

# Preparation and magnetic interactions in $\text{Cd}_{1-x}\text{Zn}_x\text{Cr}_2\text{Se}_4$ spinel

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$\text{Cd}_{1-x}\text{Zn}_x\text{Cr}_2\text{Se}_4$  is a compound with spinel structure where two important frustration effects are present. The disorder in distribution of exchange interactions is due to the substitution between zinc and cadmium on tetrahedral sites. The samples in powder form were performed by the classical ceramic method. For the crystal growth, different methods were used depending on Zn and Cd concentrations. The composition was determined by X-ray measurements of the lattice constant. The direct current susceptibility measurements show that for the whole concentration range the system obey to Curie-Weiss law at high temperature. The DC magnetization, AC susceptibility and thermoremanent magnetization measurements revealed the existence of a spin-glass-like state and reentrant spin-glass behavior at low temperatures, in the middle concentration range.

## I. Introduction

$\text{CdCr}_2\text{Se}_4$  is a ferromagnetic semiconductor with a Curie temperature  $T_C \approx 130$  K [1].  $\text{ZnCr}_2\text{Se}_4$  is an insulating helimagnet with a first order transition  $T_1 = 21$  K at which the helimagnetic macro-domains break into metamagnetic micro domains. The high temperature structure is centered tetragonal with  $T_N = 45.5$  K [2]. The magnetic order, in  $\text{CdCr}_2\text{Se}_4$  and  $\text{ZnCr}_2\text{Se}_4$ , results from the competition of nearest neighbors positive Cr-Se-Cr interaction and more distant negative Cr-Se-A-Se-Cr (A=Cd, Zn) exchange interactions. The solid solution  $\text{Cd}_{1-x}\text{Zn}_x\text{Cr}_2\text{Se}_4$  is of fundamental interest for many basic experiments:

- (i) a bicritical point (Lifshitz point) is expected in the magnetic phase diagram;
- (ii) a transition helimagnetic  $\leftrightarrow$  ferromagnetic in the middle concentration range;
- (iii) a discontinuity in the galvanomagnetic properties.

On the other hand, due to the frustration effects in this system a spin glass like behavior is expected. Numerous experimental and theoretical studies [3-15] have been devoted to the system  $\text{Cd}_{1-x}\text{Zn}_x\text{Cr}_2\text{Se}_4$ . However, some contradictions still exist between the different results given in the literature. According to A. V. Myagkov et al, single crystals of the system  $\text{Cd}_{1-x}\text{Zn}_x\text{Cr}_2\text{Se}_4$  are ferromagnetic at  $x < 0.3$ , are antiferromagnetic at  $x > 0.5$  and are spin glasses at  $x \approx 0.4$  [7]. In the magnetic phase diagram deduced from susceptibility measurements no region with only paramagnetic- spin glass was found [8]. Using the mean field theory and the ferromagnetic spin-wave theory at low temperature, the exchange

integrals were calculated up to the third nearest neighbors [12]. However, the value of  $T_C$  given for  $\text{Cd}_{0.42}\text{Zn}_{0.58}\text{Cr}_2\text{Se}_4$  is not correct and it has never been published before. Also the values of  $\theta$  used in this work are not those published by Buch et al [4]. The purpose of the present study is to investigate the magnetic properties of  $\text{Cd}_{1-x}\text{Zn}_x\text{Cr}_2\text{Se}_4$  for  $0.41 \leq x \leq 0.65$ . It will be demonstrated that there is no experimental evidence to the existence of the spin glass state in the intermediate concentration range. Since in this concentration range the magnetic properties are very sensitive to slight changes in the composition, we present here a detailed description of the preparation and the composition analysis of the samples.

## II. Samples preparation and crystal analysis

Barracough's review [16], on crystal growth of ferromagnetic semiconductors, shows that there are many attempts to have single crystals of  $\text{CdCr}_2\text{Se}_4$ : chemical vapor transport; flux growth in  $\text{CdCl}_2$ , vapor-liquid transport techniques; the largest crystals obtained have about 4 mm size. On the contrary few attempts have been made to obtain  $\text{ZnCr}_2\text{Se}_4$  crystals; Von Neida and Shick [17] described a vapor-liquid transport technique and obtained large crystals (8 mm) of  $\text{ZnCr}_2\text{Se}_4$ ; one note that the nominal temperature for growth are  $950^\circ\text{C}$  and  $875^\circ\text{C}$  for  $\text{ZnCr}_2\text{Se}_4$  and  $\text{CdCr}_2\text{Se}_4$  respectively. A difference in the nominal temperature and in crystal size exists. Only one attempt to have mixed crystals  $\text{Cd}_{1-x}\text{Zn}_x\text{Cr}_2\text{Se}_4$  have been made by Aminov et al. [18] with a view to study the magnetic properties. To prepare the solid solution  $\text{Zn}_x\text{Cd}_{1-x}\text{Cr}_2\text{Se}_4$  we have tested four growing methods used for pure  $\text{CdCr}_2\text{Se}_4$  or  $\text{ZnCr}_2\text{Se}_4$ .

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### 2.1. Solution growth (S.G.)

#### (i) Traveling heater method:

A molten  $\text{CdCl}_2$  solvent zone, with 900 °C maximum at center, moves through solid  $\text{CdCr}_2\text{Se}_4$  material [19].

#### (ii) Crystallization of $\text{CdCr}_2\text{Se}_4$ from a solution by slow cooling:

Berkowski and Piekarczyk [20] have determined the  $\text{CdCr}_2\text{Se}_4$ - $\text{CdCl}_2$  phase diagram; the best conditions of crystal growth from a  $\text{CdCl}_2$  flux are: mixture of 88 %  $\text{CdCl}_2$  mole and 12 %  $\text{CdCr}_2\text{Se}_4$  mole with starting temperature higher than 880 °C (liquidus temperature) and a cooling rate of 2 °C.h<sup>-1</sup> down to 542 °C. Berkowski obtained crystals of 2 to 4 mm size.

### 2.2. High temperature solution (H.T.S.)

In this technique, given by Larson [21], the mixture  $\text{CrCl}_3 + 2 \text{MSe}$  (M = Cd or Zn) is slow cooled from 950 °C, crystals growing in the molten mixture. Aminov [18] used this technique for growing Zn-Cd mixed crystals (950 to 700 °C in 5 days) and obtained 3 mm large crystals.

### 2.3. Chemical vapor transport (C.V.T. or C.V.D.)

Many transport agents have been used [16]. 4 mm large crystals of  $\text{CdCr}_2\text{Se}_4$  have been grown using  $\text{HCl} + \text{Cl}_2$  (molecular ration ~ 1) as transport components and with a temperature gradient between 1100 - 1000 °C [22]. For Cd-Zn mixed crystals, we used  $\text{Cl}_2 + \text{few HCl}$  with a temperature gradient between 850 - 800 °C.

### 2.4. Vapor - Liquid transport (V.L.T.)

For the first time Von Neida and Shick [17] have grown crystals of  $\text{ZnCr}_2\text{Se}_4$  and  $\text{ZnCr}_2\text{S}_4$ . ZnSe (or CdSe) and  $\text{CrCl}_3$ , in the ration 2:1 respectively, are separately cold pressed to pellet shape and placed at one end of a sealed silica vial. A gradient is established such that the tip of the tube is about 50 °C cooler than the region containing the reactants; the furnace is inclined 20 % to horizontal to promote collection of a liquid which form at the tip of the vial. Von Neida obtained imperfect 4 mm  $\text{CdCr}_2\text{Se}_4$  and 8 mm  $\text{ZnCr}_2\text{Se}_4$  crystals (in 4 days).

We have used mainly the two last techniques and have obtained large crystals of  $\text{ZnCr}_2\text{Se}_4$  (around 10 mm size) and of Zn-rich materials, but small crystals of Cd-rich compounds.

From figure 1, we see that the lattice constant of  $\text{Cd}_{1-x}\text{Zn}_x\text{Cr}_2\text{Se}_4$  system varies with the composition in accordance with the Vegard's law. The parameter is used to estimate the crystal

composition (error ~ 0.01 cation); some samples have been analyzed by electron microprobe.

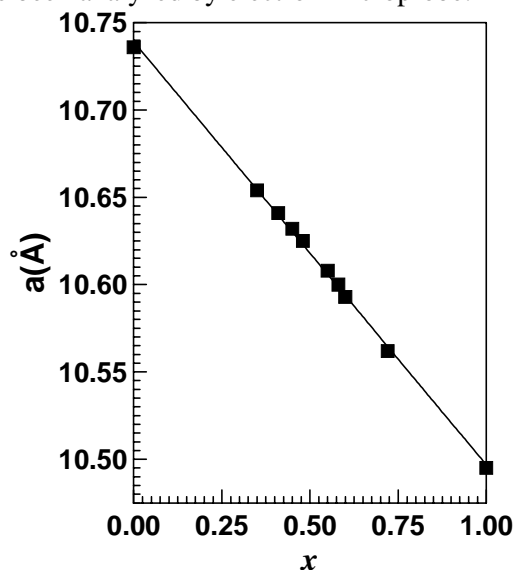


Figure 1: Variation of the spinel parameter  $a$  with the concentration of zinc in  $\text{Cd}_{1-x}\text{Zn}_x\text{Cr}_2\text{Se}_4$  solid solution.

## III. Results and data analysis

### 3.1. Direct current magnetic susceptibility

The direct current magnetic susceptibility  $\chi_{d.c}$  was measured by a DMS 4 magnetometer in the temperature range 300 - 600 K.

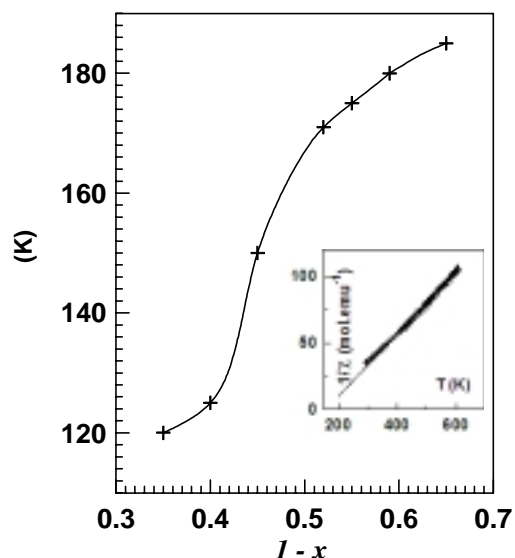


Figure 2: Variation of the Curie paramagnetic temperature  $\theta$  with the concentration  $x$  of zinc. Inset: variation of the reciprocal magnetic susceptibility  $\chi^{-1}$  of  $\text{Cd}_{0.45}\text{Zn}_{0.55}\text{Cr}_2\text{Se}_4$  as function of the temperature.

The temperature dependence of  $\chi_{d.c}$  was studied in order to determine the paramagnetic Curie temperature  $\theta$ . For all the samples  $\chi_{d.c}$  obeys a Curie Weiss law at high temperature. The least squares fit of the data to  $\chi = C / T - \theta$  yields the values of the Curie-Weiss temperature  $\theta$  presented

in Fig. 2 and the Curie Weiss constant  $C = 1.8 \pm 0.1 \text{ emu.K.mol}^{-1}$ . The effective magnetic moment  $p_{\text{eff}}$  is calculated from the value of  $C$  and the value obtained is  $p_{\text{eff}} = 3.9 \pm 0.2$  in good agreement with  $p_{\text{eff}} = 3.87$  calculated for the spin only state. The positive values of  $\theta$  indicate that the interplanar exchange interaction is ferromagnetic. The deviation of data from a straight line exhibiting the Curie-Weiss law becomes appreciable at low temperature, implying the growth of short range spin order. No linear variation of  $\theta$  is detected for the concentration range  $0.35 \leq x \leq 0.72$  in opposition of Busch et al work [4].

### 3.2. Magnetization measurements

The magnetization measurements were performed with a vibrating sample magnetometer in the temperature range 4.2 to 300 K. Two measurements procedures were used to obtain the behavior of  $M(T)$ . The sample is first cooled in zero field from the room temperature to 4.2 K. Next the measurement field is applied and the temperature is then increased and the zero-field-cooled (ZFC) curve is recorded. The temperature is next reduced without changing the measurement field, then increased again and the field cooled (FC) curve is recorded. Figure 3 displays the magnetization, with an applied field  $H_a = 0.5 \text{ mT}$ , vs. the temperature, both after cooling in zero field and after cooling in a field.

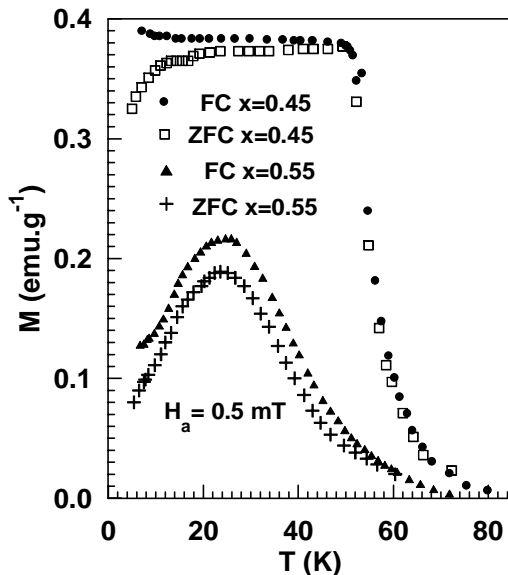


Figure 3: ZFC and FC magnetization curves for  $\text{Cd}_{0.55}\text{Zn}_{0.45}\text{Cr}_2\text{Se}_4$  and  $\text{Cd}_{0.45}\text{Zn}_{0.55}\text{Cr}_2\text{Se}_4$  with an applied field  $H_a = 0.5 \text{ mT}$ .

In the concentration range  $0.41 \leq x < 0.5$  the magnetization is reversible above the Curie temperature  $T_C$ . The decrease of the magnetization

at low temperature indicates the existence of reentrant transition to a spin glass state where no spontaneous magnetization is present.

In figure 4, we have plotted the magnetization data in the case of  $x = 0.48$  for different applied fields.

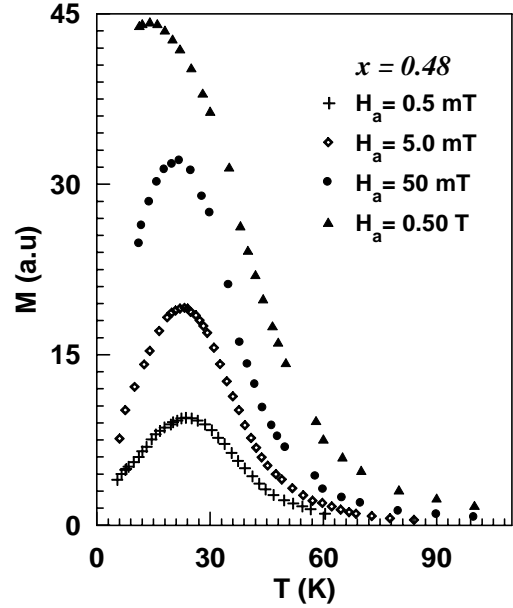


Figure 4: Magnetization curves for  $\text{Cd}_{0.52}\text{Zn}_{0.48}\text{Cr}_2\text{Se}_4$  at different applied fields.

For this sample, the measurements of the static magnetization in increasing applied fields produced  $M(H_a, T)$  curves that bears striking resemblance to the curves of figure 5 [23] which present the solution of the Sherrington-Kirkpatrick model [24] in finite field.

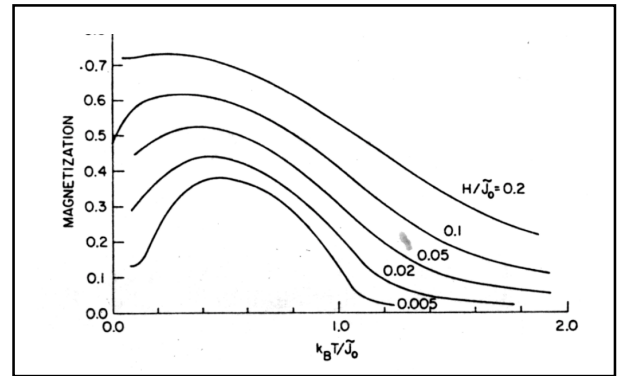


Figure 5: Solution of the Sherrington-Kirkpatrick model for the magnetization in finite fields [23]

For  $0.5 < x \leq 0.65$ , a different behavior is observed. The  $M$  vs.  $T$  curves present a broad maximum at  $T_m$ . For  $H_a \geq 5 \text{ mT}$  ZFC-FC values are identical above the magnetization maximum temperature  $T_m$ . The ZFC and FC magnetization measurements seem to demonstrate strong irreversibilities as in spin-glass state. The irreversibilities are field dependent and disappear at high field values.

However, the difference between ZFC-FC curves can be explained by the existence of hysteresis loop if  $H_a$  is less than the coercive field. This behavior cannot be related to a spin glass regime as previously proposed by A.V. Myagkov et al [7]. It is well known that the irreversibilities, metastabilities and time dependences of the frozen state are not sufficient to check out the existence of a true spin glass state. The random freezing of spins in a SG state further results in a divergence of the non-linear susceptibility which related to the spin glass order parameter. On the other hand, it must be noted that the irreversibility between the ZFC and FC curves can be observed in ordered ferromagnetic materials if the coercive field  $H_c$  increases strongly at low temperature, but in this case the ZFC magnetization is not time dependent. The behavior of the magnetization in this concentration range supports a cluster spin freezing below  $T_m$ . The antiferromagnetic interactions compete with the ferromagnetic interactions and freeze the clusters below  $T_m$ . This resembles a reentrant magnetic behavior [25].

When  $0.7 < x \leq 1$ , the temperature dependence of low field magnetization exhibits a typical antiferromagnetic behavior according to previous works [3-8].

### 3.3. Alternative current susceptibility

The alternative current susceptibility  $\chi_{ac}$  measurements are the most useful to show the dynamical properties of the disordered systems. To check out the existence of a reentrant spin glass transition at low temperature we performed out alternative current susceptibility measurements, in the concentration range  $0.4 < x < 0.5$ , at a fixed frequency  $f = 2.5 \cdot 10^{-2}$  Hz and an applied field  $H_a = 1.3 \cdot 10^{-4}$  T. Typical examples obtained for  $\text{Cd}_{0.55}\text{Zn}_{0.45}\text{Cr}_2\text{Se}_4$  and  $\text{Cd}_{0.52}\text{Zn}_{0.48}\text{Cr}_2\text{Se}_4$  are presented in Figure 6.

The behavior of  $\chi_{ac}$  can be described as follow: After the PM-FM transition, in the variation of  $\chi'_{ac}$  (real component) a plateau is observed revealing the effect of a demagnetizing field. The  $T_C$  temperature can be easily deduced. For increasing  $x$ ,  $\chi'_{ac}$  becomes differently and does not present an abrupt decrease near  $T_C$ , showing that the FM order begin to be perturbed. At lowest temperature a rapid decrease of  $\chi'_{ac}$  is observed, probably due to the disappearance of long range ferromagnetic order. For  $\chi''_{ac}$  (imaginary component) we note the presence of two peaks one relatively sharp at low temperature, the second very broad at high temperature. The susceptibility data point to existence of spin glass

like state at low temperature. The trend with the composition  $x$  reveals that the introduction of zinc ions, reinforcing the relative strength of the higher order neighbors antiferromagnetic interactions with the respect n.n. ferromagnetic one, destabilizes the ferromagnetic order, which remains, strongly perturbed. The reduction of the strength of the ferromagnetic interactions when Zn ions are introduced is clearly reflected by the decrease of the magnitude of  $\chi'_{ac}$ .

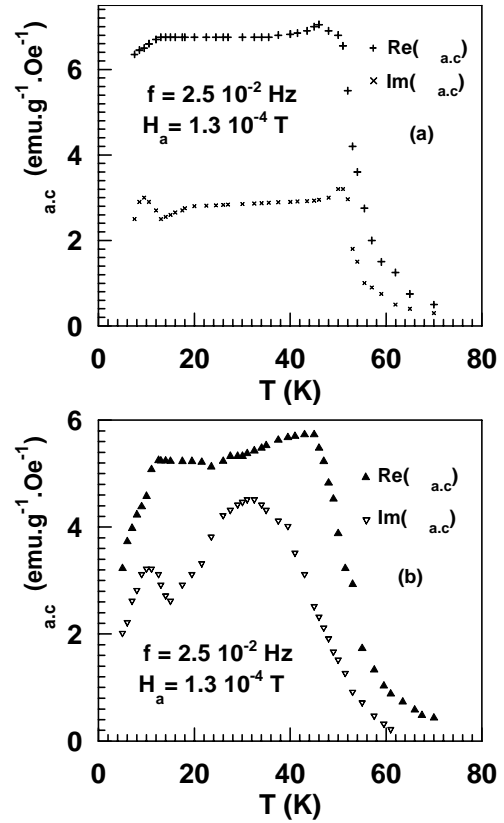


Figure 6: Temperature dependence of  $\chi_{ac}$ . (a)  $x = 0.45$ , (b)  $x = 0.48$ .

### 3.4. Thermoremanent magnetization

Thermoremanent magnetization  $TRM(T, t)$  have been performed on the sample  $\text{Cd}_{0.45}\text{Zn}_{0.55}\text{Cr}_2\text{Se}_4$  for fields 1 and 10 mT at some temperatures in the range 4.2 – 25 K. The sample was first cooled from room temperature until the measurement temperature  $T_{mes}$ . At  $T_{mes}$ , the sample is kept at constant temperature for a wait time  $t_w = 1200$  s. Then the field was switched off and the  $TRM(T, t)$  measured during 1800 s. The variation of  $TRM(T, t)$  vs.  $t$  can be fitted with the phenomenological logarithmic decay function:

$$TRM(T, t) = C(T) \exp[-p(T) \ln(t)]$$

$p(T)$  increases with the temperature as in spin glass regime, shows a maximum value at  $T_s \approx 8$  K, then decreases rapidly until  $T = 23$  K where  $TRM(T, t)$  keeps an almost constant value with time. The decrease of  $p(T)$  is not usually observed. This

behavior can be explained by the Khater relation [26] giving  $TRM(T,t)$  from a cluster model. In this model the disorder introduced by the transverse spin component  $S_t$  could explain the low temperature  $TRM(T,t)$  behavior, while at high temperature the relaxation is fast and the order evolves toward a ferromagnetic regime.

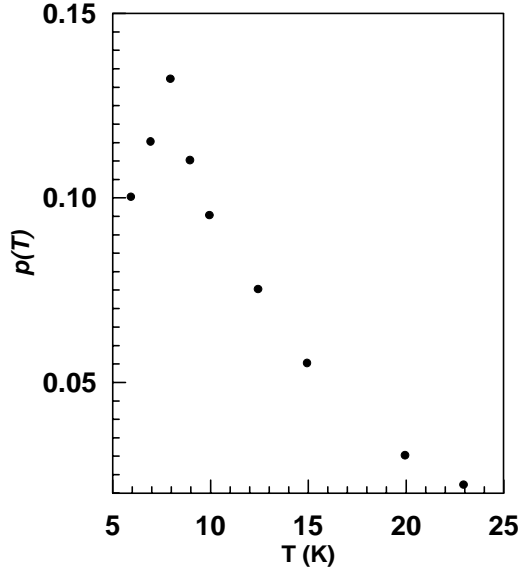


Figure 6: Temperature variation of the slope  $p(T)$  of  $\text{Ln}(TRM(T,t))$  vs  $\text{Ln}t$  curve.

#### IV. Discussion

From the measurements it is clear that  $x \leq 0.4$  the system orders ferromagnetically, while for  $0.41 \leq x \leq 0.50$  this order becomes to be perturbed and at low temperature a new transition occurs and some properties are similar to those of spin glasses. The  $\chi_{a.c}$  susceptibility trends to confirm the existence of reentrant spin glass transition. This RSG transition has been interpreted coherently within the framework provided by the transverse spin component freezing based on the molecular field theory [23,27,28].

The infinite range Sherrington-Kirkpatrick mean field theory of Ising SG systems, which is based on Edwards-Anderson magnetic frustration model, has been extended by Gabay and Toulouse for vector SG systems [28]. The phase diagram shows the existence of various magnetic phases: SG = spin-glass, F = ferromagnetic (ferri-/antiferromagnetic) phase, M1 = mixed phase 1 (M1), M2 = mixed phase 2 (M2), P = paramagnetic-phase. Thus whereas in SG and M2 phases the spin components are randomly frozen isotropically, in M1 phase only transverse spin components are randomly frozen. In the mean field SG theory, this random direction freezing of spins (SG freezing) is explained by assuming that due to the magnetic frustration in the lattice the exchange integral  $J_{ij}$ , for interaction between two spins  $S_i$  and  $S_j$  does

not have a unique value but has a Gaussian distribution and the probability  $p(J_{ij})$  for any  $J_{ij}$  value is given by,

$$p(J_{ij}) = \frac{1}{J\sqrt{2\pi}} \exp\left(-\frac{(J_{ij} - J_0)^2}{2J^2}\right)$$

where  $J_0$  is the mean field of the distribution and  $J$  is proportional to  $\Delta$  the width of the distribution.

It is not easy to relate  $J_0$  and  $J$  to any parameters which characterize the samples being studied experimentally. The moments interact through indirect exchange in this system and exchange interactions vary in sign and magnitude with intermoment distance. At low temperature, each moment points in a direction determined by several of its neighbors. Since a spin's equilibrium direction is a compromise among the random signals from several neighbors, it need not be the same as that of any other spin. Thus at low temperatures each moment's direction is fixed, but the net magnetization is zero. Finally we can assume that ferromagnetism is possible for  $x < 0.5$  as shown by the magnetization measurements. For this concentration range upon cooling, the samples pass from a PM to FM and at lower temperatures to a SG state. This is in qualitative agreement with the SK model. A drop in the ac susceptibility from its demagnetization limit is believed to be the result of a reentrant ferromagnetic-spin-glass transition. Direct measurements of the magnetization show that at low temperatures the magnetization diminishes with decreasing temperature. The existence of a ferromagnetic state has been previously checked out using scaling analysis [11,29].

For  $x > 0.5$  the measurements do not prove the existence of re-entrant state. The ferromagnetic phase (F) provided in the Gabay-Toulouse phase diagram is not well established. In this concentration range small groups of moments can interact directly. These small magnetic clusters interact via indirect exchange and can, upon cooling, freeze with random orientations. The frozen states described above have been given several names, depending on the moment concentration. Here we use the term "spin glass like state" with frozen moments but zero spontaneous magnetization.

With increasing  $x$  the system becomes more perturbed like in a local canted state. The critical analysis performed on the sample  $\text{Cd}_{0.45}\text{Zn}_{0.55}\text{Cr}_2\text{Se}_4$  shows that the paramagnetic-perturbed ferromagnetic phase transition is described by a non-classical set of critical exponents [11]. No transition from paramagnetic to spin glass state is observed for the whole composition range. The system can be described very well as a mixed phase where the

random and competition are not sufficient to lead to a spin glass state. The corresponding state may be considered as intermediate between a reentrant ferromagnetic and a true spin glass. The decrease of magnetization at low temperature indicates that samples behave like spin glasses in agreement with previous results [6].

With increasing  $x$  ( $0.6 < x \leq 0.65$ ) the magnetization curves suggest the existence of very perturbed magnetic phase domain: perhaps weak ferromagnetism or almost independent ferromagnetic clusters. The measurements performed on the samples in this concentration range show the existence of coercive field and remanent magnetization until  $T \approx 20$  K.

Hysteresis loop, thermoremanent magnetization measurements and alternative current susceptibility in presence of direct current field are in good progress for the concentration range  $0.55 \leq x \leq 0.65$  in order to establish an accurate experimental magnetic phase diagram.

### V. Conclusion

A detailed magnetic study has been performed on the  $\text{Cd}_{1-x}\text{Zn}_x\text{Cr}_2\text{Se}_4$  spinel system, which is an example of a frustrated ferromagnetic system.

(i)  $0 \leq x \leq 0.4$ : The compounds order ferromagnetically. The magnetic long-range order is established just above the Curie temperature  $T_C$ .

(ii)  $0.41 \leq x < 0.5$ : the ferromagnetic order seems to be lightly perturbed. At low temperature the properties are relevant to reentrant phenomena. A second transition is observed and related to freezing of transverse component of spin.

(iii)  $0.5 < x < 0.6$ : the magnetic long-range order is more and more perturbed with increasing the zinc concentration. The spin directions become spatially distributed and the properties prevent the development of mixed phases state rather than a true spin-glass phase.

(iv)  $0.6 < x < 0.7$ : a very perturbed magnetic phase domain occurs, probably weak ferromagnetism or almost independent ferromagnetic clusters.

(v)  $0.7 < x \leq 1$ : the system becomes helimagnetic.

### References

- [1] P. K. Baltzer, M. Robbins and P. J. Wojtowicz, J. Appl. Phys. **38**, 953 (1967).
- [2] R. Plumier, M. Lecomte, A. Miédan-Gros and M. Sougi, Phys. Rev. Lett. **55**, 239 (1975)
- [3] F. K. Lotgering, J. Phys. Chem. Solids **29**, 699 (1968).
- [4] G. Busch, B. Magyar and O. Vogt, Solid State Comm. **7**, 509 (1969).
- [5] V. E. Makhotkin, V.G. Veselago and V. Kalinnikov, Sov. Phys. Solid State **20**, 777 (1978).
- [6] R. A. Sadykov, P. L. Gruzin, A. A. Minakov, T. G. Aminov, V. G. Veselago, V. T. Kalinnikov and V. E. Makhotkin, Pis'ma Zh. Eksp. Teor. Fiz. **28**, 596 (1978).
- [7] A. V. Myagkov, A. A. Minakov and V. G. Veselago, J. E. T. P. Lett. **41**, 40 (1985); A. V. Myagkov, A. A. Minakov and V. Veselago, Sov. Phys. J.E.T.P **65**, 983 (1987).
- [8] M. Nogues and J.L. Dormann, J. Magn. Magn. Mater., **54-57**, 87 (1986).
- [9] M. Hamdoun, A. Zerhouni, A. Rachadi, H. Lassri and M. Nogues, Physica B **229**, 96 (1996).
- [10] M. Hamedoun, A. Hormatallah, A. Zerhouni, N. Benzakour, A. Belayachi and M. Nogues, Phys. Stat. Sol. (b) **203**, 521 (1997).
- [11] A. Belayachi, J. L. Dormann and M. Nogues, J. Phys. Condens. Matter **10**, 1599 (1998).
- [12] N. Benzakour, M. Hamedoun, M. Houssa, A. Hormatallah and F. Mahjoubi, Phys. Stat. Solidi B **212**, 335 (1999).
- [13] F. Mahjoubi, M. Hamedoun, F.Z. Bakkali, A. Hormatallah, A. Benyoussef, J. Magn. Magn. Mater. **221**, 359 (2000).
- [14] K. Afif, A. Benyoussef, M. Hamedoun and A. Hormatallah, Phys. Stat. Solidi A **171**, 571 (1999).
- [15] K. Afif, A. Benyoussef, M. Hamedoun and A. Hormatallah, Phys. Stat. Solidi B **219**, 383 (2000).
- [16] K. G. Barraclough, Prog. Crystal Growth Charact. **1**, 57 (1977) and citations included.
- [17] A. R. Von Neida and L. K. Shick, J. Appl. Phys. **40**, 1013 (1969).
- [18] T. G. Aminov et al, Izv. Akad. Nank SSSR, neorg. mater. **13**, 1300 (1976).
- [19] N. Hemmat et al, Proc. ACCG, Conf. on Crystal Growth, p. 79 (1969).
- [20] M. Berkowski and W. Piekarczyk, J. Crystal Growth **40**, 253 (1977).
- [21] G. H. Larson and A. W. Sleight, Phys. Letters **28 A**, 203 (1968).
- [22] P. Gibart, L. Goldstein, J. L. Dormann and M. Guittard, J. Crystal Growth, **24/25**, 147 (1974).
- [23] D. Sherrington and S. Kirkpatrick, Phys. Rev. Lett., **35** (1975) 1792
- [24] M.B. Salamon, K. V. Rao, and Y. Yeshurun, J. Appl. Phys. **52** (3) (1981) 1687
- [25] R.N. Bhowmik and R. Ranganathan, J. Magn. Magn. Mater., **247**, 83 (2002).
- [26] A. Khater, J. Ferré and P. Meyer, J. Phys. C: Solid State Phys. **20**, 1857 (1987).
- [27] J. R. L. de Almeida and D. J. Thouless, Phys. Rev. A **11**, 983 (1978)
- [28] M. Gabay and G. Toulouse, Phys. Rev. Lett. **47** 201 (1981).
- [29] A. Belayachi, Doctorat d'Etat, Rabat (1996).