$. The corresponding change of the spin temperature and the magnitude of the resulting magnetization $M_{ad} = \chi_{ad} H^*$ result from the condition that for any adiabatic process the work done by the magnetic field is equal to the change of the internal energy of the spin subsystem [20,30]. This leads to

$$\chi_{ad} = \chi - T(H^* \partial\chi/\partial T)^2/2c_S . \quad (5)$$

Taking H^* = 10 kOe, the values for the spin heat capacity c_S, and the isothermal magnetic susceptibility $\chi(T)$ suitable for $Cd_{0.85}Mn_{0.15}Te$ at 2 K [25,31], we get $(\chi - \chi_{ad})/\chi = 5\%$, which corresponds to the increase of the spin temperature by 0.25 K.

Finally, we turn to the experimentally observed [8–11] dependence of τ_f^{-1} on L_W, the width of the quantum well, shown in Fig. 2. We suggest that in addition to a slight increase of τ_f/τ_{SS} with L_W, the dependence $\tau_f(L_W)$ results

from a rather strong decrease of τ_{SS}^{-1} with the lowering of Mn concentration x visible in Fig. 1 as well as from the obvious fact that the penetration of the exciton into the barriers decreases with increasing L_W. As we have already noted, in the case of the layered structures τ_f^{-1} represents an average value over the relevant *magnetic* layers, the interface acting as an additional layer of an intermediate Mn concentration [25]. Thus, τ_f^{-1} decreases with increasing L_W if the Mn concentration in the well is smaller than in the barriers and increases otherwise. Actually, the former should be much stronger, especially for nonmagnetic or weakly magnetic wells. Experimental results summarized in Fig. 2 follow these expectations.

In summary, our phenomenological description of the recent experimental results [4–11] demonstrates that the relaxation of the photocarrier energy associated with spin alignment occurs adiabatically at the expense of the interaction among the localized spins. This is in contrast to the ordinary thermalization process which proceeds via the coupling to phonons. Our analysis indicates that the conservation of the spin momentum is broken by strong nonscalar spin-spin interactions which shift the spin response time down to picosecond range. While the proposed model is consistent with the results for layered structures, a quantitative description of the data must await further progress in control and understanding of the magnetic properties of the interfaces.

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