Temperature dependence of interactions in diluted magnetic semiconductors

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Coupling between the infinite and finite clusters of a spin system with the reentrant transition and spin-glass state has been studied as a function of temperature in the range from 4 K to 300 K. Thin films of $CdCr_{2x}In_{2-2x}Se_4$ with $0.8 \leq x \leq 0.95$ were investigated. The magnetic properties of $CdCr_{2x}In_{2-2x}Se_4$ spinel are very sensitive to the substitution of Cr by In, which leads to a random distribution of magnetic interactions. As-deposited samples had a multilayered structure of Cr/Cd-Cr-In-Se/Cr. After heat treatment we have obtained a uniform single film of controlled composition. The temperature dependence of linewidth and line shift of the ferromagnetic resonance, in parallel geometry, was used to compute the coupling parameters: M, which describes the resonance interaction between the finite and infinite clusters, and D, which is related to the relaxation processes. The models of randomly distributed spins proposed by Anderson and Continentino were applied. The model of a two-level system with asymmetric double-well potentials predicts a frequency shift due to the coupling between the infinite ferromagnetic network and finite clusters. We assume that the height of a barrier introduced in this model is a linear function of the temperature. The significant temperature dependence of coupling parameters was obtained for samples in the spin-glass state with Cr concentration $x = 0.85$ and 0.8. We have found that M and D are not single-valued functions of indium concentration (dilution level). We have also determined the temperature dependence of $(M-D)$, which has an intuitive meaning as being responsible for the interaction of nondamped magnons of the infinite cluster with a two-level system. The expression $(M-D)$ was found to be a linear function of the temperature.

I. INTRODUCTION

Diluted ferromagnetic systems exhibit specific properties that can be controlled by the dilution level.[1–7] When the concentration of magnetic atoms is changed, we have the following magnetic phases: the ferromagnetic state (FM) with the reentrant transition (REE) and spin-glass state (SG). These phases are considered as consisting of an infinite ferromagnetic network (IFN) with long-range ferromagnetic order (magnons) and finite spin clusters (FC). The finite clusters are randomly distributed in IFN. Note that this description differs in details from that proposed in our previous paper.[6] Our previous microscopic picture of SG excluded the presence of IFN. Experimental data obtained from the ferromagnetic resonance and magnetic balance measurements show that, above the freezing temperature, the samples exhibit properties typical for a long-range-ordered spin system.

The ferromagnetic state with the reentrant transition is characterized by high fluctuations of the exchange constant at the temperature closer to REE transition than evolves in SG state.

The diluted ferromagnetic system is macroscopically characterized by (i) the magnetization (saturation magnetization), the temperature behavior of which depends on the magnetic phase; (ii) the unidirectional magnetic anisotropy; (iii) other properties such as electrical transport, optical and magneto-optical parameters.

Therefore, we expect the system to show some interesting variations of magnetic, transport, and optical properties with the dilution levels that, on a microscopic scale, would be explained by an interaction between IFN-FC. We concentrated on $CdCr_{2x}In_{2-2x}Se_4$ thin films with $0.8 \leq x \leq 0.95$. This composition corresponds to a range of magnetic phases: (i) ferromagnetic state with the reentrant transition (REE)—a sample with the composition of $CdCr_{1.9}In_{0.1}Se_4$; (ii) spin glass SG—the samples with the composition of $CdCr_{1.7}In_{0.3}Se_4$ and $CdCr_{1.6}In_{0.4}Se_4$.

These spin systems are characterized by different magnetic excitations and coupling interactions in comparison with the ferromagnetic state. We also expect the unidirectional magnetic anisotropy to be attributed to the spin-glass state. The interactions are described by Anderson's model[8] which was significantly developed by Continentino.[2,9]

The models consider spin systems to be in metastable states, illustrated by two-level systems (TLS), which are related to the sense of rotation of the spin direction. In our notation TLS corresponds to FC. The energy of TLS is described by an asymmetric double-well potential which is a random quantity altered by a particular configuration of spins surrounding the two minima. Apart from spin-wave excitations in IFN, additional magnetic excitations due to the metastable ferromagnetic states on both sides of double-well potentials are expected. In general, the effective interaction has three components: exchange interactions in IFN which are responsible for excitation of spin waves (magnons), two-level systems interactions being relevant in the finite clusters, and also coupling between magnons and TLS.

The ferromagnetic resonance (FMR) at microwave frequency is one of the most sensitive tools for studies of the interaction on the microscopic scale. These interactions alter the resonance peak-to-peak linewidth Γ because all relaxation processes have their own contribution to it. The line shift δH due to the unidirectional magnetic anisotropy can be calculated from the position of the resonance peak. The analytical formulas used for a discussion of our results are presented in Sec. III. These formulas include two intrinsic parameters that characterize the interaction between the infinite ferromagnetic network (IFN) and finite spin clusters (FC):[2,3,9]

(i) M, which characterizes the resonant interaction between spin waves and two-level systems (IFN and FC) in which only TLS with energy splitting approximately equal to the magnon energy take part.

(ii) D, which describes the relaxation processes—perturbation of interaction between the finite and infinite clusters due to the modulation of TLS by spin-wave propagating in the infinite ferromagnetic network. Then the intra-FC relaxation processes are in some way hidden in the value of D. As a feedback, the magnons with shifted frequency should be emitted or absorbed. This is also considered as a scattering process between magnons of IFN and TLS—treated as a defect.

The indirect interaction between FC can be modulated by magnons of the ferromagnetic network (IFN). The temperature dependence of linewidth Γ and line shift δH was used to compute $M(T)$ and $D(T)$ dependence.

As it results from our calculations, the dependence of $M(T)$ and $D(T)$ are nonmonotonic functions of dilution level, exhibiting the maximum for $x=0.85$. The value of $(M-D)$ could be interpreted as a contribution to a process which comes from the interaction between nondamped magnons and TLS. The temperature dependence of this parameter reflects the assumed temperature dependence of the barrier height.

II. EXPERIMENT

Thin films of $CdCr_{2x}In_{2-2x}Se_4$ with $0.8 \le x \le 0.95$ deposited in a high-vacuum system were investigated by means of the ferromagnetic resonance. As-deposited samples are in the form of a multilayer structure: Cr/Cd-Cr-In-Se/Cr. This structure gives, after heat treatment, a uniform single film with the controlled magnetic properties. A single film is obtained as a result of diffusion processes and interface interactions during heat treatment. We scan the thickness of the buffer and top layers of Cr from 20 Å to 100 Å with a thickness of the middle layer of Cd-Cr-In-Se larger than 3000 Å. It was found that the thickness of Cr of about 50 Å gives the best uniform single films with good adhesion. To get a required magnetic phase we changed the composition of the middle layer. Therefore, the final parameters of a single film depend on the initial structure of a multilayer. All details of the thin films' deposition have been presented in our previous paper.[6,10]

The magnetic balance technique was applied for determination of $M_s(T)$. Experimentally found $M_s(T)$ was also confirmed by the theoretical formulas (see Ref. 6) for reentrant transition,

$$[M(0)-M_s(T)]/M(0) = BT^{3/2}\xi(3/2)\sum_{n=1}^{\infty}[\exp(-n\Delta_r/k_BT)/n^{3/2}]\,, \quad (1)$$

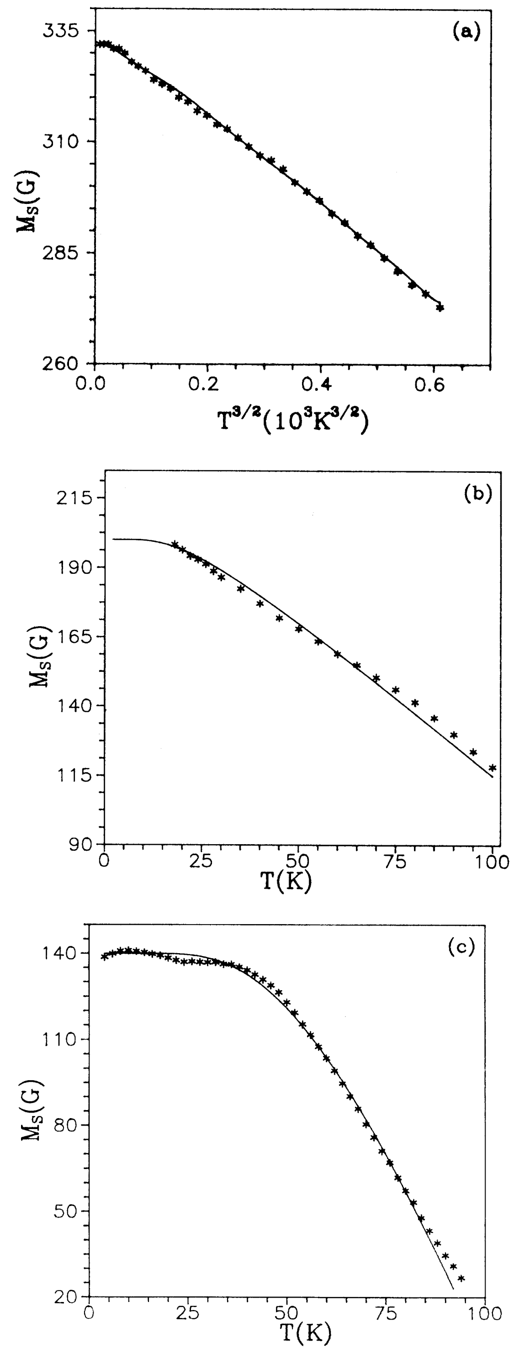

FIG. 1. (a) M_s vs $T^{3/2}$ for $CdCr_{1.9}In_{0.1}Se_4$ thin film. The solid line represents the best fit of Eq. (1) to the experimental data with $\Delta_r=5$ K. (b) M_s vs T for $CdCr_{1.7}In_{0.3}Se_4$ thin film. The solid line represents the best fit of Eq. (2) to the experimental data with $\Delta_s=60$ K, $C_s=0.35$. (c) Temperature dependence of M_s for $CdCr_{1.6}In_{0.4}Se_4$ thin film. The solid line represents the best fit of Eq. (2) to the experimental data with $\Delta_s=140$ K, $C_s=5.4$.

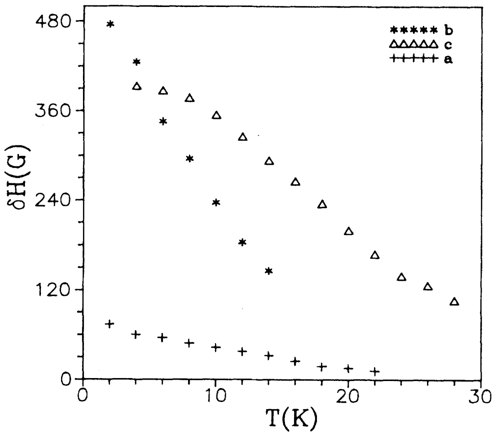

FIG. 2. Temperature dependence of line shift for (a) $CdCr_{1.9}In_{0.1}Se_4$, (b) $CdCr_{1.7}In_{0.3}Se_4$, (c) $CdCr_{1.6}In_{0.4}Se_4$.

and spin-glass state,

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

Figures 1(a), 1(b), and 1(c) present the temperature dependence of the saturation magnetization M_s of investigated samples. As shown, this behavior corresponds to the REE [Fig. 1(a)] and SG [Figs. 1(b) and 1(c)].[5,6]

Temperature dependence of the linewidth Γ and the line shift δH (Refs. 2 and 3) allows us to determine the interaction parameters M and D of the spin systems as well as their temperature dependence. X-band microwave spectrometer (FMR) was used to find $\delta H(T)$ and $\Gamma(T)$ within the temperature range extending from 4.2 to 300 K.

The line shift δH is due to the induced anisotropy field arising from Dzyaloshinsky-Moriya (DM) interactions.[11] The details of the model and calculation of DM interaction predict that the unidirectional magnetic anisotropy for alloys with randomly located spins depends on the value of the effective internal field.[12] Thus we expect different values of the unidirectional magnetic anisotropy

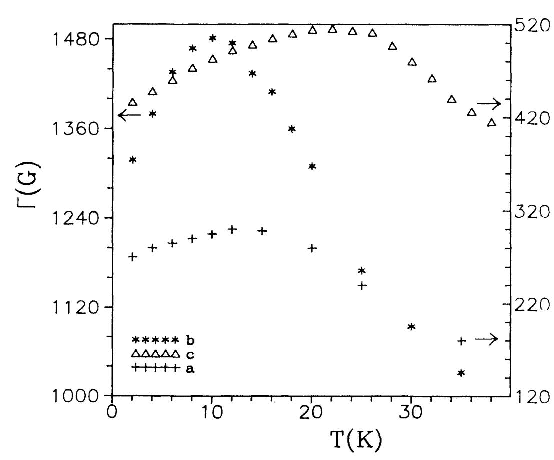

FIG. 3. Temperature dependence of the resonance linewidth for (a) $CdCr_{1.9}In_{0.1}Se_4$, (b) $CdCr_{1.7}In_{0.3}Se_4$, (c) $CdCr_{1.6}In_{0.4}Se_4$.

field for parallel and perpendicular ferromagnetic resonance.

It is well known that in both cases the resonance fields differ significantly due to the effective internal field. In this paper we are discussing the case of parallel resonance and, from the following equation,

$$(\omega/\gamma)_{\parallel}^2=(H_{\parallel}+\delta H)(H_{\parallel}+\delta H+4\pi M_s) \ , \qquad (3)$$

temperature dependence of δH is found. In Eq. (3), $\gamma=g\mu_B/\hbar$ is the gyromagnetic ratio of precessing moments, $\omega=2\pi\nu$, ν is a microwave frequency.

Figures 2 and 3 present the temperature dependence of δH and Γ, respectively.

III. RESULTS AND DISCUSSION

The Hamiltonian which describes magnons, two-level systems, and their coupling has the following form:[2]

$$H=\sum_k \epsilon_k a_k^\dagger a_k+(\varepsilon/2)\sigma^z+(\Delta/2)\sigma^x+G\sigma^z S^x \ . \qquad (4)$$

The first term describes magnons. The $a_k^\dagger$ and a_k are the creation and annihilation operators, respectively. The second term is responsible for the energy difference between metastable ferromagnetic states and ε represents this difference. The third term corresponds to the quantum-mechanical tunneling between magnetic metastable states, and Δ is the tunneling frequency. Then the second and third terms describe the energy of TLS. The last term in Eq. (4) is the energy due to the coupling between magnons and TLS and G is the coupling constant. As it was presented in Ref. 2, after a diagonalization procedure, the Hamiltonian can be rewritten as

$$H=\sum_k \epsilon_k a_k^\dagger a_k+[(\varepsilon^2+\Delta^2)^{1/2}/2]\sigma^z + (D\sigma^z+M\sigma^x)S^x \ , \qquad (5)$$

where

$$D=G\varepsilon/(\varepsilon^2+\Delta^2)^{1/2} \ , \qquad (6a)$$

$$M=G\Delta/(\varepsilon^2+\Delta^2)^{1/2} \ . \qquad (6b)$$

Equations (6a) and (6b) give the analytical formula for M and D for which we determine their temperature dependence.

The resonance frequency is expected to be shifted in the ferromagnetic resonance experiment. That gives a particular contribution to the linewidth and lineshift. We do not intend to present all details of the calculations that are reported in Ref. 2. The final formulas for the temperature dependence of the linewidth and line shift, after Weissenberger, Elschner, and Continentino,[3] have two components:

$$\Gamma=\Gamma_1+\Gamma_2 \ , \qquad (7)$$

$$\delta H=\delta H_1+\delta H_2 \ . \qquad (8)$$

Subscripts 1 and 2 correspond to components which include coupling parameters M and D, respectively.

Using the results of Weissenberger, Elschner, and Continentino[3] for the linewidth and line shift, one obtains

$$\Gamma_1=[2\pi n_0 M^2/(\hbar\gamma N_0)]\tanh[\hbar\omega/(2k_B T)] , \quad (9)$$

$$\Gamma_2=[4\pi n D^2/(\hbar\gamma N_0)][k_B T/V_0]\exp(-T/T_0)] , \quad (10)$$

$$(\delta H)_1=[2n_0 M^2]/(\hbar\gamma N_0)\ln(T/T^*) , \quad (11)$$

$$(\delta H)_2=[4nD^2/(\hbar\gamma N_0)]\exp(-T/T_0), \quad (12)$$

where N_0 is a number of magnetic ions per unit volume, T_0 is a temperature of the linewidth maximum, n_0 is the density of the magnetic tunneling states (TLS) per unit volume and unit energy, n is the distribution of an energy level.

The parameter n needs some comments; $n=N/V$, V is the barrier height, N can be found from the fraction of spins in finite clusters over those in the infinite cluster.

The two-level system (TLS) introduced by Anderson, Halperin, and Varma[8] is characterized by the height of the potential barrier V between metastable states of TLS. The formula for the barrier height $V(T)$ should reflect the temperature dependence of the source of the barrier. We assume that the unidirectional magnetic anisotropy energy constant K attributed to the spin-glass state is related to the asymmetric double-well potentials. So it is important to have access to a proper experiment which gives the temperature dependence of K. Since we have a strict formula (from the position of the resonance field for parallel geometry), for the unidirectional field δH [see Eq. (3)] we can find the character of the temperature dependence of $\delta H(T)$ (see Fig. 2) and the values of the unidirectional anisotropy constant $4\pi K$ ($K=M_s\delta H$).

Figure 4 presents the temperature dependence of $4\pi K$. As is seen in Fig. 4, there is a linear relationship between $4\pi K$ and T. The results are in agreement with those reported in Ref. 12. In our numerical calculation we used the following formula for the barrier height $V(T)$:

$$V(T)=V_0(1-T/T^*) . \quad (13)$$

The temperature dependence of V was also considered in Refs. 13 and 14. Table I presents the values of parameters used for computing M and D. The value of V_0 was in some sense chosen arbitrarily for the sample with REE. It is the same as V_0 in Refs. 2 and 3 because, according to the phase diagram, the magnetic state of our sample is identical to that of the specimens studied there. For the other two samples the value of V_0 was obtained from the estimation of $K_{(\mathrm{REE})}/K_{(\mathrm{SG})}$ for $T=0$ (see Fig. 4). The N_0 was calculated from the well-known formula

$$N_0=M_s/(g\mu_B S) , \quad (14)$$

where S is the spin of the magnetic atom (for Cr $S=\frac{3}{2}$).

The density of the magnetic tunneling states n_0 was taken after the literature data.[15,16] As we see in Table I the highest value of n_0 is reached in a sample with REE transition and decreases while the dilution increases. Due to the validity of the model only data for $T \le T^*$ are taken into account.

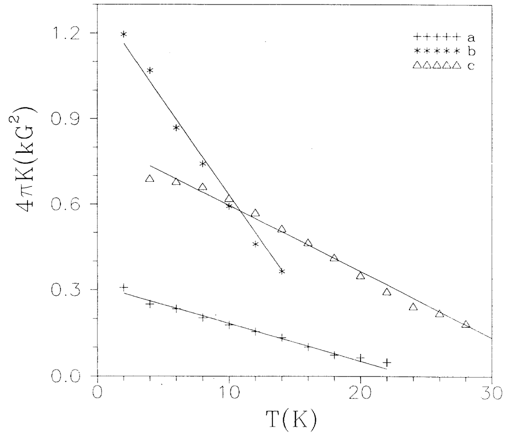

FIG. 4. Temperature dependence of $4\pi K$ for (a) $CdCr_{1.9}In_{0.1}Se_4$, (b) $CdCr_{1.7}In_{0.3}Se_4$, (c) $CdCr_{1.6}In_{0.4}Se_4$. The solid lines were determined from the linear regression procedure.

The distribution of energy levels n requires the information on the value of N/N_0 and we take the data from Ref. 3 in which, in our opinion, the properties of investigated samples are similar to ours.

Figures 5(a), 5(b), and 5(c) present the temperature dependence of coupling constants M and D. The results differ from those presented by Weissenberger, Elschner, and Continentino.[3] In Ref. 3 there is no visible temperature dependence of M and D, which is not surprising because the authors take into their calculation a constant value of the barrier height independent of the temperature. We suppose that our results are close to the real energetic state of TLS. For SG states at low temperature $D>M$; at least such a tendency could be seen. It means that the dominant interaction takes place between damped magnons and TLS. With an increasing temperature the damping effect is decreased so the main contribution to the interaction results from the resonance interaction between magnons and TLS. The values of $M(T)$ and $D(T)$ for chromium concentration $x=0.85$ (less diluted SG state) are the highest in comparison with other samples within the investigated temperature range. This

TABLE I. The value of parameters used for computing the coupling constants M and D.

Composition	n_0 [10^{22}/(eV cm^3)]	N_0 (10^{22}/cm^3)	n ($T=0$) [10^{22}/(eV cm^3)]	V_0 (eV)	T_0 (K)
$CdCr_{1.9}In_{0.1}Se_4$	16	1.2	36	0.01	13
$CdCr_{1.7}In_{0.3}Se_4$	13	0.7	9	0.04	10
$CdCr_{1.6}In_{0.4}Se_4$	10	0.5	12	0.03	22

could be explained as being related to the influence of magnons of IFN on the interaction between FC (indirect interaction). The value of the $M(T)$ and $D(T)$ is the smallest for the REE sample within the temperature range from 2 to 15 K. This sample is near the ferromagnetic threshold.[4] So the relevant excitations are the spin waves and the interactions which are described by M and D are weak.

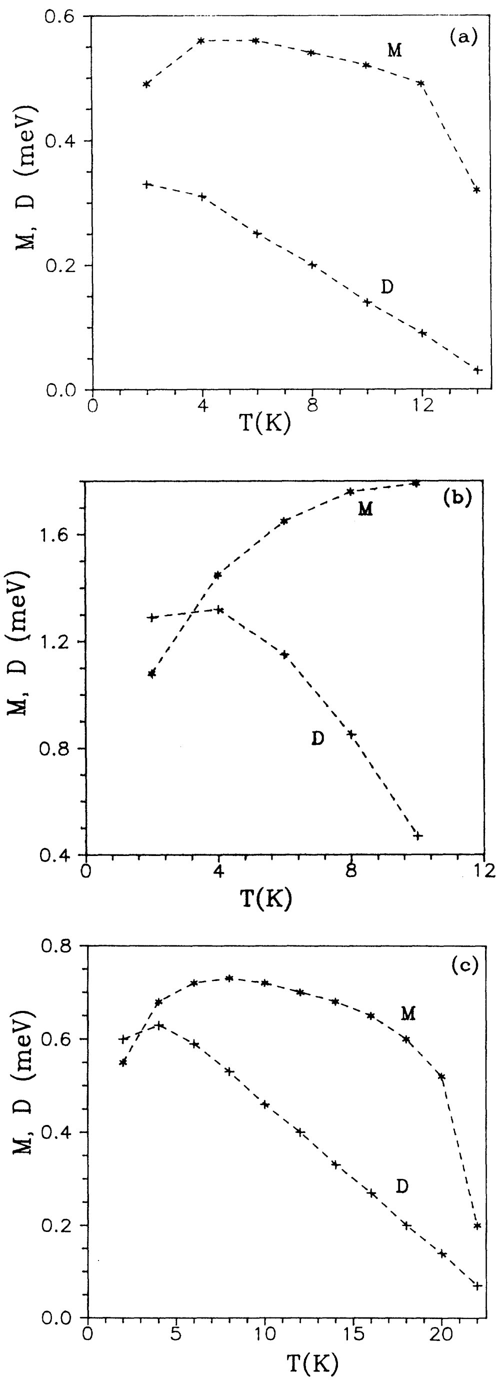

FIG. 5. The coupling constants M and D vs temperature for (a) $CdCr_{1.9}In_{0.1}Se_4$, (b) $CdCr_{1.7}In_{0.3}Se_4$, (c) $CdCr_{1.6}In_{0.4}Se_4$. The dashed line through the points is a guide for the eye.

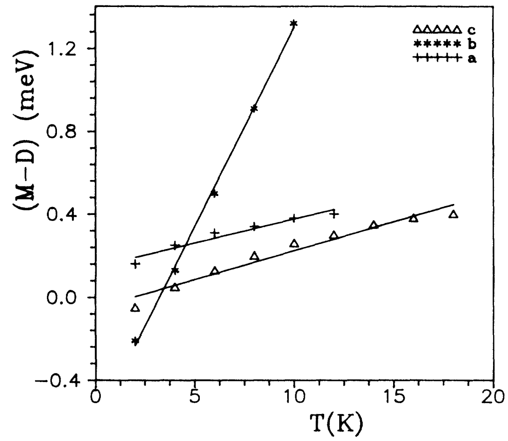

FIG. 6. Variation of $(M-D)$ with temperature for (a) $CdCr_{1.9}In_{0.1}Se_4$, (b) $CdCr_{1.7}In_{0.3}Se_4$, (c) $CdCr_{1.6}In_{0.4}Se_4$. The solid lines were determined from the linear regression procedure.

Figure 6 presents the temperature dependence of $(M-D)$ for three investigated samples. The value of $M-D$ can be treated as a measure of interaction between nondamping magnons and TLS. The linear relationship between $(M-D)$ and T is seen. We expected that this dependence should reproduce the results of the temperature dependence of the barrier height but the slope of $V(T)$ is negative. We also expect that in FMR experiment in the frequency range from 3 to 36 GHz, for example, the frequency dependence of M and D could be an extra source of information due to the wide range of magnon energies.

IV. CONCLUSIONS

The coupling between IFN and FC as well as the influence of magnons on the coupling were discussed in this paper. Investigated samples of $CdCr_{2x}In_{2-2x}Se_4$ with $0.8 \leq x \leq 0.95$ have the temperature dependence of linewidth that can be fitted quite well to the universal plot $\Delta T/\Gamma_1$ vs $(T/T_0)\exp(-T/T_0)$ which is valid for a large number of alloys with randomly located spins.[6,17] It is therefore suggested to apply the models presented by Anderson, Halperin, and Varma[8] and Continentino,[2] for SG materials. Moreover, the temperature dependence of the saturation magnetization confirms that according to the phase diagram, our samples belong to the class of materials in REE and SG.

The models of alloys with randomly located spins predict two coupling parameters M and D defined by Eqs. (6a) and (6b). In order to determine these parameters we applied the ferromagnetic resonance technique. The relaxation processes are sensitive to the microscopic interactions. They manifest themselves by the value of resonance linewidth. We modified the assumptions of applied models taking into account the temperature dependence of the barrier height which characterized the energetic state of asymmetric double-well potentials. The results for M and D in Eqs. (9)–(12) are also temperature dependent as is demonstrated in Figs. 5(a), 5(b), and 5(c).

The results we obtained could be summarized as follows:

(i) Unidirectional magnetic anisotropy is a linear function of temperature (see Fig. 4) and also has the highest value for the sample with $x = 0.85$ and decreases while dilution level decreases. It remains in agreement with the model of diluted spins.

(ii) We related the height of potential barrier V to this unidirectional anisotropy. So the temperature dependence of V [see Eq. (13); $V = V_0(1 - T/T^*)$] is concluded to be similar to the temperature dependence of $4\pi K$.

(iii) M and D are temperature dependent and are sensitive to the dilution level. Also the maximum value of M was obtained for a sample with $x = 0.85$ in the wide temperature range. It corresponds to SG but less diluted. This may be explained by the magnons that influence the inter-TLS's interaction.

(iv) We introduce an extra factor $(M - D)$ of the interaction which describes the contribution to the interaction from nondamped magnons only. Since the damping effect is strongly related to the height of the barrier,[2] the $(M - D)$ is a linear function of T, with the positive slope.

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