

Optical Studies of Mn²⁺ Spin Resonance in CdMnTe Quantum Wells

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The optical properties of diluted magnetic semiconductors and their quantum well structures have been widely investigated from the viewpoint of the modification of the electronic states induced by the presence of magnetic ions. The reciprocal effect of the influence of electronic states on magnetic ions has been less studied, particularly in the case of quantum well structures for which the small number of magnetic ions makes them difficult to be detected in traditional EPR or magnetisation measurements. In this paper we report the investigation of Mn²⁺ magnetic resonance in CdMnTe quantum well structures by means of purely optical methods: resonant Raman scattering and optically detected magnetic resonance. Both methods show that the classical description of Mn²⁺ ions in terms of isolated paramagnetic $S = 5/2$ spins is inadequate in these systems, in spite of the low concentrations of Mn²⁺.

We have studied a single quantum well structure consisting of a modulation doped CdMnTe magnetic well between nonmagnetic barriers of CdMgTe. The sample was characterised by magneto-photoluminescence, photoluminescence excitation and Raman scattering studies. A Ti:Sapphire tuneable laser was used for excitation, with a power density kept below $1 \mu\text{W}/\text{cm}^2$. The low temperature of the sample was confirmed by the Mn²⁺ spin temperature, calculated from the anti-Stokes to Stokes line intensity ratio to be $T=1.7\text{K}$. The collected light was analysed in a double-grating spectrometer. For the optically detected resonance (ODMR) measurements changes in the photoluminescence spectrum were monitored as the sample was irradiated with microwaves and the magnetic field slowly swept through resonance. In Raman scattering experiments, we observe resonant spin-flip transitions of Mn²⁺ ions and of conduction band electrons. The magnetic field dependence of the electron spin-flip Raman shift value allowed us to evaluate the value of Mn²⁺ ion concentration in the Cd_{1-x}Mn_xTe quantum well as $x = 0.54\%$.

We observe multiple spin-flip transitions of the Mn²⁺ ions, with a number of transitions greater than 5 in both Voigt and Faraday backscattering geometry in external magnetic fields up to 28 T (see Figure 1). In the Faraday geometry we used circularly polarised, $\sigma+$ or $\sigma-$, incident and collected light. The multiple spin-flip signals are observed within the energy range of the exciton high-energy spin state branch. Thus, an out-going resonance was deduced as a necessary condition for this effect. Independent of light polarisation no signal was observed on the low-energy exciton branch. In contrast, in the Voigt configuration, where the effect is generally much stronger and more lines can be seen, multiple spin-flip transitions are observed on both exciton branches.

The observation of multiple Mn²⁺ spin-flip transitions in the Voigt geometry is in agreement with previous reports [1,2]. The multiple spin-flips were explained as an indication of the creation of a collective magnetic moment of several Mn²⁺ spins, formed within the extent of the photo-created exciton wave function as the result of the hole-Mn²⁺ ion exchange interaction. However, the existence of multiple spin-flip signals in the Faraday configuration was not predicted by the above theory. Surprisingly enough, we observe multiple transitions also in this configuration. Furthermore, the maximum peak intensities of consecutive peaks, defined as the maximum observed intensity of one peak for all excitation energies, is well described by an exponential function. This differs from the Poisson distribution reported in [1] for the Voigt geometry.

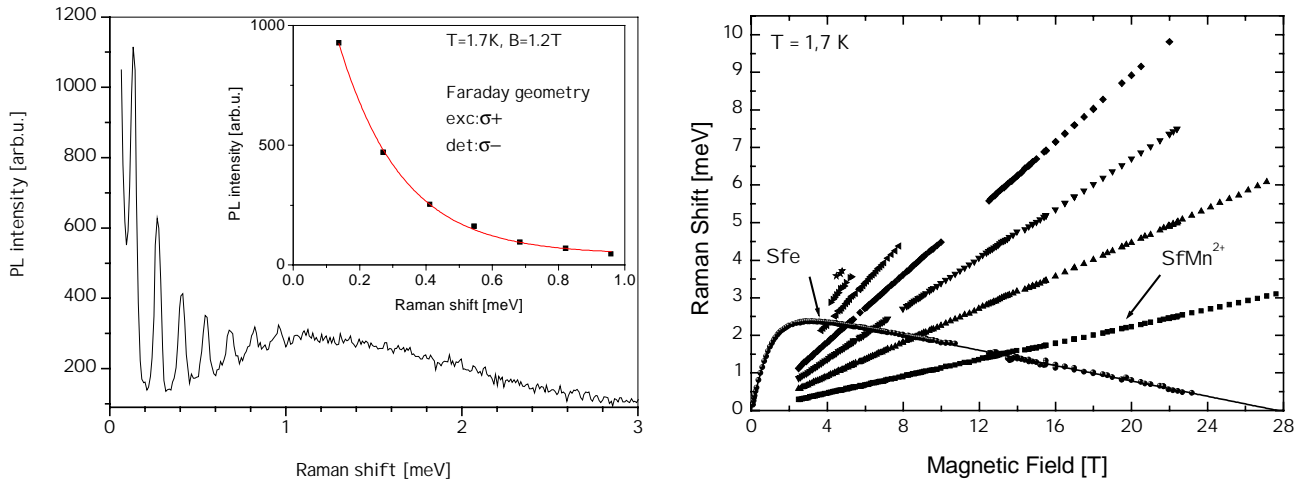


Figure 1. Left figure shows multiple Mn²⁺ spin flips (n=7) for the Faraday geometry. The exponential function describes consecutive peak intensity (inset). Right figure shows the magnetic field dependence of Raman shift of Mn²⁺ ions and electronic spin-flip.

The ODMR measurements showed a significant blue shift of the photoluminescence spectrum at the magnetic field corresponding to Mn²⁺ spin transitions at the energy of the incident microwaves (see Figure 2). This blue shift, when plotted against the magnetic field, exhibits a complicated structure, suggestive of a hyperfine splitting due to interactions between Mn²⁺ electronic and nuclear spins. However, their number was greater than the expected number of hyperfine transitions (6).

Together, these observations clearly demonstrate that the behaviour of Mn²⁺ ions in a quantum well requires a description taking into account the exchange interactions with free carriers, and cannot be considered as simple paramagnetic spin 5/2 ions.

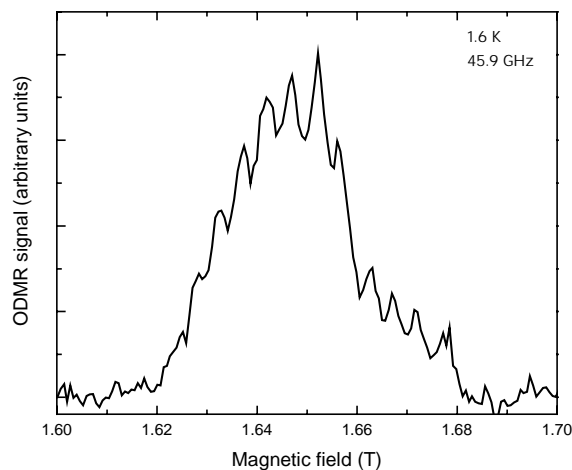


Figure 2. Optically detected magnetic resonance spectrum. Note the multiple lines superimposed on the main structure.

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