Critical lines of magnetic semiconductor thin films: Experiment

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The irreversibilities between the field-cooled and zero-field-cooled dc magnetization were used to determine the field and composition dependence of the spin-glass freezing temperature in $CdCr_{2-2x}In_{2x}Se_4$ thin films. The magnetic ordering was confirmed by the temperature dependence of induced magnetization M and unidirectional magnetic anisotropy field H_{an} determined from ferromagnetic resonance data (4.2–120 K). The experimentally determined H-T phase diagram shows two instability lines: the Gabay-Toulouse-type (GT line) and the Almeida-Thouless-type (AT line) for thin films of $CdCr_2Se_4$:In with reentrant transition and the AT line for $CdCr_{2-2x}In_{2x}Se_4$ in the spin-glass state. The AT and GT lines obey the relation $\tau=[(n+1)(n+2)/8]^{1/3}\ (h_{eff})^{2/3}$ and $\tau=[(n^2+4n+2)/(4(n+2)^2)]\ (h_{eff})^2$, respectively, for the normalized effective field $h_{eff}=h_a+h_m$. The first term in h_{eff} stands for the external magnetic field, while the second is related to the internal field of the infinite ferromagnetic network (long-range ordering). The value of h_m determined from the H-T phase diagram was found to be dependent on indium concentration. [S0163-1829(97)02709-4]

I. INTRODUCTION

The chalcogenide spinel of $CdCr_2Se_4$ exhibits specific properties that can be controlled by the dilution level. When the concentration of magnetic atom is changed, the following magnetic phases are obtained:

(i) for $CdCr_2Se_4$:In, the ferromagnetic state (FM) with the reentrant transition (REE),

(ii) for $CdCr_{2-2x}In_{2x}Se_4$ ($0<x\leqslant 0.4$), spin-glass state (SG).

The ferromagnetic state (FM) with the reentrant transition (REE) is characterized by high fluctuations of the exchange constant at the temperature close to REE transition. This state further evolves into spin-glass (SG) state. On the microscopic scale this system is considered as consisting of an infinite ferromagnetic network (IFN) with long-range ferromagnetic order and finite spin clusters (FSC's) randomly distributed in IFN. In this system, the amount of IFN significantly dominates over FSC, in the volume of a sample. The finite-spin clusters are described by the Anderson[1] and Continentino[2] models. The models consider a system of spins to be in metastable states, illustrated by two-level systems (TLS's), which are related to the sense of rotation of the spin direction. The energy of TLS's is described by an asymmetric double-well potential which is a random quantity.

For such a system the two instability lines are obtained in the H-T phase diagram: the de Almeida-Thouless (AT) line,[3] which indicates the onset of longitudinal freezing and Gabay-Toulouse (GT) line for the transverse freezing.[4] For $CdCr_{2-2x}In_{2x}Se_4$ ($0<x\leqslant 0.4$) in the spin-glass state, the ratio of IFN to FSC depends on the dilution level. With increasing amount of indium the FSC's dominate over the IFN. This system exhibits the AT instability line.

This microscopic picture of both REE and SG systems develops the problem of instability of the AT and GT lines.[3–5] Introducing the infinite ferromagnetic network, the internal magnetic field, related to IFN, has to be taken under consideration in the description of the instability lines.

With respect to irreversibility, we have concentrated on the comparison between experimentally determined H-T irreversibility lines and critical lines described by the AT and GI models. The disordered magnetic system is macroscopically characterized by

(i) the temperature dependence of induced magnetization M, which includes the nonzero density of states in the energy gap of magnons dispersion relation,

(ii) the unidirectional magnetic anisotropy field H_{an}, which originates from the Dzaloshinsky-Moriya interaction (DM).

Thin films of $CdCr_{2-2x}In_{2x}Se_4$ (for $0\leqslant x\leqslant 0.4$) were obtained by rf sputtering. The chosen preparation method of the samples allows us to change the atomic composition in a wide range. The thickness of the investigated films was about 5000 Å, thus the samples were considered as three-dimensional magnetic medium. The basic parameters $M(T)$ and H_{an} (T), that classify the magnetic phase of samples, were determined from the ferromagnetic resonance (FMR) and superconducting quantum interference device (SQUID) data. The freezing lines were found from the onset of irreversibilities between the field-cooled (FC) and zero-field-cooled (ZFC) dc magnetization. We analyzed the phase diagram (H-T) in terms of the De Almeida-Thouless model[3] for the Ising's spin and of Gabay-Toulouse model[4,6] for Heisenberg spins. From the comparison between the theoretically predicted critical line and experimental data, the value of internal magnetic field h_m of IFN was determined.

II. MAGNETIZATION AND ANISOTROPY IN MAGNETIC DISORDERED STATE

The chalcogenide spinel of $CdCr_{2-2x}In_{2x}Se_4$, with respect to the amount of In, belongs to a different class of magnetic ordering. For $CdCr_2Se_4$:In the ferromagnetic state with transition to the spin-glass state (REE) is obtained. Diluting samples by In (x increases) the spin-glass state is expected (SG).[7–9] For each type of magnetic ordering the typi-

cal temperature dependence of the induced magnetization is predicted:

(i) State with REE. The temperature dependence of magnetization is described by relation:[7]

$$[M(0)-M(T)]/M(0)$$

$$=BT^{3/2}\xi(3/2)\sum_{l=1}^{\infty}[\exp(-l\Delta_r/k_BT)/l^{3/2}], \quad (1)$$

where

$$B=\xi(3/2)[g\mu_B/M(0)](k_B/4\pi D_s)^{3/2}.$$

$\xi(3/2)$ stands for the Riemann ξ function, Δ_r is the energy gap, and D_s is the spin-wave stiffness constant.

(ii) SG state. $M(T)$ is well represented by the relation[7]

$$[M(0)-M(T)]/M(0)=C_s/[\exp(\Delta_s/k_BT)-1]. \quad (2)$$

where C_s is responsible for the density of states at the energy gap of the magnon dispersion relation and Δ_s is a measure of the intercluster interaction.

In the disordered state, an unidirectional magnetic anisotropy is present. The microscopic phenomena responsible for this anisotropy are explained by the Dzyaloshinsky-Moriya interaction (DM).[10,11] The DM interaction is due to the spin-orbit scattering of the conduction electrons by the nonmagnetic impurities.

On the macroscopic scale, this anisotropy manifests itself as an internal magnetic field, so called the unidirectional magnetic anisotropy field (H_{an}).[7,12] H_{an} depends on the field-induced remanent magnetization and keeps the direction of the cooling field. It means that the spins have some memory of the cooling field direction.

The unidirectional magnetic anisotropy field is influenced by the dilution level and, being related to the induced magnetization, is also temperature dependent. The relation of $H_{an}(T)$ is known only from the experimental data, since no comprehensive model of the temperature dependence of H_{an} exists. It was reported in Refs. 7 and 12, that the unidirectional magnetic anisotropy field H_{an} for $CdCr_{2-2x}In_{2x}Se_4$ thin films decreases with increasing temperature and is dependent on the amount of In (H_{an} increases with increasing dilution level). We classify the magnetic state of thin films of $CdCr_{2-2x}In_{2x}Se_4$ on the basis of the temperature and concentration dependence of the field-induced magnetization and the unidirectional magnetic anisotropy field.

III. TRANSITION LINES–MEAN-FIELD THEORIES

The magnetic disordered state comprises a system of spins coupled by random Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange and the DM interaction. A set of spins is considered to be frozen in a random direction below the freezing temperature. In general, the random magnetic anisotropy provides mixing of the longitudinal- and transverse-spin components.

The DM interactions modify the results of the mean-field theory.[13,14] The transition line for the Ising model has been given by De Almeida–Thouless[3] (AT line) and indicates the onset of longitudinal freezing, while the transition line for the Heisenberg spins has been described by Gabay-Toulouse[4] (GT line) and is related to the transverse freezing.

The AT line for the Ising model is defined by the dependence of the freezing temperature on the effective magnetic field and is given by relation:[4,5]

$$\tau=[(n+1)(n+2)/8]^{1/3}h_{\rm eff}^{2/3}, \quad (3)$$

where n is the spin component, $\tau=1-T_g(H)/T_g(0)$, $h_{\rm eff}=h_a+h_m$ is the normalized effective field, $h_a=\eta g S_{\rm eff}u_BH_a/k_BT_g(0)$, η is an average size of the FSC clusters,[15] $h_m=J_0m$, is the normalized internal magnetic field of IFN, and $T_g(H)$ is the freezing temperature.

The internal magnetic field h_m is related to IFN, where m is the normalized macroscopic induced magnetization of IFN and J_0 is the magnetic coupling constant. The transition occurs for the vector spins with n components (Heisenberg model) along the GT line given by

$$\tau=\{(n^2+4n+2)/[4(n+2)^2]\}h_{\rm eff}^2. \quad (4)$$

It was reported in literature[14–16] that weakly diluted samples with REE exhibit AT and GT instability lines. For this kind of magnetic order two critical temperatures of irreversibility are experimentally detected and this phenomenon is a source of mixing of longitudinal and transverse spin freezing. When temperature increases, the crossover from the GT line to the AT line occurs, Kotliar and Sompolinsky[17] have found that the crossover effect takes place for $d=h_{\rm eff}^{2/3}$ ($d=D/J$, the ratio of strength of Dzaloshinsky-Moriya with respect to the RKKY interaction). For the SG state the AT line only characterizes the instability of the spin system.

IV. SAMPLES AND MEASUREMENTS METHODS

A. Technology

We investigated thin $CdCr_2Se_4$ films of chromium spinel with controlled concentration of In in the lattice. Thin films of $CdCr_2Se_4$:In exhibit reentrant transition (REE), diluted samples of $CdCr_{2-2x}In_{2x}Se_4$ with $0<x\leqslant 0.4$ are in the spin-glass state (SG). It should be emphasized that the uniformity of the studied material is very difficult to obtain. The parameters of deposition process have to be carefully chosen and tested with proper accuracy. Films were deposited by the rf sputtering technique on Corning glass substrates. The device is equipped with a two-cathode system and rotatable substrate holder of controlled temperature. We used the chromium cathode for depositing the buffer and overlayer films. The powdered CdSe cathode uniformly spotted with Cr_2Se_3 and In_2Se_3 was used for sputtering Cd-Cr-In-Se sublayers. The thickness of the buffer and top layers of Cr was scanned from 20 to 100 Å whereas a thickness of the middle layer of Cd-Cr-In-Se was larger than 3000 Å. We have found that the thickness of Cr of about 50 Å gives good adhesion. As-deposited samples are in the amorphous state and have a form of multilayer structure of Cr/Cd-Cr-In-Se/Cr. Heat treatment provides uniform polycrystalline single films with the required composition. The processes of crystallization were carried out in a Kristalloflex 4H x-ray-diffraction apparatus at temperatures ranging from 300 to 850 K and for different annealing times from 0.5 to 5 h. The x-ray-diffraction patterns were recorded *in situ* after each stage of heat treatment in the argon atmosphere had been completed.

It was found that the samples reached the polycrystalline state after being annealed at 790 K for 1 h. The composition of samples was analyzed by means of an x-ray microprobe (ARL SEMQ microanalyzer) and Auger spectroscopy (Riber LAS-620). The film thickness was measured by means of Talysurf 4 profilometer. Other details of the preparation techniques are described in Ref. 12.

B. Measurements of $M(T)$ and $H_{an}(T)$

The magnetic states with REE and SG were confirmed by the experimentally determined temperature dependence of induced magnetization M and the presence of the unidirectional magnetic anisotropy field H_{an}. The value of H_{an} increases with the amount of In.

We determined the temperature dependence of M and H_{an} by means of the ferromagnetic resonance experiment (FMR) at microwave frequency within the temperature range from 4 to 200 K. In the FMR technique, the position of resonance peak depends on the total value of the internal magnetic field H_{int}. For thin films, apart from the external magnetic field H, the shape magnetic anisotropy field $H_u = 4\pi M$ (M is the induced magnetization) and the unidirectional magnetic anisotropy field H_{an} contribute to H_{int}. In the FMR experiment for thin films, two characteristic geometries, perpendicular, and parallel, are used for determining the basic magnetic parameters.[7,12] In general, different internal effective fields alter the spin system in both geometries so one could expect different values of H_{an} for perpendicular and parallel resonance. For the uniform mode of FMR for perpendicular and parallel geometry we have the dispersion relation in the form:

$$(\omega/\gamma)_{\perp} = H_{\perp} + H_{an}^{\perp} - 4\pi M_{\perp}, \tag{5}$$

$$(\omega/\gamma)_{\parallel}^2 = (H_{\parallel} + H_{an}^{\parallel})(H_{\parallel} + H_{an}^{\parallel} + 4\pi M_{\parallel}), \tag{6}$$

where $\omega = 2\pi v$, v is the microwave frequency (X band), g is the gyromagnetic ratio of processing moments, $H_{\perp}$ and $H_{\parallel}$ are the resonance fields in perpendicular and parallel geometry of the experiment, respectively. In our case the conditions: $(\omega/\gamma)_{\perp} = (\omega/\gamma)_{\parallel}$ and $M_{\perp} = M_{\parallel}$ are kept during the whole resonance experiment.

For investigated thin films the resonance field versus temperature, for both geometries, drops at low temperatures by the same amount. Thus, following Refs. 7, 18, and 19, we have assumed that $H_{an}^{\perp} = H_{an}^{\parallel}$. For the chosen ($X$ band) microwave frequencies we used Eqs. (5) and (6) for determining the temperature dependence of M and H_{an}.

C. Determination of the H-T phase diagram

To derive the H-T phase diagram, different techniques are applied: torque measurements,[16] field cooled (FC) and zero-field-cooled (ZFC) dc magnetization,[15] Mossbauer effect,[20,21] and ac susceptibility. In our investigations we used the FC and ZFC dc magnetization measurements carried out by SQUID. The dc magnetization was measured as a function of temperature for a set of external magnetic fields applied within the film plane. FC and ZFC dc magnetizations were detected during experiment, performed always in the same sequence: the sample was cooled from 220 to 4.2 K in zero field, a small magnetic field (H_a) was applied and the magnetization data were collected for temperature changing from 4.2 to 220 K (ZFC data). For the same value of H_a, FC data were recorded from 4.2 to 220 K. The experiment was done for several values of H_a up to the limiting case when ZFC and FC dc magnetization overlap.

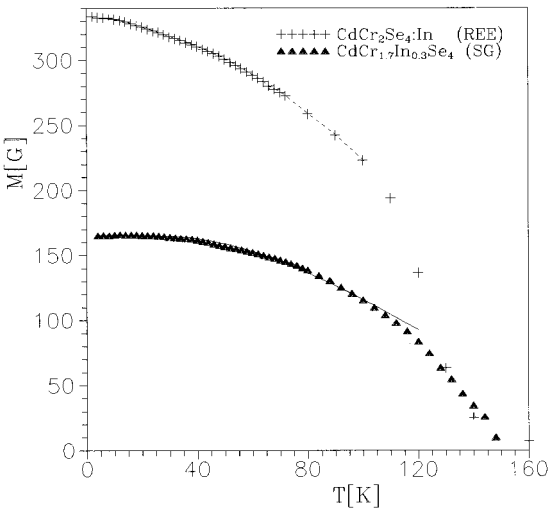

FIG. 1. Temperature dependence of the magnetization. Dashed line represents the best fit of Eq. (1) to experimental data ($\Delta_r = 5$ K) for a thin $CdCr_2Se_4$:In film. Solid line represents the best fit of Eq. (2) to the experimental data ($C_s = 1.7$, $\Delta_s = 190$ K) for $CdCr_{1.7}In_{0.3}Se_4$ thin film.

On the basis 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. From the relationship between ΔM and T the freezing temperature $T_g(H)$ was established, then the H-T phase diagram was found.

The $\Delta M(T)$ curve exhibits one or two critical temperatures depending on the sample composition. Following Ref. 15 we defined T_g as a temperature below which the first onset of irreversibilities is seen, T_{CRO} as a temperature below which ΔM increases relatively rapidly.

If only one critical temperature is present, the AT or GT line can be established. When two critical temperatures are found, the phase diagram has the AT and GT lines.[15]

V. EXPERIMENTAL RESULTS AND DISCUSSION

We present results for thin films of $CdCr_2Se_4$:In—REE and $CdCr_{1.7}In_{0.3}Se_4$ and $CdCr_{1.3}In_{0.7}Se_4$—SG. The REE and SG states of thin films were confirmed by the temperature dependence of the field-induced magnetization and the unidirectional magnetic anisotropy field. We present the $M(T)$ and $H_{an}(T)$ experimental data for thin films $CdCr_2Se_4$:In with the reentrant transition and $CdCr_{1.7}In_{0.3}Se_4$ in the spin-glass state.

The data of irreversibilities between the FC and ZFC magnetization are demonstrated for the samples with REE and in the SG state. The final data are shown in a form of H-T phase diagram for $CdCr_2Se_4$:In, $CdCr_{1.7}In_{0.3}Se_4$, and $CdCr_{1.3}In_{0.7}Se_4$ samples.

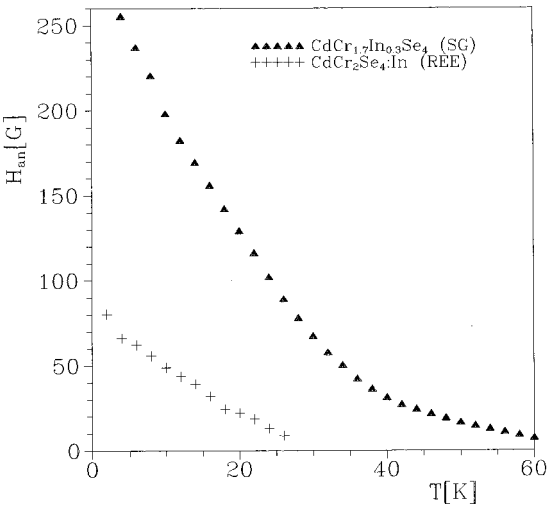

FIG. 2. Unidirectional magnetic anisotropy field as a function of temperature for $CdCr_2Se_4$:In and $CdCr_{1.7}In_{0.3}Se_4$ thin films.

A. Magnetization and unidirectional magnetic anisotropy field

Figure 1 presents the temperature dependence of magnetization, taken from FMR data as well as the calculated one, for two samples: $CdCr_2Se_4$:In and $CdCr_{1.7}In_{0.3}Se_4$. The dashed curve in Fig. 1 indicates the modified Bloch's law, according to Eq. (1), for the sample with reentrant transition. The value of energy gap $\Delta_r=5$ K was taken as a fitting parameter. The solid line in Fig. 1 is theoretically found using Eq. (2), for the sample in the SG state. In this case we have the fitting parameters $C_s=1.7$ and $\Delta_s=190$ K. The good agreement between theory and experimental data is seen in both cases.

Figure 2 shows the experimentally determined temperature dependence of the unidirectional magnetic anisotropy field. The results of $H_{\mathrm{an}}(T)$, presented in Fig. 2, are typical for the samples with REE and in the SG state.[7,18]

B. Irreversibility of dc magnetization

The onset of the irreversibilities between the FC and ZFC dc magnetization—SQUID techniques, was used for determining the critical lines. As an example we present in Fig. 3 the low-field magnetization of a $CdCr_2Se_4$:In thin film. From the data shown in Fig. 3 the irreversible part of magnetization defined as $\Delta M=M_{\mathrm{FC}}-M_{\mathrm{ZFC}}$ vs temperature was determined.

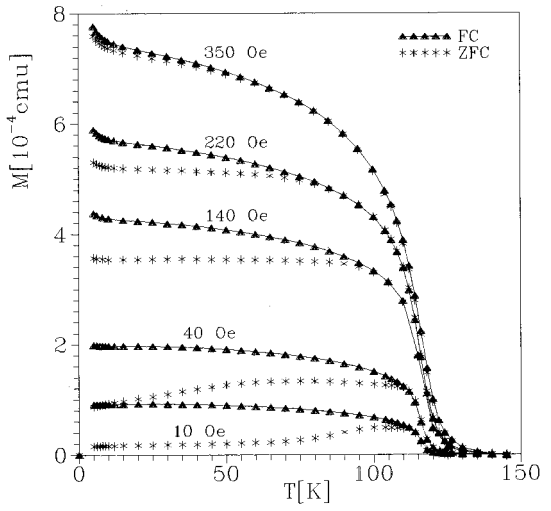

FIG. 3. ZFC and FC dc magnetization for a $CdCr_2Se_4$:In thin film.

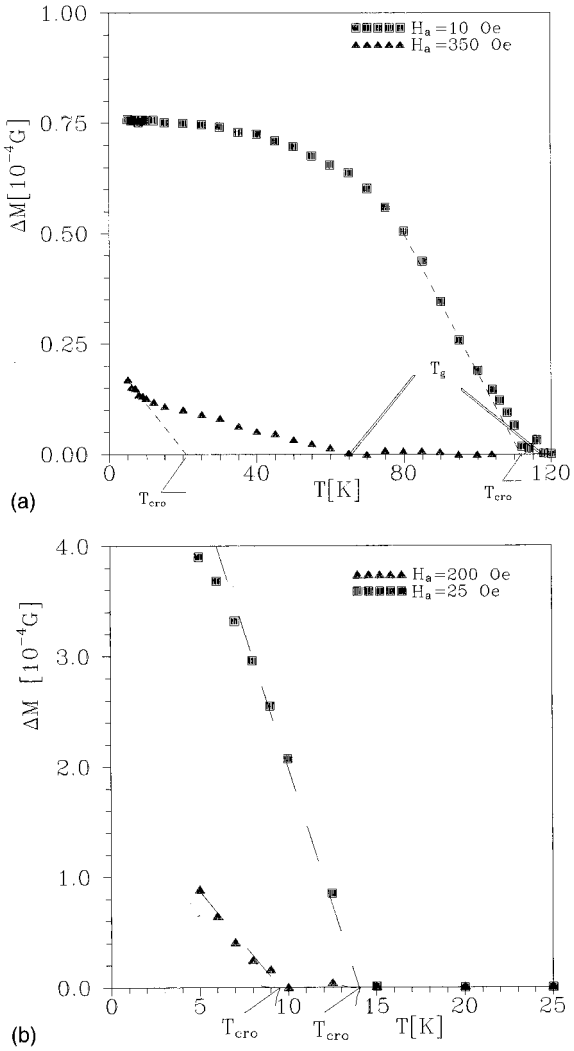

FIG. 4. (a) Irreversible part of ΔM as a function of temperature for $CdCr_2Se_4$:In and (b) $CdCr_{1.7}In_{0.3}Se_4$ thin films, respectively.

Figure 4(a) presents the data of ΔM for $CdCr_2Se_4$:In (REE) for two external magnetic fields: $H_a=10$ and 350 G. The character of both curves differs significantly, but for each of them two critical temperatures T_g and T_{CRO} can be found.[15] The T_g corresponds to $\Delta M(T)=0$ and T_{CRO} is defined as the temperature below which ΔM is increasing relatively rapidly. The data of $\Delta M(T)$ for the samples of $CdCr_{1.7}In_{0.3}Se_4$, and $CdCr_{1.3}In_{0.7}Se_4$ exhibit only one critical temperature T_{CRO}.

As an example of the temperature dependence of ΔM, for the sample in the spin-glass state, Fig. 4(b) shows the experimental data for a thin film of $CdCr_{1.7}In_{0.3}Se_4$. The data are presented for two values of external magnetic field $H_a=25$ and 200 G. The character of the curve reveals only one critical temperature T_{CRO} for both fields. It seems to be in a good agreement with prediction of the Ising model for a diluted magnet when longitudinal spin freezing prevails in the critical phenomena.

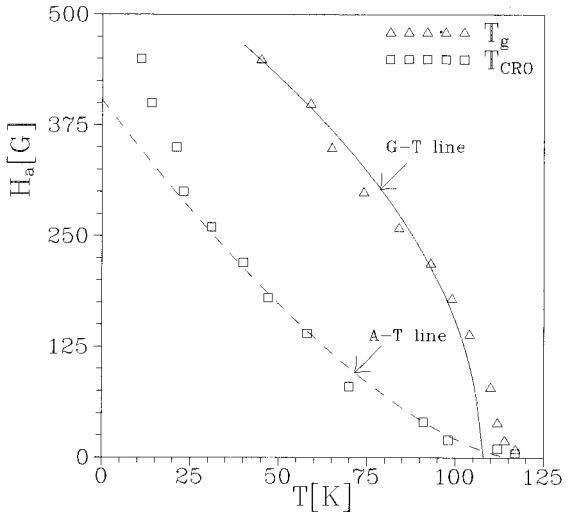

FIG. 5. H_a-T (external field-temperature) phase diagram of a $CdCr_2Se_4$:In thin film with reentrant transition.

C. The phase diagrams

We obtained the H_a-T phase diagram with (i) AT and GT critical lines for thin films with the reentrant transition and (ii) the AT line for thin films in the spin-glass state. Figure 5 presents the experimentally determined H_a-T curves for thin films of $CdCr_2Se_4$:In (REE). It is seen that above $T=110$ K the character of the GT line changes to the AT line. Note that the $T_g(0)=107$ K and $T_{CRO}(0)=120$ K. The results are similar in character to those presented in Ref. 15 for bulk $CdCr_{1.7}In_{0.3}S_4$. Figure 6 shows the AT lines for $CdCr_{1.7}In_{0.3}Se_4$ [$T_{CRO}(0)=17.7$ K] and $CdCr_{1.3}In_{0.7}Se_4$ [$T_{CRO}(0)=17.4$ K] thin films. We have also analyzed the experimental data using the linear relationship between $h_{\rm eff}$ and τ in the form of [see Eq. (3)]

$$h_{\rm eff}=\{8/[(n+1)(n+2)]\}^{1/2}\tau_1^{3/2}, \tag{7}$$

where $\tau_1=1-T_{CRO}(H)/T_{CRO}(0)$ predicted by de Almeida–Thouless (AT line) with $n=1$, and in the form of [see Eq. (4)]

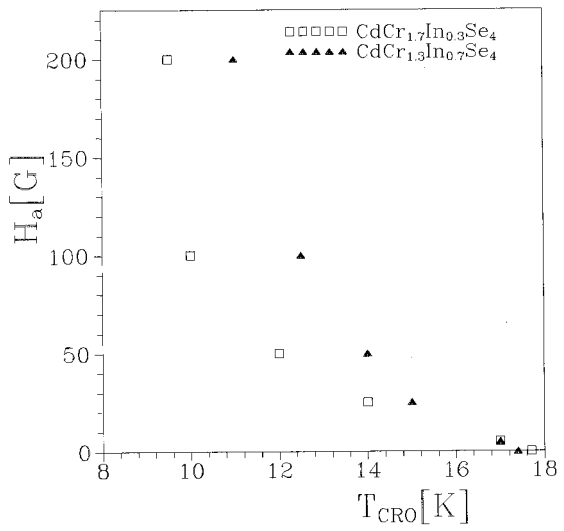

FIG. 6. H_a-T (external field-temperature) phase diagram of $CdCr_{1.7}In_{0.3}Se_4$ and $CdCr_{1.3}In_{0.7}Se_4$ thin films, respectively.

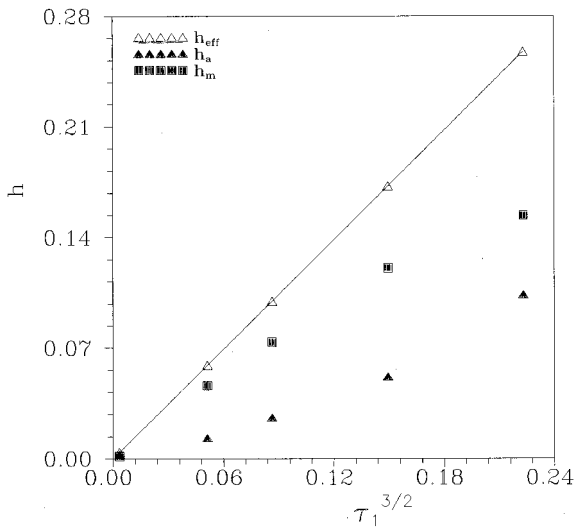

FIG. 7. The $h_{\rm eff}$, h_a, and h_m versus $\tau_1^{3/2}$ for a thin $CdCr_{1.3}In_{0.7}Se_4$ film.

$$h_{\rm eff}=2(n+2)/(n^2+4n+2)^{1/2}\tau_2^{1/2}, \tag{8}$$

where $\tau_2=1-T_g(H)/T_g(0)$ predicted by Gabay-Toulouse (GT line) with $n=3$.

The value of $T_g(0)$ and $T_{CRO}(0)$ as well as $T_g(H_a)$ and $T_{CRO}(H_a)$ are known from the experimental data of $\Delta M(T)$. Then values of τ_1 and τ_2 could be calculated for the AT and GT critical lines, respectively. The prefactor in Eqs. (7) and (8) was taken from the theory. The value of $h_{\rm eff}$ already defined as superposition of two terms, $h_{\rm eff}=h_a+h_m$, can be determined from Eqs. (7) and (8).

For each value of h_a the internal magnetic field h_m, related to the infinite spin clusters, was calculated for each sample. As an example, we present in Fig. 7, the relations between $h_{\rm eff}$, h_a, and h_m and $\tau_1^{3/2}$ for the sample of $CdCr_{1.3}In_{0.7}Se_4$.

The data for $CdCr_2Se_4$:In (with REE) and $CdCr_{2-2x}In_{2x}Se_4$ (in the SG state) with $x=0.15$ and $x=0.35$ thin films are collected in Table I for $\tau_1=\tau_2=0.20$ as an example. The reduced field: $h_{\rm eff}$, h_a, and h_m were already defined in paragraph 3. We have taken $g=2$, $S=3/2$ for Cr^{3+} and $\eta=60$ (average size of finite spins cluster FSC).[15]

In some models of the spin-glass state the system is considered as having the form of clusters embedded in IFN. Below freezing temperature, spins in a single cluster are aligned and all clusters are randomly distributed in the sample volume. In this microscopic picture of disordered state, the distribution of clusters size is also taken into account as well as the composition dependent size of a cluster.[9,22] We are not able to perform the susceptibility measurements that allow us to determine the cluster size in the indirect way.[9] Therefore we have to start from the simplest case, assuming that the average size of clusters (FSC), is stable, not altered by a dilution level within the examined range of indium content. Only the number of FSC is affected by the dilution level.

We have taken the size of clusters equivalent to 60 spins, which corresponds to about 3–4 spins along one direction of lattice. The lattice parameter of $CdCr_2Se_4$ equals 10.75 Å and the Cr-Cr distance is 3.63 Å. It gives about 27 cluster

TABLE I. Critical temperatures and reduced field for different compounds.

Sample	$T_{CRO}(0)$ (K)	$T_g(0)$ (K)	h_{eff}	h_a	h_m	$J_0=1-h_a/h_{eff}$
$CdCr_2Se_4$:In						
(AT line)	120		0.100	0.002	0.098	0.98
(GT line)		107	0.921	0.020	0.901	0.98
$CdCr_{1.7}In_{0.3}Se_4$						
(AT line)	17.7		0.100	0.012	0.088	0.88
$CdCr_{1.3}In_{0.7}Se_4$						
(AT line)	17.4		0.100	0.026	0.074	0.74

spins in a unit cell. In our calculations, we estimate the value of cluster size above the unit cell, taken one more spin cluster (four spins along every direction on the lattice), then the size of the clusters is 64. Finally we have taken 60 as the average size of the spin clusters.

We have found that the exchange constant $J_0=1-h_a/h_{eff}$ (Ref. 9) of IFN (infinite ferromagnetic network) is dilution-level dependent in such a way that with increasing In concentration J_0 decreases. It is reported in Refs. 14, 15, and 17 that the dynamics of vector spin glasses leads to a crossover from Heisenberg spins ($h_{eff}\sim\tau_2^{1/2}$) to Ising spins ($h_{eff}\sim\tau_1^{3/2}$). For higher magnetic field the critical line in the magnetic field—temperature plane have the part similar to the GT line. With decreasing magnetic field there is part of the $H(T_g)$ line which has the field temperature dependence of that typical for the AT line. The change from GT to AT line takes place at the crossover field $h_{eff}^{2/3}=d$ ($d=D/J$, defined in Sec. III) which is altered by the random magnetic anisotropy.

In Fig. 5, the two critical AT and GT lines for thin film of $CdCr_2Se_4$:In (REE) are present. Both lines are merging into one which is AT like, at $H_a=25$ G. There is the relation $h_{eff}^{2/3}\propto d$. We put with approximation $h_{eff}^{2/3}=d$ and $D=9$ K was found. A similar effect was also expeirmentally confirmed in Ref. 15. There is an analytical formula for a relationship between microscopic DM interaction constant D and macroscopic unidirectional magnetic anisotropy energy constant K ($K=H_{an}M$).[23] The formula consists of the standard expressions of Ruderman-Kittel-Kasuya-Yosida (RKKY) constant and a DM vector.

The authors of Refs. 16 and 24 have obtained satisfactory agreement between the calculated and experimentally determined values of the anisotropy constant, for bulk samples of CuMn alloys doped with Au and Ni, in the spin-glass state. In our case, at the present state of the investigations of polycrystalline samples, we cannot perform the theoretical calculations of d due to the complicated crystallographic structure of the chalcogenide spinel.

VI. CONCLUSIONS

The microscopic picture of the disordered magnetic system with finite-spin clusters (FSC's) and the infinite ferromagnetic network (IFN) with the long-range magnetic ordering modifies the critical AT and GT lines of the H-T phase diagram. The IFN are the source of an internal magnetic field which contributes to longitudinal and/or transverse spins freezing. In this paper we investigated thin films of magnetic semiconductors: (i) $CdCr_2Se_4$:In (with REE), (ii) $CdCr_{2-2x}In_{2x}Se_4$ (in the SG state) with $x=0.15$ and $x=0.35$.

The samples were obtained by rf sputtering. The type of disordered magnetic state has been studied by the temperature and composition dependence of the induced magnetization and unidirectional magnetic anisotropy field.

The critical lines were found from the onset of irreversibilities between the field-cooled (FC) and zero-field-cooled (ZFC) dc magnetization. The H_a-T phase diagram for samples with REE has two critical lines AT and GT, while for samples in the SG state one AT critical line was found. On the basis of the H_a-T phase diagram the internal magnetic field h_m related to IFN was determined. This field is dilution-level dependent; decreasing with the increasing amount of indium. Such an effect was expected since the ratio of IFN/FSC increases when dilution level is getting smaller. This leads to the increase in the reduced magnetization m and, as a result the increase in h_m is observed. It remains a challenge for us to determine reliably the internal magnetic field of the long-range-ordered clusters. Also the value of $d=D/J$, the ratio of strength of Dzaloshinsky-Moriya with respect to the RKKY interaction, was determined in the case of the thin films with reentrant transition.

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