TEMPERATURE DEPENDENCE OF THE ESR LINE INTENSITY IN $HgCr_2Se_{4-x}S_x$

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The temperature dependence of the ESR line intensity in $HgCr_2Se_{4-x}S_x$ ($x = 0, 0.25, 0.5, 1, 3, 3.5, 3.75, 4$) at the temperature range of $90 \div 300$ K was investigated. The anomalous increase of the intensity was observed for all samples. The anomalous increase can be explained by the gradual transition from the EPR phenomenon to the FMR phenomenon connected with the short-range magnetic ordering. For $HgCr_2S_4$ the further decrease of intensity with decreasing temperature is connected with a gradual creation of antiferromagnetic spiral magnetic ordering.

1. Introduction

The magnetic semiconductors $HgCr_2Se_{4-x}S_x$ have normal spinel structure [1]. $HgCr_2Se_4$ is a Heisenberg type ferromagnetic, having the Curie temperature 108—111 K [2, 3]. The compounds $HgCr_2Se_{4-x}S_x$ are ferromagnetics [3] for $x = 0, 0.25, 0.5$. The magnetic susceptibility for $HgCr_2S_4$ satisfies the Curie–Weiss law at high temperatures. The asymptotic Curie temperature $\theta = +105$ K. The positive and large value of θ gives evidence of predominance of ferromagnetic type interaction. The antiferromagnetic spiral type magnetic ordering exists at low temperatures. The temperature of phase transition $T_N = 36$—60 K [2, 4]. The uncertainity in T_C and T_N arises probably from the presence of impurities. The magnetic ordering in $HgCr_2Se_{4-x}S_x$ origins in the superexchange ferro and antiferromagnetic interactions.

The anomalous increase of line intensity was observed for $HgCr_2Se_4$ [7]. The purpose of this paper is to report the measured temperature dependences of line intensity in $HgCr_2Se_{4-x}S_x$.

2. Results of measurements

The ESR absorption spectra were measured with the RE-1301 X-band spectrometer in the temperature range 90 to 300 K. The measurements were carried for polycrystalline samples of $HgCr_2Se_{4-x}S_x$ with $x = 0, 0.25, 0.5, 1, 3, 3.5, 3.75, 4$. The ESR spectrum

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consisted of a single lorentzian shape line. The temperature broadening of line width arising from spin-lattice relaxation was observed for all samples [5]. The critical broadening of the line width near the Curie point was observed for compounds with $x = 0, 0.25, 0.5, 1$. This effect is weaker for remaining compounds with $x = 3, 3.5, 3.75, 4$.

At temperatures higher than $200 \div 240$ K the experimental dependences of intensities of ESR line $I = I(T)$ are consistent with a temperature dependence $I \sim T^{-1}$ (Fig. 2).

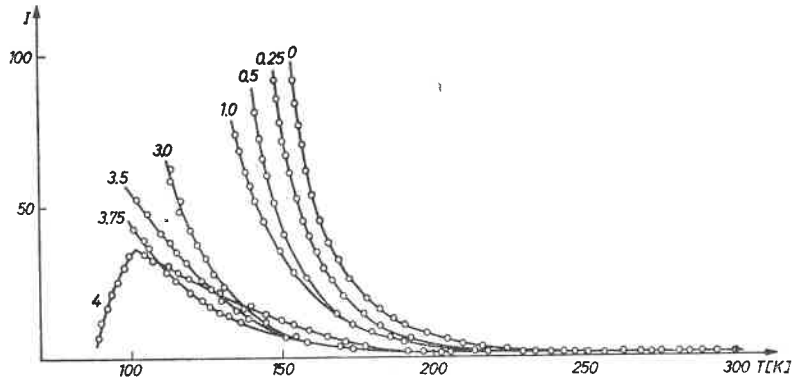

Fig. 1. Relative intensity of the ESR line as a function of temperature for $HgCr_2Se_{4-x}S_x$ with $x = 0$, 0.25, 0.5, 1.0, 3.0, 3.5. 3.75, 4.0 (the intensity at temperature 300 K is 1)

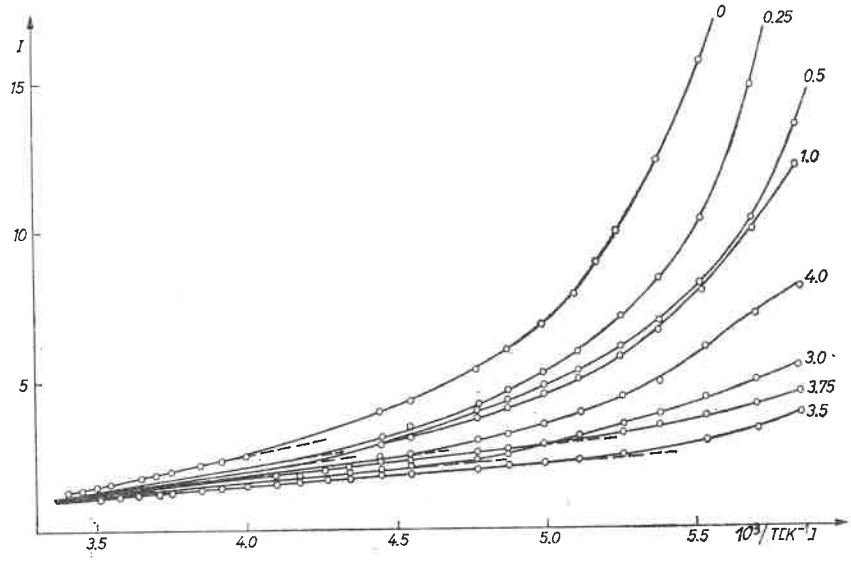

Fig. 2. Relative intensity of the ESR line as a function of inverse temperature for $HgCr_2Se_{4-x}S_x$

The intensities anomalously increase with further decreasing temperature (Fig. 1). The temperature dependence of $I(T)$ is different for $HgCr_2S_4$. At temperature range 300 K to about 100 K the increase of intensity is also observed. The rapid decrease of intensity to the zero value with decreasing temperature from 100 K to 90 K is observed (in the limit of spectrometer sensitivity).

3. *Discussion of results*

Theory of EPR predicts the following dependence of line intensity on temperature [6]:

$$I \sim T^{-1}.$$

The measured line intensities satisfy this dependence for all tested compounds at temperatures higher than 200—240 K. For this temperature range the condition $T \gtrsim 2T_C$ is satisfied, where T_C is the Curie temperature. The anomalous increase of I appears at temperatures $T \lesssim 2T_C$.

The anomalous increase of I can be connected with a gradual transition from the EPR phenomenon to the FMR phenomenon [7]. The increase of intensity is several times greater than increase of the static magnetic susceptibility (near the Curie point). There exists a short-range ordering [8] at the temperature range $T_C < T \lesssim 2T_C$. An increase of short-range ordering occurs with decreasing temperature. Then the motions of spins become correlated resulting in the coherent precession of the the total magnetic moment of the sample.

For $HgCr_2S_4$ the increase of ESR line intensity with decreasing temperature from 300 K to about 100 K can be also explained by increase of short-range ferromagnetic type ordering. The magnetic measurements give evidence of a predominance of ferromagnetic interaction at this temperature range [2].

The resonance frequency is very high for the spiral antiferromagnetic structure [8, 9]. Therefore the decrease of ESR line intensity (at the temperature range from 100 K to 90 K) can be explained by processes of creating the spiral antiferromagnetic type ordering. The decay of ESR line was observed for antiferromagnetic samples near the Neel point [11].

Also the roentgenographic investigations of $HgCr_2S_4$ [10] indicate some anomaly in the temperature dependence of a lattice constant at temperatures lower than about 100 K. This anomaly is connected with the existence of magnetic interactions of the non-ferromagnetic type.

4. *Conclusions*

The measurements of temperature dependences of the ESR line intensity make possible investigations of processes of creation of the magnetic ordering. The anomalous increase of the ESR line intensity with decreasing temperature can be connected with the ferromagnetic short-range ordering. The increase of a space range of magnetic ordering leads gradually to coherent precession of total magnetic moment of the sample.

The decrease of I for $HgCr_2S_4$ is connected with the process of creating the spiral antiferromagnetic type ordering.

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