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## Study of impurity states in magnetic semiconductors by multi-quantum spin-echo spectroscopy

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### 1. Introduction

The change of type and concentration of charge carriers in 3d-magnetic semiconductors is realized on the base of heterovalent doping by diamagnetic impurities. The doping causes the change of electrical properties such well as the modification of magnetic properties of these materials. For example, the magnetic crystallographic anisotropy constant and the temperature magnetic ordering increase with Ag<sup>1+</sup>-doping and decrease with In<sup>3+</sup>-doping in model magnetic semiconductor CdCr<sub>2</sub>Se<sub>4</sub>. To study the nature of the heterovalent doping on the microscopic level we have used the <sup>53</sup>Cr NMR in CdCr<sub>2</sub>Se<sub>4</sub>. Taken into account the quadrupole splitting of <sup>53</sup>Cr spectra we have applied the multi-quantum spin-echo spectroscopy method, which is a high resolution NMR method in magnets with quadrupole nuclei [1- 3].

### 2. Results and discussion

The spectra of multi-quantum echo  $V_{4\tau}$  in undoped and Ag<sup>1+</sup> and In<sup>3+</sup> doped crystals are different significantly. The spectra are broadened and shifted into high frequency range in CdCr<sub>2</sub>Se<sub>4</sub>:Ag crystals. The inhomogeneous broadening and low frequency shift take place in CdCr<sub>2</sub>Se<sub>4</sub>:In crystals. The temperature measurements of the main frequency maximum  $V_{4\tau}$  show that the 3/2-Bloch law is true for wide temperature interval. These measurements have been used to define the exchange stiffness constant A, which was found  $A = 3,35 \cdot 10^{-10}$  Oe/sm<sup>2</sup> in undoped crystals, and  $A = 4 \cdot 10^{-10}$  Oe/sm<sup>2</sup> in Cd<sub>1-x</sub>Ag<sub>x</sub>Cr<sub>2</sub>Se<sub>4</sub> (x=0,03) crystals. The growth of exchange stiffness constant correlates with the Curie temperature and show the increase of exchange interaction at Ag-doping. The CdCr<sub>2</sub>Se<sub>4</sub>:In crystals have opposite tendency.

At heterovalent doping the Cr-ions change his valence and impurity states (molecules) Cr<sup>4+</sup>- Ag<sup>1+</sup>- Cr<sup>3+</sup> and Cr<sup>2+</sup>- In<sup>3+</sup>-Cr<sup>3+</sup> are formed near impurity. The exchange and spin-orbital interactions inside such molecules are different from

similar interactions on the outside.

We have found that the speed of multi-quantum echo  $V_{4\tau}$  decay at 77 K is greater in CdCr<sub>2</sub>Se<sub>4</sub>:Ag than in CdCr<sub>2</sub>Se<sub>4</sub>:In. The oscillations of spin-echo amplitude  $V_{4\tau}$  have been detected as a result of detailed measurements of the dependences of echo-amplitude V on the delay time  $\tau$  between two excited pulses. On the base of oscillations period the quadrupole constants have been valued in doped crystals. The lower NMR Q-factor of Ag-molecules causes appropriate properties of electron systems of such molecules. Really, the measurements of the electron paramagnetic resonance linewidth shown that the linewidth in CdCr<sub>2</sub>Se<sub>4</sub>:Ag into 6 times greater than in CdCr<sub>2</sub>Se<sub>4</sub>:In at the close impurity concentration. This mean that the spin-orbital coupling in CdCr<sub>2</sub>Se<sub>4</sub>:Ag - molecules is greater than in CdCr<sub>2</sub>Se<sub>4</sub>:In - molecules .

### 3. Conclusion

The impurity states (molecules) appear as a result of heterovalent doping by diamagnetic impurities in the 3d magnetic semiconductors of CdCr<sub>2</sub>Se<sub>4</sub>. The Cr<sup>4+</sup>-Ag<sup>1+</sup>- Cr<sup>3+</sup> states (molecules) take place in Ag-doped crystals and Cr<sup>2+</sup>-In<sup>3+</sup>-Cr<sup>3+</sup> states (molecules) take place in In-doped crystals. The high exchange and spin-orbit interaction is observed for Ag-states with respect to the In-states.

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