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SCATTERING DYNAMICS OF FREE EXCITONS ON Fe^{++} IONS IN $Cd_{1-x}Fe_xSe$

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Inelastic scattering of excitons on Fe^{++} ions in $Cd_{1-x}Fe_xSe$ was studied by resonant Raman scattering. Polarization measurements were done using a modulation technique, allowing for the first time to determine the full polarization state of the detected light. The obtained results were compared to a simple calculation in an incoherent model of scattering on Fe^{++} ions.

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1. Introduction

Raman scattering cascade involving intra-Fe^{++} excitations, reported first by D. Scalbert et al. [1] provides a powerful tool to study inelastic scattering of excitons on Fe^{++} ions. The first two excited states of the Fe^{++} ion in CdSe have symmetries A_2 and E and lie 12.8 cm^{-1} and 17.6 cm^{-1}, respectively, above the ground state A_1. The cascade consists of multiple Raman lines involving one or many excitations of the A_2 and E states.

We shall describe the cascade in terms of hot luminescence — creation of a free exciton, which undergoes a sequence of inelastic scatterings on Fe ions losing an energy of $\Delta E_{12} = E_{A2} - E_{A1}$ at each scattering, where E_{A1} and E_{A2} are respectively energies of the ground and the first excited state of the Fe^{++} ion [1]. The exciton states will be numbered by index $n = 0, 1, \ldots$

For each exciton state we introduce three time constants representing its scattering kinetics: τ^n — total lifetime, T_{2^n} — polarization relaxation time, T_{Fe^n} — time constant representing probability of inelastic scattering on Fe^{++} ions.

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2. Experiment

Raman scattering experiments were performed in a standard setup using a Coherent CR599 CW dye laser as an excitation source, a GDM 1000 double grating monochromator and a GaAs-cathode photomultiplier in photon counting regime.

Backscattering configuration was used with light propagation along the crystal hexagonal axis c. Measurements were done in magnetic field $\boldsymbol{B} \parallel c$. Polarization modulation was applied to the (linearly polarized) exciting beam using a photoelastic modulator followed by a Fresnel quarter-wave prism set horizontally (Fig. 1).

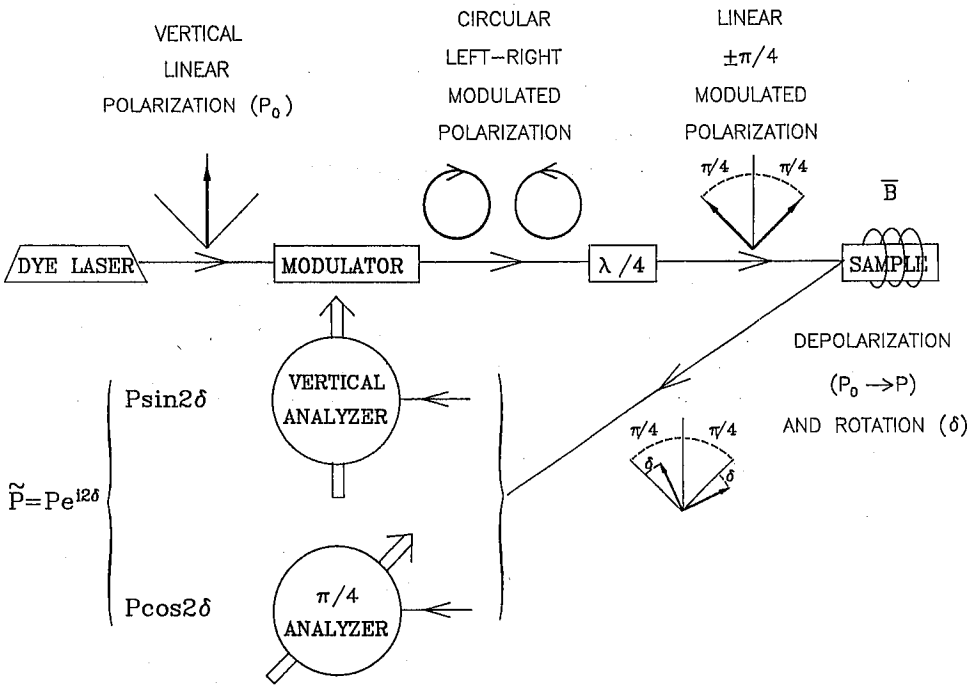

Fig. 1. Polarization measurements.

This setup produces linear polarization whose direction oscillates around the original polarization of the incident beam with the frequency and amplitude determined by the modulator (50 kHz and $\pm 45°$ in our case). A linear polarizer was placed in front of the monochromator. The photon counting system was triggered by a reference signal from the modulator producing two counts I_{45} and I_{-45} allowing to determine the polarization degree $P = (I_{45} - I_{-45})/(I_{45} + I_{-45})$ and the average intensity $(I_{45} + I_{-45})/2$. In order to distinguish between effects due to depolarization and rotation of polarization direction, the measurements were done for two positions of the linear polarizer: parallel or at 45° to the polarization of the incident laser beam. The polarization degree values measured for the two positions represent real and imaginary part of a complex polarization $\tilde{P} = P\exp(2i\delta)$ where δ is the angle of polarization rotation of the scattered light in the magnetic field.

3. Results and discussion

In order to determine the time constants involved we use the procedure outlined in Ref. [1], improved by indexing the time constants with replica order n:

1) Zero-field polarization degree $P^n(0)$ yields the ratio $T_2^n/(T_2^n + \tau^n) = P^n(0)/P^{n-1}(0)$.

2) Two LO-phonon replica of the original cascade (line intensities I_2^n) yields the ratio $\tau^{n+1}/T_{Fe}^n = I_2^{n+1}/I_2^n$.

3) Determination of the exciton Zeeman splitting $\hbar\omega_0$ corresponding to the half-width of the Lorentzian depolarization in the magnetic field allows to obtain the polarization lifetime $T = (1/\tau + 1/T_2)^{-1}$ from the condition $\omega_0 T^n = 1$. Ion–carrier exchange integrals from Ref. [2] were used.

Figure 2 shows results of polarization measurement for exciting light photon energy $\hbar\omega_{exc} = 14843\ cm^{-1}$ together with theoretical curves. While at low fields

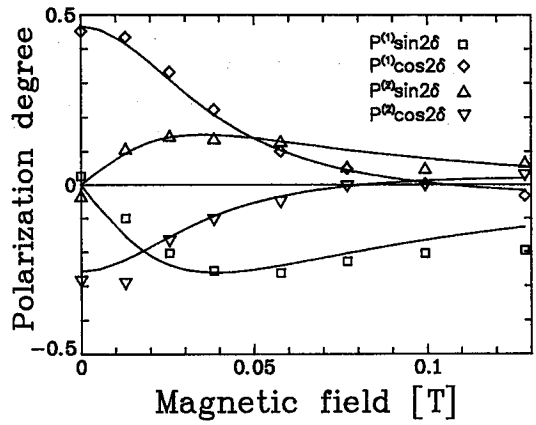

Fig. 2. Polarization of Raman lines in magnetic field for the first and the second scattering on Fe^{++}. Excitation photon energy $\hbar\omega_{exc} = 14843\ cm^{-1}$. Lines correspond to fits with Lorentzian expression.

the curves describe quite well the experimental data, discrepancies appear at higher fields. They probably originate from circular dichroism, not included in the model. The obtained values of time constants are summarized in Table, which contains also results for T_{Fe} calculated using a simple incoherent model of scattering governed by ion–carrier exchange described by a Heisenberg term [2]. We obtain a qualitative agreement supporting the presented physical image.

We point out that the method used in this work allows to measure very short time values due to the exchange enhancement of Zeeman splittings. It creates therefore perspectives in studies of other fast processes (e.g. phonon cascades) in DMS.

TABLE

Time constants obtained from fitting procedure described in text. All time constants are in ps.

$\hbar\omega_{exc} = 14843$ cm^{-1}				$\hbar\omega_{exc} = 14854$ cm^{-1}				
n	0	1	2	n	0	1	2	3
τ		1.8	1.6	τ		7	4	6
T_2		1.8	2	T_2		10	8	5
T_{Fe}		2	20	T_{Fe}		6.5	40	
$T_{Fe^{th}}$	8	21		$T_{Fe^{th}}$	6	8		

Acknowledgment

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