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FORMATION TIME OF MAGNETIC POLARONS*

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It is shown that non-scalar spin-spin interactions rather than spin-lattice coupling or spin diffusion control the dynamics of the magnetization formation visible in time-resolved luminescence and SQUID magnetometry.

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Progress in the application of short light pulses for monitoring of dynamical processes has made it possible to trace the emerging of exciton magnetic polarons (EMP) after optical injection of carriers across the band gap in diluted magnetic semiconductors (DMS) [1-6]. In this paper we discuss mechanisms which might account for the fast dynamics of the polaron formation in DMS.

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, (iii) a nonlinear term $\hat{H}_M = \hat{H}_M(r, t)$, which describes the exchange interaction with the time-dependent polarization of the surrounding localized spins,

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

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

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

Finally, $G(r, t)$ is the response function of the spins. As a starting point we assume that it can be described by Bloch equations with the diffusion term included.

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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 $a^2/4D < t < \tau_S$, where τ_S is the spin relaxation time. In this time domain the diffusion 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 [7] is in conflict with the experimental findings [1–4], as they point to the exponential dependence of E_p on t .

In order to determine the influence the shrinking of the carrier wave function accompanying the polaron formation may have on its dynamics we solved the Schrödinger equation taking the nonlinear history-dependent term (1) into account with $D = 0$. Our method gives access to the important time range $t \leq \tau_S$, inaccessible in the analytical model of Kavokin et al. [8], and therefore provides quantitative information on the relation between τ_S and τ_f , a time characterizing the exponential dependence $E_p(t)$.

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 we included a square well potential with the radius and depth treated as adjustable parameters. Other parameters characterizing the most thoroughly studied system $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ are already known to a good accuracy [9]. The results of our computations of $E_p(t)$

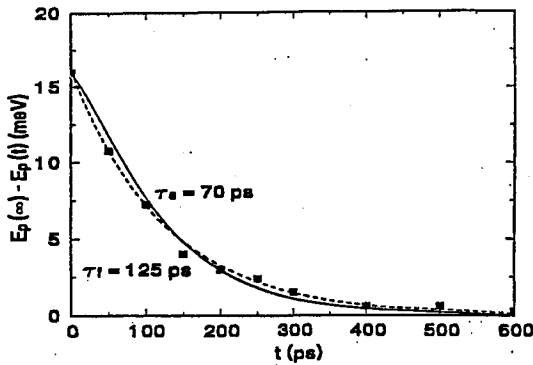


Fig. 1. Determination of polaron formation time τ_f and spin-relaxation time τ_S by fitting a simple exponential function (dashed lines) or solutions of the Schrödinger equation (solid lines) to the experimental time dependencies of the Stokes shift of exciton luminescence in $\text{Cd}_{0.85}\text{Mn}_{0.15}\text{Te}$ [4]. Initial depth and radius of the localizing well is 15 meV and 35 Å, respectively.

compared to the time dependence of the luminescence Stokes shift, as measured by Mackh et al. [4] for $\text{Cd}_{0.85}\text{Mn}_{0.15}\text{Te}$ at 1.8 K, are presented in Fig. 1. As shown, the solution of the Schrödinger equation with $\tau_S = 70$ ps leads to an almost exponential dependence $E_p(t)$ characterized by a decay constant of $\tau_f = 125$ ps.

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 which control the dynamics of EMP formation. As shown in Fig. 2, the experimentally obtained [10, 11] spin-lattice relaxation rate τ_{SL}^{-1} is by

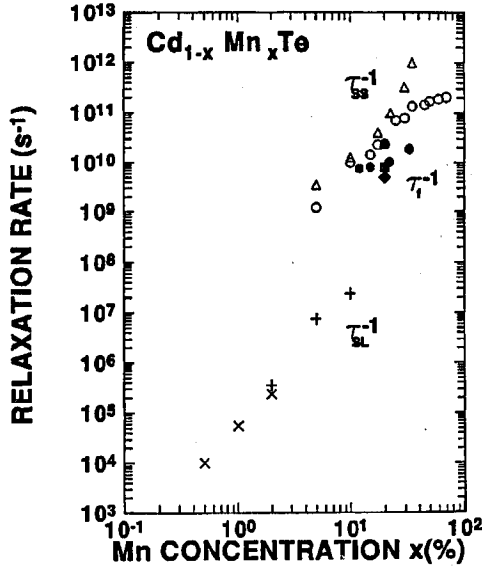


Fig. 2. Spin-spin relaxation rate (empty circles [13] and triangles [14]) and spin-lattice relaxation rate at 5 K (crosses [10], pluses [11]) compared to polaron formation rate (full triangle [2], square [3], circles [4], and diamond [5]).

more than two 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_l agrees very well with the spin-spin relaxation rate τ_{ss}^{-1} deduced from the EPR studies [12, 13]. 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 [10, 14], 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 non-scalar part of the spin-spin interactions is strong enough, a non-zero magnetization can be formed adiabatically already at $\tau_{sl} > t \geq \tau_{ss}$. The magnitude of the resulting magnetization $M_{ad} = \chi_{ad}H^*$ is given by 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 [10, 15]. This leads to

$$\chi_{ad} = \chi - T(H^* \partial \chi / \partial T)^2 / 2c_s. \quad (3)$$

Taking $H^* = 10$ kOe and the values for the spin heat capacity c_s and the isothermal magnetic susceptibility $\chi(T)$ suitable for $\text{Cd}_{0.85}\text{Mn}_{0.15}\text{Te}$ at 2 K [9, 16] we get $(\chi - \chi_{ad})/\chi = 5\%$, which corresponds to the increase in the spin temperature by 0.25 K.

In summary, our analysis implies that the magnetization induced by a burst of photocarriers is formed adiabatically in time of the order of τ_{SS} . Moreover, for an appropriately large heat capacitance of the spin reservoir, the magnitude of that magnetization is close to its isothermal value. Thus, the relaxation of the photocarrier energy associated with spin alignment occurs at the expense of the interaction among the localized spins, the conservation of the spin momentum being broken by strong non-scalar spin-spin interactions which shift the spin response time down to picosecond range. This fast dynamics constitutes an important property of DMS in view of their foreseen application in optoelectronic devices.

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