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H-T PHASE DIAGRAM OF MAGNETIC SEMICONDUCTOR THIN FILMS — EXPERIMENT

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We used the irreversibilities between the field cooled and zero field cooled dc magnetization to determine the field and composition dependence of the spin-glass freezing temperature in $\text{CdCr}_2\text{Se}_4 : \text{In}$ (REE) and $\text{CdCr}_{2-2x}\text{In}_{2x}\text{Se}_4$ ($0.15 \leq x \leq 0.35$) (SG) thin films. The *H-T* phase diagram of samples with REE has two instability lines: the Gabay-Toulouse-type (G-T) and the De Almeida-Thouless-type (A-T) while samples in SG state are characterized by the A-T line. The A-T line of thin films was used for calculation the normalised internal magnetic field h_m of infinite spin clusters with long range ordering.

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Chalcogenide spinels of CdCr_2Se_4 exhibit specific properties that can be controlled by the dilution level. When the concentration of magnetic atoms is changed, the following magnetic phases are obtained:

(i) for $\text{CdCr}_2\text{Se}_4 : \text{In}$, the ferromagnetic state (FM) with the reentrant transition (REE). The system in microscopic scale is considered as consisting of an infinite ferromagnetic network (IFN) with long range ferromagnetic order and finite spin clusters (FSC) randomly distributed in IFN. The amount of IFN significantly dominates the FSC, in the volume of sample. The system has two instability lines at the *H-T* phase diagram: the De Almeida-Thouless (A-T) line [1], which indicates the onset of longitudinal freezing and Gabay-Toulouse (G-T) line for the transverse freezing [2].

(ii) for $\text{CdCr}_{2-2x}\text{In}_{2x}\text{Se}_4$ ($0 \ll x \leq 0.4$), spin-glass state. With increasing amount of indium the FSC dominates over IFN. This system exhibits the A-T instability line. This microscopic picture of both REE and SG develops the problem of the instability A-T and G-T lines [1-4]. Introducing the IFN, the internal magnetic field, related to IFN, has to be taken under consideration in the description of instability lines.

The disordered magnetic system is macroscopically characterised by:

(i) the temperature dependence of induced magnetization M , which includes the non-zero density of states in the energy gap [5],

(ii) the unidirectional magnetic anisotropy field H_{an} originated from the Dzaloshinsky–Moriya interaction (D–M).

Thin films of $\text{CdCr}_{2-2x}\text{In}_{2x}\text{Se}_4$ (for $0 \leq x \leq 0.4$) obtained by rf sputtering were investigated in the temperature range from 4 K to 200 K. The basic parameters $M(T)$ and $H_{an}(T)$, which classify the magnetic phase of samples, were determined from ferromagnetic resonance (FMR) [5] and SQUID data. The H_{an} was also determined from the displacement of $M-H$ loop. The hysteresis loops were measured after cooling the samples from 120 K to 4.2 K in the external magnetic field H_{FC} . The magnetic anisotropy induced by the field cooling is unidirectional with a single easy direction of magnetization parallel to H_{FC} [6]. For a chosen value of H_{FC} several cycles of loops were recorded, each loop up to different external magnetic field H_{max} . Asymmetrical loops were obtained until H_{max} has reached the value of H_{an} that was ascribed to the unidirectional magnetic anisotropy. A further increase in H_{max} produces the reversible, symmetrical $M-H$ loops. Figure 1 presents the two cycles of $M-H$ loops for thin films of $\text{CdCr}_{1.3}\text{In}_{0.7}\text{Se}_4$. The $M-H$ loop is asymmetrical for $H_{max} \leq 300$ Oe it means that $H_{an} = 300$ Oe. The freezing lines were found from the onset of irreversibilities between the field cooled (FC) and zero field cooled (ZFC) dc magnetization. We analysed the phase diagram (H_a-T) in terms of De Almeida–Thouless model [1] and of Gabay–Toulouse model [2, 4].

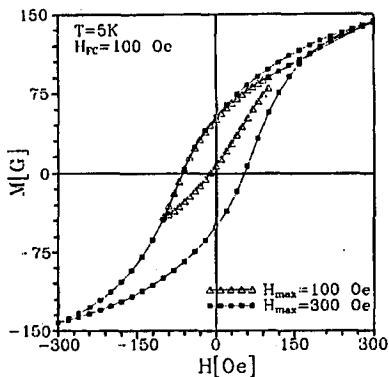


Fig. 1. $M-H$ loop for $\text{CdCr}_{1.7}\text{In}_{0.3}\text{Se}_4$ thin film taken at $T = 5$ K, $H_{FC} = 100$ Oe.

In our investigations we used the FC and ZFC dc magnetization measurements done by SQUID. The dc magnetization (M_{FC} and M_{ZFC}) as a function of temperature was measured for a set of external magnetic fields H_a applied within the film plane. On the base of these experimental data, for the fixed value of H_a , the irreversibility of magnetization ($\Delta M = M_{FC} - M_{ZFC}$) vs. temperature was obtained. Then H_a-T phase diagram was found. Depending on the samples composition the character of $\Delta M(T)$ curve exhibits one or two critical temperatures. Following the paper [7] we defined:

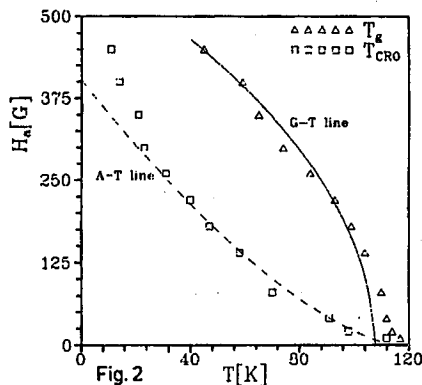


Fig. 2. H_a - T phase diagram of $\text{CdCr}_2\text{Se}_4:\text{In}$ thin film (REE).

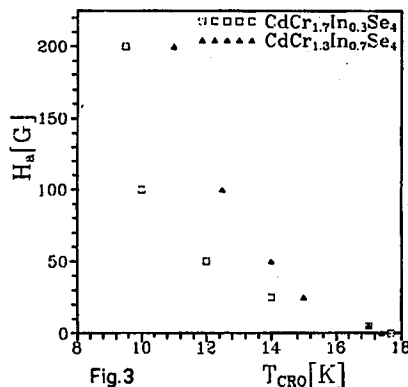


Fig. 3. H_a - T phase diagram of $\text{CdCr}_{1.7}\text{In}_{0.3}\text{Se}_4$ and $\text{CdCr}_{1.3}\text{In}_{0.7}\text{Se}_4$ thin films (SG).

- T_g as the temperature below which the first onset of irreversibilities is seen,
- T_{CRO} is the temperature below which ΔM increases relatively rapidly.

If only one critical temperature is present the A-T or G-T line can be established. When two critical temperatures are found, the phase diagram has the A-T and G-T lines [7]. Figure 2 presents the experimentally determined H_a - T curves for the thin films of $\text{CdCr}_2\text{Se}_4:\text{In}$ (REE). It is seen that above $T = 100$ K the character of G-T line changes to A-T line. Figure 3 shows the A-T lines for $\text{CdCr}_{1.7}\text{In}_{0.3}\text{Se}_4$ and $\text{CdCr}_{1.3}\text{In}_{0.7}\text{Se}_4$ thin films. We also have analysed the experimental data using the linear relationship between h_{eff} and τ^α in the form predicted by De Almeida-Thouless (A-T line):

$$h_{\text{eff}} = (8/(n+1)/(n+2))^{1/2} \tau_1^{3/2},$$

where $\tau_1 = 1 - T_{\text{CRO}}(H)/T_{\text{CRO}}(0)$ and $n = 1$

and in the form predicted by Gabay-Toulouse (G-T line):

$$h_{\text{eff}} = 2(n+2)/(n^2 + 4n + 2)^{1/2} \tau_2^{1/2},$$

where $\tau_2 = 1 - T_g(H)/T_g(0)$ and $n = 3$.

The reduced field $h_{\text{eff}} = h_a + h_m$.

The normalized external field $h_a = \eta g S_{\text{eff}} \mu_B H_a / k_B T_g(0)$ for G-T line and for A-T line $h_a = \eta g S_{\text{eff}} \mu_B H_a / k_B T_{\text{CRO}}(0)$, with $g = 2$, $S_{\text{eff}} = 3/2$ for Cr^{3+} and $\eta = 60$ (average size of clusters [7]). The internal field h_m is related to IFN, $h_m = mJ_0$ and m is the normalized macroscopic induced magnetization, J_0 is the exchange constant of IFN. The internal magnetic field h_m , calculated for each value of h_a , was found to be dilution level dependent. The h_m decreases when amount of indium is increasing. The data of $\text{CdCr}_2\text{Se}_4:\text{In}$ (REE) and $\text{CdCr}_{2-2x}\text{In}_{2x}\text{Se}_4$ (SG) thin films are collected in Table for $\tau_1 = \tau_2 = 0.20$. We found that the exchange constant J_0 of IFN, decreases with increasing In concentration.

TABLE

Reduced field for different compounds.

Sample	h_{eff}	h_a	h_m	$J_0 = 1 - h_a/h_{\text{eff}}$
CdCr ₂ Se ₄ : In				
*	0.100	0.002	0.097	0.98
**	0.921	0.020	0.901	0.98
CdCr _{1.7} In _{0.3} Se ₄ *	0.100	0.012	0.088	0.88
CdCr _{1.3} In _{0.7} Se ₄ *	0.100	0.026	0.074	0.74

*A-T line, **G-T line.

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