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Influence of temperature on the critical fields in $ZnCr_{2-x}Al_xSe_4$ antiferromagnets

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ABSTRACT

The critical fields were investigated by the ac susceptibility $\chi_{ac}(B)$ and the magnetic moment $\mu(B)$ in the antiferromagnetic semiconductors of the $ZnCr_{2-x}Al_xSe_4$ spinel system in the compositional range $0.13 \le x \le 0.4$. The both dependences $\chi_{ac}(B)$ and $\mu(B)$ revealed two maxima at critical fields H_{c1} characteristic for metamagnetic threshold where the simple spin spiral transforms into conical magnetic structure and at H_{c2} where the conical magnetic structure transforms into ferromagnetic phase. With increasing temperature a peak at critical field H_{c2} shifts into lower values of the magnetic field while a peak at H_{c1} remains unchanged. These results point to a strong antiferromagnetic superexchange interaction in the range of magnetic ordering which does not significantly depend on temperature, Al content and magnetic field. The opposite behaviour in the case of the ferromagnetic exchange interaction was observed.

1. Introduction and experimental

Within the last decade the fascinating physics of spinels such as the gigantic Kerr rotation in $FeCr_2S_4$ [1], the colossal magnetoresistance in Cu doped $FeCr_2S_4$ [2], the multiferroic behaviour and colossal magnetocapacitive in $CdCr_2S_4$ and $HgCr_2S_4$ [3], and the large magnetostriction and negative thermal expansion in $ZnCr_2Se_4$ [3] have been observed. Previous neutron-diffraction investigations carried out on the p-type $ZnCr_2Se_4$ semiconductor revealed a complex antiferromagnetic order below 21 K with a strong ferromagnetic interaction evidenced by a large positive Curie–Weiss temperature of 115 K. The spin structure having a ferromagnetic arrangement in the (0 0 1) planes is dominated by the antiferromagnetic arrangement between the spins in adjacent (0 0 1) planes with a turning angle of 42° [4,5]. Recently, the structural and magnetic investigations of $ZnCr_{1.85}Al_{0.15}Se_4$ and $ZnCr_{1.77}Al_{0.23}Se_4$ single crystals [6] showed an antiferromagnetic phase transition at $T_N \approx 22$ K with Curie–Weiss temperatures of 108 and 114 K, respectively, as well as a metamagnetic transition at the critical field H_{c1} of about 15 kOe for $T = 1.72$ K, the breakdown of the conical spin arrangement at the critical field H_{c2} about 70 kOe for $T = 4.2$ K, and a decreasing saturation magnetic moment from 2.5 to 2.15μ_B per Cr atom, respectively, with increasing Al content.

Powder samples of the $ZnCr_{2-x}Al_xSe_4$ spinel series were obtained in the range $0.13 \le x \le 0.4$ using a ceramic method. The chemical composition of polycrystals was determined by wavelength-dispersive X-ray fluorescence spectrometry. The X-ray diffraction revealed a single-phase material with the cubic spinel structure (*Fd3m*). The ac susceptibility was measured at three different temperatures using a Lake Shore 7225 ac susceptometer up to 60 kOe in the range of magnetic ordering and at internal oscillating magnetic field $H_{ac} = 1$ Oe with internal frequency $f = 120$ Hz. The magnetic moment was measured with the aid of a step-magnetometer at 4.2 K and in applied external fields up to 140 kOe.

The present contribution reports the ac susceptibility and the magnetic moment measurements of the antiferromagnetic $ZnCr_{2-x}Al_xSe_4$ spinel system below the ordering temperature.

2. Results and discussion

Fig. 1 presents the ac magnetic susceptibility, χ_{ac}, vs. magnetic field H up to 60 kOe of the $ZnCr_{2-x}Al_xSe_4$ spinels with $x = 0.13$ (a), 0.22 (b), 0.33 (c) and 0.4 (d) at different temperatures and at internal oscillating magnetic field $H_{ac} = 1$ Oe with internal frequency $f = 120$ Hz. Below the Néel temperature T_N the χ_{ac} *(H)* shows a peak at the critical field $H_{c1} = 12.5$ kOe characteristic for metamagnetic transition where the simple spin spiral changes into conical magnetic structure. Both in the compositional range $0.13 \le x \le 0.4$ and in the range of magnetic ordering the values of H_{c1} remain almost constant. With increasing temperature the second critical field H_{c2} (where the conical structure changes into ferromagnetic phase) shifts into lower magnetic fields. Fig. 2 presents the mag-

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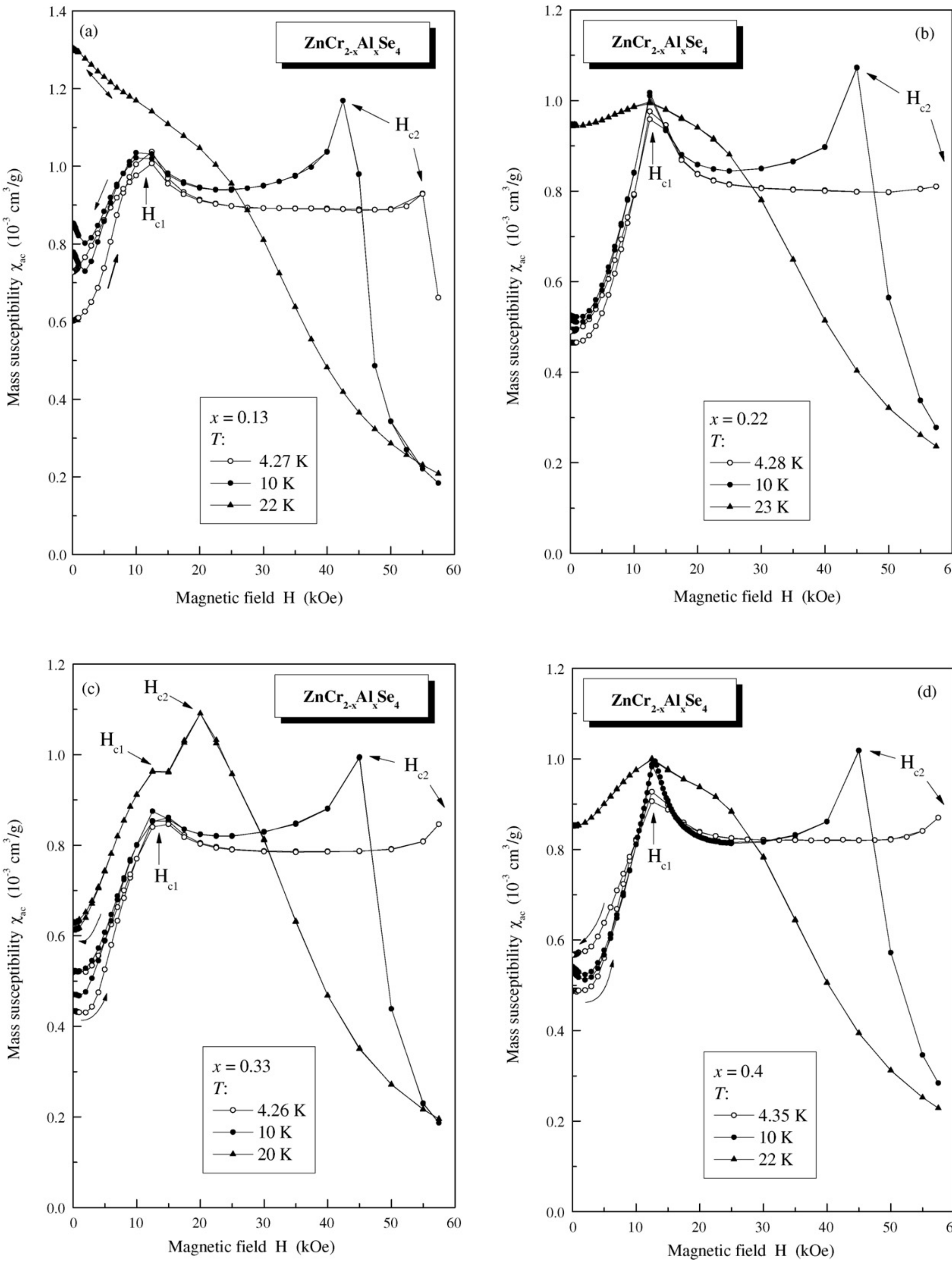

Fig. 1. AC mass susceptibility χ_{ac} vs. external magnetic field H of the $ZnCr_{2-x}Al_xSe_4$ spinels with $x = 0.13$ (a), 0.22 (b), 0.33 (c) and 0.4 (d) at different temperatures and at internal oscillating magnetic field $H_{ac} = 1$ Oe with internal frequency $f = 120$ Hz. The critical fields H_{c1} and H_{c2} are indicated by arrows.

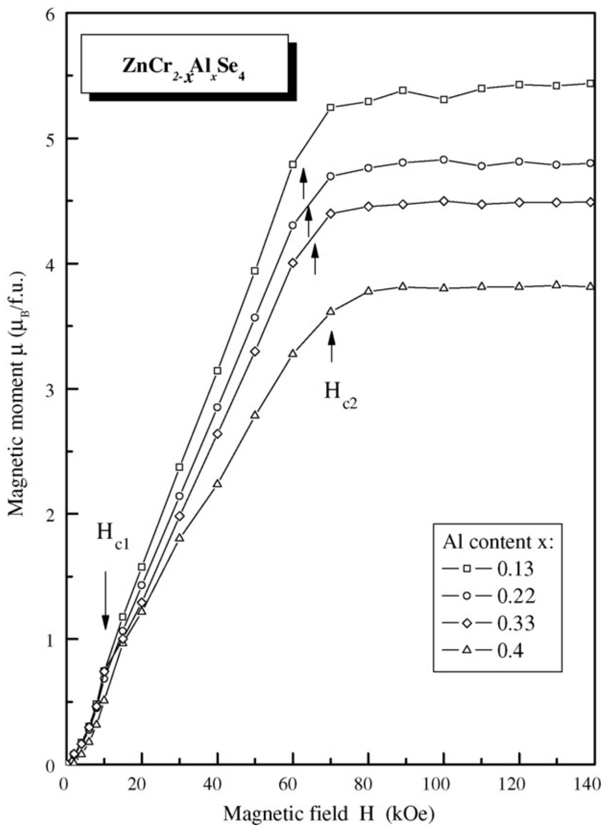

Fig. 2. Magnetic moment μ vs. external magnetic field H at $T = 4.2$ K of the $ZnCr_{2-x}Al_xSe_4$ spinel system with $x = 0.13$, 0.22, 0.33 and 0.4. The critical fields H_{c1} and H_{c2} are indicated by arrows.

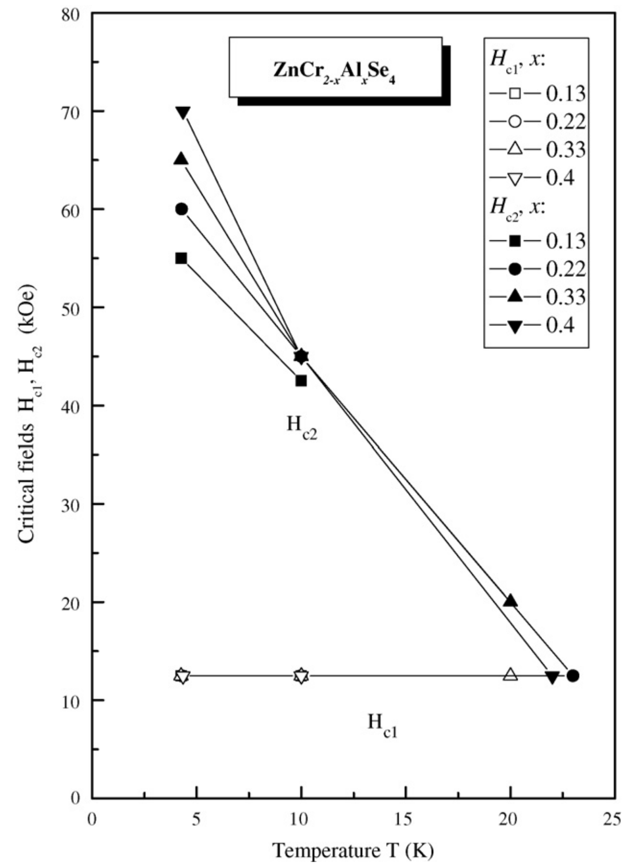

Fig. 3. The critical fields H_{c1} and H_{c2} vs. temperature T of the $ZnCr_{2-x}Al_xSe_4$ spinels with $x = 0.13$, 0.22, 0.33 and 0.4.

netic moment, μ, vs. magnetic field H up to 140 kOe. Here, the first critical field $H_{c1} = 12.5$ kOe is determined at the inflection point of the $\mu(B)$ curve and the second critical field H_{c2} – at the saturation point. The saturation magnetic moment μ_{sat} decreases from $5.35\mu_B$ for $x = 0.13$ to $3.83\mu_B$ for $x = 0.4$. For comparison, $\mu_{sat} = 5.74\mu_B$ at 4.2 K in pure $ZnCr_2Se_4$ spinel was observed [7]. A change of critical field vs. temperature is depicted in Fig. 3. From Figs. 2 and 3 it follows also that with increasing Al content x in a sample H_{c2} shifts into higher values below 10 K. In the vicinity of the ordering temperature $T_N \approx 23$ K both critical fields, H_{c1} and H_{c2}, disappear. The above mentioned results are connected with a dilution of the chromium sub-lattice by non-magnetic Al ions which play a role of spin defects in the completely filled majority spin t_{2g} orbital of the Cr^{3+} band. A shift of H_{c2} into lower magnetic fields with increasing temperature means that the short-range ferromagnetic (FM) interaction weakens while the long-range antiferromagnetic (AFM) superexchange interaction remains still strong because the position of peak at H_{c1} does not depend on temperature in the range of magnetic ordering.

In conclusion, a shift of H_{c2} is sensitive to temperature, spin defects and magnetic field. The behaviour of critical fields well correlates to the FM exchange and AFM superexchange interactions in the spinels under study.

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