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ANISOTROPIC BOUND MAGNETIC POLARON IN $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$

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Abstract. – Bound magnetic polarons (BMP) resulting from exchange coupling between Mn^{++} ion spins and anisotropic hole of an acceptor in hexagonal CdMnSe are studied through time resolved donor-acceptor luminescence. A crystal-field model is used to fit the temperature dependent binding energy and the potential barrier for orientational relaxation.

Experiments

Exchange interaction between magnetic ions and weakly bound carriers creates BMP in semimagnetic semiconductors [1]. In CdMnSe , although Mn^{++} spins have essentially isotropic magnetic properties, when they couple to a Γ_9 hole at the top of the valence band of this wurtzite type crystal, a very anisotropic $A^\circ\text{BMP}$ is anticipated, with a strong tendency to align its total magnetic moment along the c axis.

As done previously in CdMnTe [2], $A^\circ\text{BMP}$ effect in $\text{Cd}_{0.95}\text{Mn}_{0.05}\text{Se}$ was demonstrated through the temperature dependence of its binding energy E_b . It was deduced from the study of donor-acceptor (DA) recombination, in a high sensitivity time resolved measurement (see Fig. 1).

Time evolution of $A^\circ\text{BMP}$ orientation was studied through the time-dependent circular polarization P_c of DA emission for propagation $\parallel c$ under two different conditions: I) action of a magnetic field $B \parallel c$ with an unpolarized pump above band-gap [3]; II) optical pumping with σ^+ light in the tail of A° to conduction band absorption at zero field [4].

Figure 2 shows the time evolution of P_c under conditions I). The slowing down of P_c evolution when T decreases reflects the dynamics of $A^\circ\text{BMP}$ orientation, as explained in the model below. The fact that P_c increases with time, for $T > 5$ K indicates that the degree of initial $A^\circ\text{BMP}$ orientation P_0 is smaller than its equilibrium value P_{th} . Indeed, our sample being n type, A° is created by capture of a free hole by A^- , since we pump above the band gap. In the box around A^- , the N spins undergo magnetic fluctuations when the field is already present and one can evaluate the probability P_z that the fluctuation points along the field direction z . It looks as if $P_0 = P_z$, which means that BMP develops its magnetic moment along the z component of the magnetic fluctuation present at the time of hole capture, whatever its angular momentum. Since P_{th} results from the orientation by the same field of a BMP near saturation, then it is clear that $P_{th} > P_z = P_0$.

Optical pumping, which works through a site selection process [4], provides at best a value $P_c \sim 8\%$

Fig. 1. – Polaron binding energy as a function of temperature for acceptors in $\text{Cd}_{0.95}\text{Mn}_{0.05}\text{Se}$. The curve is the result of theory and the circles, the experimental data.

Fig. 2. – Time evolution of circular polarization P_c of DA luminescence in $\text{Cd}_{0.95}\text{Mn}_{0.05}\text{Se}$ at difference temperatures, for a constant ratio $B/T \sim 0.1$ Tesla K^{-1} .

which stays constant for more than $5 \mu\text{s}$ at 1.8 K. This is another proof that $A^\circ\text{BMP}$ orientation is totally frozen at low T . At the same T , under conditions I) one observes a decrease of P_c with time delay: this is a qualitative proof of the influence of donor spin polarization P_e upon P_c . When $P_e \neq 0$ as under conditions I), a spin dependent recombination process induces the decrease of P_c with time delay.

Theoretical model and discussion

A simple model for anisotropic $A^\circ\text{BMP}$ was obtained [5] by adding a uniaxial crystal-field term to the spherical Heisenberg model previously used for cubic crystals. The bound hole is treated as a parti-

cle of total angular momentum $\mathbf{j} = 3/2$. The separation between the A band ($j_z = \pm 3/2$) and B band ($j_z = \pm 1/2$) is given by the uniaxial term. The effective Hamiltonian is thus written:

$$H = -\mathbf{J} \cdot \sum_i \mathbf{S}_i - D(j_z^2 - 9/4) - \mu_B g_{Mn} B \sum_i S_{iz} - \mu_B g_h B j_z. \quad (1)$$

Here, $\mathbf{S}_i$ denotes the spin ($S = 5/2$) of the Mn^{++} ion at the site $\mathbf{R}_i$ and $b_i = |\varphi(\mathbf{R}_i)|^2$, where $\varphi(\mathbf{R})$ is the envelope function of the acceptor ground state. The hole- Mn^{++} exchange parameter is $J = -\beta/3$ in the usual notation; $D = 11.5$ meV according to experiment [6] in CdSe. The magnetic field B is applied along the c axis. Note that we neglect Mn-Mn AF exchange interaction, which is much smaller than the polaron coupling within the hole orbit.

H is then reduced to an exactly soluble model by making a finite-radius constant-coupling approximation. Defining $\sigma = \sum_{i=1}^N \mathbf{S}_i$, where N is the number

of Mn^{++} spins equally coupled to the acceptor, we note that σ and $M = \sigma_z + j_z$ are good quantum numbers. Here, $\sigma = NS, NS-1, \dots, 0$ (assuming even N). The energy levels $\varepsilon_{\sigma M \ell}$ are obtained by diagonalizing H in the subspace $\{|\sigma, \sigma_z = M - m \rangle | j, j_z = m \rangle\}$. The curve in figure 1 shows the theoretical zero-field polaron binding energy $E = -\langle \varepsilon \rangle$ as a function of temperature. The only free parameter for obtaining this fit to our experimental data is $N_0 \beta = -0.81$ eV, which is 73 % of the free hole value [7]. This reduction is about the same as that predicted for CdMnTe [8]. Other parameters were the acceptor binding energy, 100 meV in CdSe leading to a Bohr radius of 8 Å, giving $N \sim 12$ Mn^{++} ions.

As pointed out in reference [3], both E and equilibrium polarization can be interpreted at low temperature within the Ising approximation, which neglects the B -like ($j_z = \pm 1/2$) states ($D \rightarrow \infty$). The most interesting aspect of the crystal-field model is that it provides a framework for discussing the time resolved polarization measurements that revealed a relatively long relaxation time (Fig. 2), suggesting a thermally activated behaviour. Indeed, the ground state of $A^\circ \text{BMP}$ in zero field is doubly degenerate: $M = \pm(NS - 3/2)$ corresponding to orientation parallel or antiparallel of the c axis. Relaxation induces transitions in the $\{\sigma, M, 1\}$ space, but presumably with selection rules like $\delta M = \pm 1$ or ± 2 ; as a consequence, BMP orientation has to be understood as a multi-step process of similar nature as BMP formation. If a potential barrier exists between the two ground states, the orientational relaxation rate is given by: $1/\tau = (1/\tau_0) \exp(-E_a/k_B T)$ where τ_0 can be identified with the BMP formation time and the activation en-

Fig. 3. - Energy levels in the crystal field model for $\sigma = NS$ as a function of $M = \sigma_z + j_z$. The lower branch exhibits the anisotropy energy K .

ergy E_a in zero field is the anisotropy energy K . Now, K at $T = 0$ can be calculated as follows. Let us fix $\sigma = NS$ and plot the eigenvalues ε as a function of M (see Fig. 3). The lowest of the four branches thus obtained represents the energy of the polaron at saturation as a function of its orientation with respect to the c axis. As expected, it is a symmetric curve with a maximum at $M = 0$. The difference between this maximum and the ground-state gives $K = 15$ meV, with the parameters used. Let us add that for $\sigma < NS$, the corresponding lower branch stays above. K should decrease with increasing T and B .

Experimental data indicate a broad distribution of E_a , centered around 10 meV. The width may be related to alloy disorder. Thus, the simple uniaxial model provides a reasonable interpretation of the activation energy. τ_0 was found of the order of 10^{-9} sec.

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