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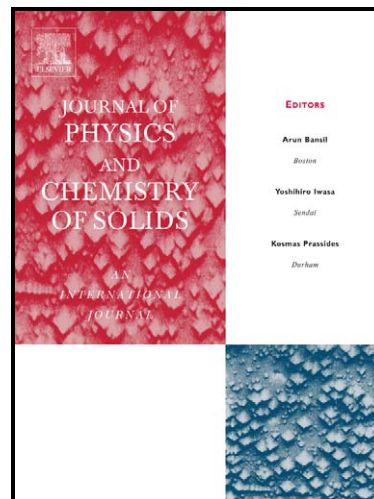
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Spin-glass-like behaviour and magnetic order in

Mn[Cr_{0.5}Ga_{1.5}]S₄ spinels

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Abstract

Magnetization and susceptibility were investigated as a function of temperature and magnetic field in polycrystalline Mn[Cr_{0.5}Ga_{1.5}]S₄ spinel. The dc susceptibility measurements at 919 Oe showed a disordered ferrimagnetic behavior with a Curie-Weiss temperature $\theta_{\text{CW}} = -55$ K and an effective magnetic moment of $5.96 \mu_{\text{B}}$ close to the spin-only value of $6.52 \mu_{\text{B}}$ for Cr³⁺ and Mn²⁺ ions in the $3d^3$ and $3d^5$ configurations, respectively. The magnetization measured at 100 Oe revealed the multiple magnetic transitions with a sharp maximum at the Néel temperature $T_{\text{N}} = 3.9$ K, a minimum at the Yafet-Kittel temperature $T_{\text{YK}} = 5$ K, a broad maximum at the freezing temperature $T_{\text{f}} = 7.9$ K,

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and an inflection point at the Curie temperature $T_C = 48$ K indicating a transition to paramagnetic phase. A large splitting between the ZFC and FC magnetizations at a temperature smaller than T_C suggests the presence of spin-glass-like behaviour. This behaviour is considered in a framework of competing interactions between the antiferromagnetic ordering of the $A(\text{Mn})$ sublattice and the ferromagnetic ordering of the $B(\text{Cr})$ sublattice.

Keywords: A. Spin glass; B. Ferrimagnetic ordering; C. Chalcogenide

1. Introduction

The MnCr_2S_4 parent is an insulating ferrimagnetic normal spinel at low temperatures with three different magnetic structures: Néel ferrimagnet, Yafet-Kittel and tridimensional with the Yafet-Kittel-Néel transition at $T_{\text{YK}} = 5.5$ K, the Curie-Weiss temperature $\theta_{\text{CW}} = 10$ K, the Curie temperature $T_C = 66$ K and the spontaneous magnetic moment per molecule of $1.27 \mu_B$ [1]. MnCr_2S_4 is a semiconductor [2] above 80 K for which the Mott's variable-range-hopping mechanism of conductivity is suggested [3] and shows the n - p phase transition at about 326 K [4]. Two λ anomalies observed in the heat capacity of MnCr_2S_4 are accompanied both with ferrimagnetic ordering at 65 K and an additional antiferromagnetic ordering of the $A(\text{Mn})$ sublattice at 4.8 K [3]. An electron-spin-resonance study of MnCr_2S_4 single crystals revealed a positive cubic

magneto-crystalline anisotropy below T_C [5]. The X-ray photoelectron spectra showed that in MnCr_2S_4 the Mn ions are bivalent in a $3d^5$ and the Cr ions are trivalent in a $3d^3$ electronic configurations [6]. The effective magnetic moment per molecule $\mu_{\text{eff}} = 7.62 \mu_B/\text{f.u.}$ is close to the spin-only value $p_{\text{eff}} = 8.06 \mu_B/\text{f.u.}$ (see Table 1).

The second end member $\text{Ga}_{2/3}\text{Cr}_2\text{S}_4$ is an insulating ferromagnetic normal spinel with the Curie temperature $T_C = 56 \text{ K}$ and the Curie-Weiss temperature $\theta_{\text{CW}} = 40 \text{ K}$ [7]. For the same compound Nakatani [8] found $T_C = 30 \text{ K}$, $\theta_{\text{CW}} = 20 \text{ K}$ and $\mu_{\text{eff}} = 3.53 \mu_B/\text{Cr}$ for a sample with spinel structure and $T_N = 10 \text{ K}$, $\theta_{\text{CW}} = -95 \text{ K}$ and $\mu_{\text{eff}} = 3.60 \mu_B/\text{Cr}$ for a sample with vacancy-ordered spinel structure. Later, Belov *et al.* [9] found a spin-glass-like behaviour in $\text{Ga}_{2/3}\text{Cr}_2\text{S}_4$ with the freezing temperature $T_f = 4.5 \text{ K}$ and the Curie-Weiss temperature $\theta_{\text{CW}} = 10 \text{ K}$ and Koroleva *et al.* [10] the magnetic moment per molecule of $0.4 \mu_B$, obtained in a field of 50 kOe at 4.2 K . Some magnetic and electrical parameters of the border spinels MnCr_2S_4 and $\text{Ga}_{2/3}\text{Cr}_2\text{S}_4$ are collected in Table 1.

In the isostructural $\text{MnCr}_{2-2x}\text{In}_{2x}\text{S}_4$ spinel system the re-entrant spin-glass (RSG) and spin-glass-like (SG) behaviour of magnetization caused by the fluctuating competitive ferromagnetic (FM) and antiferromagnetic (AFM) interactions at low temperatures in the substitution range $0.0 \leq x \leq 0.5$ have been observed [11]. The most pronounced RSG behavior of magnetization was observed in the sample with the highest dilution in the substitution range. The spontaneous magnetic moment vanishes in the range $0.15 \leq x \leq 0.5$ and the susceptibility of the samples has a non-monotonic temperature dependence with

the maximum near $T_m = 16-17$ K that shifts slightly with the substitution [11]. The dc magnetization measurements using the high field extrapolation of the modified Arrot plots in $\text{MnCr}_{2-2x}\text{In}_{2x}\text{S}_4$ single crystals showed an influence of In for Cr substitution on the para- to ferromagnetic phase transition [12]. Also, the non-monotonic temperature dependences of Kouvel-Fisher effective susceptibility exponent for disordered samples and its increase with increasing In concentration were established [12].

The present contribution reports the magnetic properties for MnCr_2S_4 ferrimagnet doped with high content of the non-magnetic Ga ions (75 %) located in octahedral sites of the spinel structure. It was thus natural to expect the strong spin rearrangement of the magnetic moments and/or an appearance of a new magnetic transition.

2. Experimental

A powder sample of the $\text{Mn}[\text{Cr}_{0.5}\text{Ga}_{1.5}]\text{S}_4$ spinel was obtained with the aid of a ceramic method [13]. The X-ray diffraction made with the aid of the SIEMENS D5000 diffractometer revealed a single-phase material with the cubic spinel structure of the $(\text{A})[\text{B}_2]\text{X}_4$ type (where $A = \text{Mn}$, $B = \text{Cr}$ and $X = \text{S}$) and the space group $Fd3m$ (No. 227). The brackets $()$ and $[\]$ mean the tetra- and octahedral coordination, respectively. The lattice parameter $a = 1003.11(1)$ pm and the positional anion parameter $u = 0.383(2)$ were established. A Rietveld

refinement of the spinel structure showed an octahedral coordination [B] of the Ga ions.

Magnetization, dc and ac susceptibilities were measured using a step-magnetometer at 4.2 K and in applied external fields up to 150 kOe, a Faraday type Cahn RG automatic electrobalance in the temperature range 4.2-300 K and in applied external magnetic field $H_{dc} = 919$ Oe as well as a Lake Shore 7225 ac susceptometer/dc magnetometer in the temperature range 2-80 K and at different internal oscillating magnetic fields H_{ac} changing from 1 to 10 Oe with internal frequency $f = 120$ Hz, respectively. The magnetization was measured in applied external magnetic field $H_{dc} = 100$ Oe and 1 kOe and in two different cooling modes. In the zero-field-cooled (ZFC) mode the sample was first cooled down in the absence of an external magnetic field and then investigated while heating in a given magnetic field. The field-cooled (FC) mode usually followed ZFC run when the same magnetic field was set on at high temperatures and measurements were performed with decreasing temperature. For both modes, the cooling process always started from the paramagnetic state.

The dynamic ac magnetic susceptibilities were measured in the ZFC mode. Before the proper ac measurements a testing procedure was done. The in-phase χ' and out-of-phase χ'' components of the ac susceptibility were recorded simultaneously as a function of temperature and: 1) at different external static fields H_{dc} changing from 0 to 1 kOe, 2) at different internal oscillating field H_{ac} changing from 1 to 10 Oe with the internal frequency $f = 120$ Hz. A testing procedure described above and depicted in Fig. 1 showed that the increasing

external static field H_{dc} suppresses the fine magnetic structure while the increasing internal oscillating field H_{ac} reveals a spin dynamics in a system for $\chi'(T)$ and the energy losses for $\chi''(T)$. Therefore, the signals of the second (χ_2) and third (χ_3) harmonics associated with non-linear susceptibilities were detected as a function of temperature using an oscillating field of 5 Oe without applied static magnetic field. One can conclude that the fine magnetic structure is sensitive on a kind and strength of magnetic field.

3. Results and discussion

Figure 2 shows a temperature dependence of dc mass susceptibility, $\chi_\sigma(T)$, of the $\text{Mn}[\text{Cr}_{0.5}\text{Ga}_{1.5}]\text{S}_4$ spinel. The curve deviates downward from its linear part characteristic for ferrimagnetic coupling of magnetic moments. The negative value of the Curie-Weiss temperature $\theta_{CW} = -55$ K suggests a strong antiferromagnetic component. The effective magnetic moment $\mu_{\text{eff}} = 5.96 \mu_B/\text{f.u.}$ estimated from equation: $\mu_{\text{eff}} = 2.83\sqrt{MC_\sigma}$, where the molar mass $M = 313$ g and the Curie constant $C_\sigma = 1.417 \cdot 10^{-2} \text{ K}\cdot\text{cm}^3/\text{g}$ taken from experiment is quite close to the spin-only value ($p_{\text{eff}} = 6.52 \mu_B/\text{f.u.}$). This value was calculated from equation: $p_{\text{eff}} = \sqrt{p_{\text{Mn}}^2 + 0.5p_{\text{Cr}}^2}$, where $p = 2\sqrt{S(S+1)}$ for a Mn^{2+} ion ($S = 5/2$) with $3d^5$ and for a Cr^{3+} ion ($S = 3/2$) with $3d^3$ electronic configurations. It means that the orbital moment of both magnetic ions is totally quenched due to strong crystalline electric field interactions.

A shape of magnetic isotherms, $\sigma(H)$, made up to 60 kOe shows a lack of saturation and a nonlinear increase of magnetization with increasing magnetic field (Fig. 3), indicating that the spontaneous magnetization occurs in the Mn[Cr_{0.5}Ga_{1.5}]S₄ sample. It disappears going into the paramagnetic limit ($T_C = 48$ K). Similar behaviour of $\sigma(H)$ has been observed for polycrystalline MnCr_{2-2x}In_{2x}S₄ spinel with $x = 0.3$ [11]. An increase of magnetic field even up to 140 kOe at 4.2 K also did not give the saturation magnetization; however, appearing hysteresis loop accompanied with a clear step in magnetization may suggest a strong rearrangement spin process in a sample (Fig. 4).

Figure 5 presents a temperature dependence of magnetization, $\sigma(T)$, at $H_{dc} = 100$ Oe (lower picture) and 1 kOe (upper picture) measured in ZFC and FC cooling modes. There is no difference between ZFC and FC magnetizations at 1 kOe and the fine magnetic structure is not visible. Generally, a curve shape at 1 kOe suggests a ferrimagnetic ordering. Next, a curve, $\sigma(T)$, at $H_{dc} = 100$ Oe (lower picture) shows the multiple magnetic transitions with fine magnetic structure. At low temperatures we observe a collinear antiferromagnetic order of magnetic moments with a sharp peak at the Néel temperature $T_N = 3.9$ K. This ordering is ascribed to the A(Mn) sublattice because it correlates well with the temperature dependence of the local magnetic moments of the Mn ions obtained by neutron diffraction [14]. A minimum at $T_{YK} = 5$ K is attributed to a transition from a collinear Néel magnetic structure to a triangular Yafet-Kittel type [15]. Our value of the Yafet-Kittel temperature $T_{YK} = 5$ K for Mn[Cr_{0.5}Ga_{1.5}]S₄ spinel is close to 5.5 K for MnCr₂S₄ [15], pointing out the same mechanism connected

with a canting of the Mn moments (assigned to strong AFM A - A interactions) with respect to the Cr moments (assigned to FM via the sulfur ions superexchange B - S - B interactions [16]). A broad maximum at the freezing temperature $T_f = 7.9$ K and a large splitting between the ZFC and FC magnetizations with decreasing temperature are characteristic for a spin-glass-like behavior. Probably, in this case it is not a conventional spin glass with randomly oriented magnetic moments but it has a re-entrant character connected with a bond frustration of competing AFM and FM exchange interactions of nearly equal strength. No similar behaviour in the parent MnCr_2S_4 spinel was observed. So, a presence of the re-entrant spin-glass state in the $\text{Mn}[\text{Cr}_{0.5}\text{Ga}_{1.5}]\text{S}_4$ spinel maybe a consequence of dilution of magnetic $B(\text{Cr})$ sublattice by the non-magnetic Ga ions. The temperature dependence of magnetization, $\sigma(T)$, at $H_{\text{dc}} = 100$ Oe between the freezing temperature $T_f = 7.9$ K and the Curie temperature $T_C = 48$ K indicates rather a ferrimagnetic order, although at T_C the $B(\text{Cr})$ sublattice usually orders ferromagnetically in the case of sulfur spinels [16], whereas the $A(\text{Mn})$ sublattice still remains quasiparamagnetic, just as in MnCr_2S_4 [3].

The higher harmonics of ac susceptibility, resulting from hysteresis and nonlinearity of magnetization [17] are helpful for interpretation of spin arrangement in the spinel under study, too. The second $\chi_2(T)$ and third $\chi_3(T)$ harmonics presented in Figs. 6 and 7 have one feature in common: they vanish below T_f and above T_C in accordance with the simple molecular field theory [18]. We suggest the following scenario of magnetic ordering between T_f and T_C

in the $\text{Mn}[\text{Cr}_{0.5}\text{Ga}_{1.5}]\text{S}_4$ spinel. The in-phase second harmonic $\chi_2'(T)$ is negative and has a large minimum close to T_C indicating a parallel spin arrangement in this temperature region as dominant. The out-of-phase second harmonics $\chi_2''(T)$ is positive and has a high maximum also close to T_C relating to the energy dissipation (Fig. 6). The in-phase third harmonic $\chi_3'(T)$ has a positive large peak and a negative small one close to T_f indicating a ferrimagnetic spin arrangement in this temperature region as dominant. This state may result from the fact that the longitudinal component of the magnetic moment of the $A(\text{Mn})$ sublattice is ordered antiparallel to the moment of the $B(\text{Cr})$ sublattice (ordered parallel), whereas the transverse component of the moment of the $A(\text{Mn})$ sublattice is fully disordered [3]. The second negative small cusp observed for $\chi_3'(T)$ confirms a parallel spin arrangement at T_C . Similarly, the out-of-phase third harmonics $\chi_3''(T)$ is positive and has a high maximum close to T_f relating to the energy lost (Fig. 7). In general, the out-of-phase harmonics are usually associated with the energy losses, connected for example with the magnetic-domain-wall motion or with rotation of magnetization within domains [19].

Figure 8 presents a dependence of ac mass susceptibility vs. magnetic field, $\chi_{ac}(H)$, up to 60 kOe for $\text{Mn}[\text{Cr}_{0.5}\text{Ga}_{1.5}]\text{S}_4$ spinel. No critical fields characteristic, *e.g.* for a metamagnetic transition and/or a transition from a conical spin arrangement into ferromagnetic phase [20] were observed. Only a small hysteresis loop at 4.3 K is visible in the insets of Figs. 3 and 8 confirming a strong rearrangement spin process in a sample. A lack of critical fields means

that the sample under study is far from saturation magnetization. It concerns also the $A(\text{Mn})$ and $B(\text{Cr})$ magnetic sublattices treated alone. This suggestion is in agreement with the observed saturation magnetization of $6 \mu_{\text{B}}$ found in high magnetic fields of 300 kOe for MnCr_2S_4 [21], where the magnetic moment of the Mn ions becomes perpendicular to the Cr moment. It takes effect that the longitudinal component of the moment of the Mn ions becomes zero and only the Cr moment remains visible in the saturation state [3].

4. Conclusions

In conclusion, we have discovered the spin-glass-like behaviour and complex magnetic order in $\text{Mn}[\text{Cr}_{0.5}\text{Ga}_{1.5}]\text{S}_4$ spinel by observation of magnetization at low magnetic fields up to 100 Oe in ZFC and FC cooling regime. A dilution of magnetic sublattice $B(\text{Cr})$ by nonmagnetic Ga-ions ensures a bond frustration resulting from a competition between AFM and FM exchange interactions of nearly equal strength. The dc magnetic field of 1 kOe destroys the fine magnetic structure with multiple magnetic transitions conserving the ferrimagnetic ordering of magnetic moments.

Acknowledgements

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Table 1

Magnetic and electrical parameters of the border compounds MnCr_2S_4 and $\text{Ga}_{2/3}\text{Cr}_2\text{S}_4$. T_{YK} , T_{N} , T_{f} , θ_{CW} and T_{C} are the Yafet-Kittel, Néel, freezing, Curie-Weiss and Curie temperatures, respectively, C_{m} is the molar Curie constant, μ_{eff} is the effective magnetic moment per molecule, p_{eff} is the theoretical effective magnetic moment per molecule, ρ is the electrical resistivity at room temperature, E_{A} is the activation energy at $T > 260$ K and S is thermopower at 313 K.

Spinel	T_{YK} (K)	θ_{CW} (K)	T_{C} (K)	C_{m} (Kcm^3/mol)	μ_{eff} ($\mu_{\text{B}}/\text{f.u.}$)	p_{eff} ($\mu_{\text{B}}/\text{f.u.}$)	ρ (Ωcm)	E_{A} (eV)	S ($\mu\text{V}/\text{K}$)	Refs
MnCr_2S_4	5.5	10	66	7.25	7.62	8.06	$4 \cdot 10^9$	0.3	-143	[1-4]
$\text{Ga}_{2/3}\text{Cr}_2\text{S}_4$	-	40	56	-	-	-	$4 \cdot 10^{12}$	-	-	[7,10]
	-	20	30	3.11	4.99	5.48	-	-	-	[8] ¹⁾
	$T_{\text{N}}=10$	-95	-	3.23	5.09	5.48	-	-	-	[8] ²⁾
	$T_{\text{f}}=4.5$	10	-	-	-	-	-	-	-	[9]

¹⁾Spinel structure. ²⁾Vacancy-ordered spinel structure.

Figure captions

Fig. 1. Zero field ac susceptibilities $\chi'(T)$ and $\chi''(T)$ recorded simultaneously as a function of temperature T with internal frequency $f = 120$ Hz at oscillating magnetic fields of 1 and 10 Oe where different external static fields H_{dc} changing from 0 to 1 kOe. The arrows indicate an increase of (ac) and (dc) magnetic fields.

Fig. 2. Mass susceptibility χ_σ and $1/\chi_\sigma$ vs. temperature T recorded at $H_{dc} = 919$ Oe. The dashed line indicates a Curie-Weiss behavior.

Fig. 3. Zero field cooled dc mass magnetization M vs. magnetic field H up to 60 kOe at 4.3, 7.2, 9.9, 14.9 and 39.4 K. Inset: the onset of the $\sigma(H)$ dependence up to 1.0 kOe showing a hysteresis loop for $T = 4.3$ K.

Fig. 4. Zero field cooled dc mass magnetization M vs. magnetic field H up to 140 kOe at 4.2 K. A run of magnetic field is indicated by arrows.

Fig. 5. Temperature dependence of zero field cooled (ZFC) and field cooled (FC) dc mass susceptibility χ_σ recorded at $H_{dc} = 100$ Oe and 1 kOe. The temperatures of T_N , T_{YK} , T_f and T_C are indicated by arrows.

Fig. 6. Temperature dependence of the real χ_2' and imaginary χ_2'' components of zero field second harmonic of ac susceptibility with internal frequency $f = 120$ Hz at oscillating field $H_{ac} = 5$ Oe. The temperatures of T_f and T_C are indicated by arrows.

Fig. 7. Temperature dependence of the real χ_3' and imaginary χ_3'' components of zero field third harmonic of ac susceptibility with internal frequency $f = 120$ Hz at oscillating field $H_{ac} = 5$ Oe. The temperatures of T_f and T_C are indicated by arrows.

Fig. 8. Zero field cooled ac mass susceptibility χ_{ac} vs. magnetic field H at 4.3, 7.9, 12.7, 17.4 and 36.7 K. Inset: the onset of the $\chi_{ac}(H)$ dependence up to 1.0 kOe showing a hysteresis loop for $T = 4.3$ K.

Fig 1

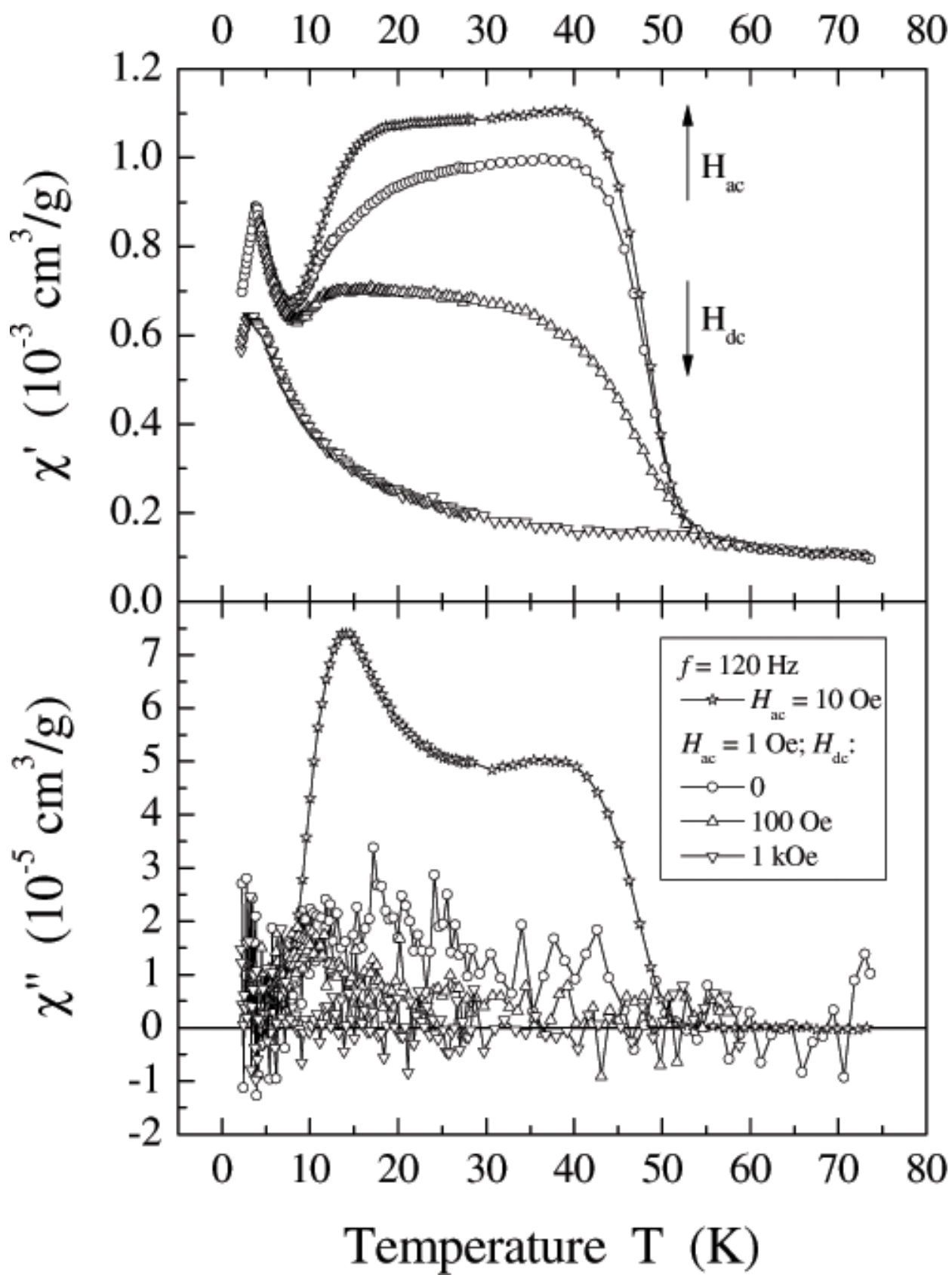


Fig 2

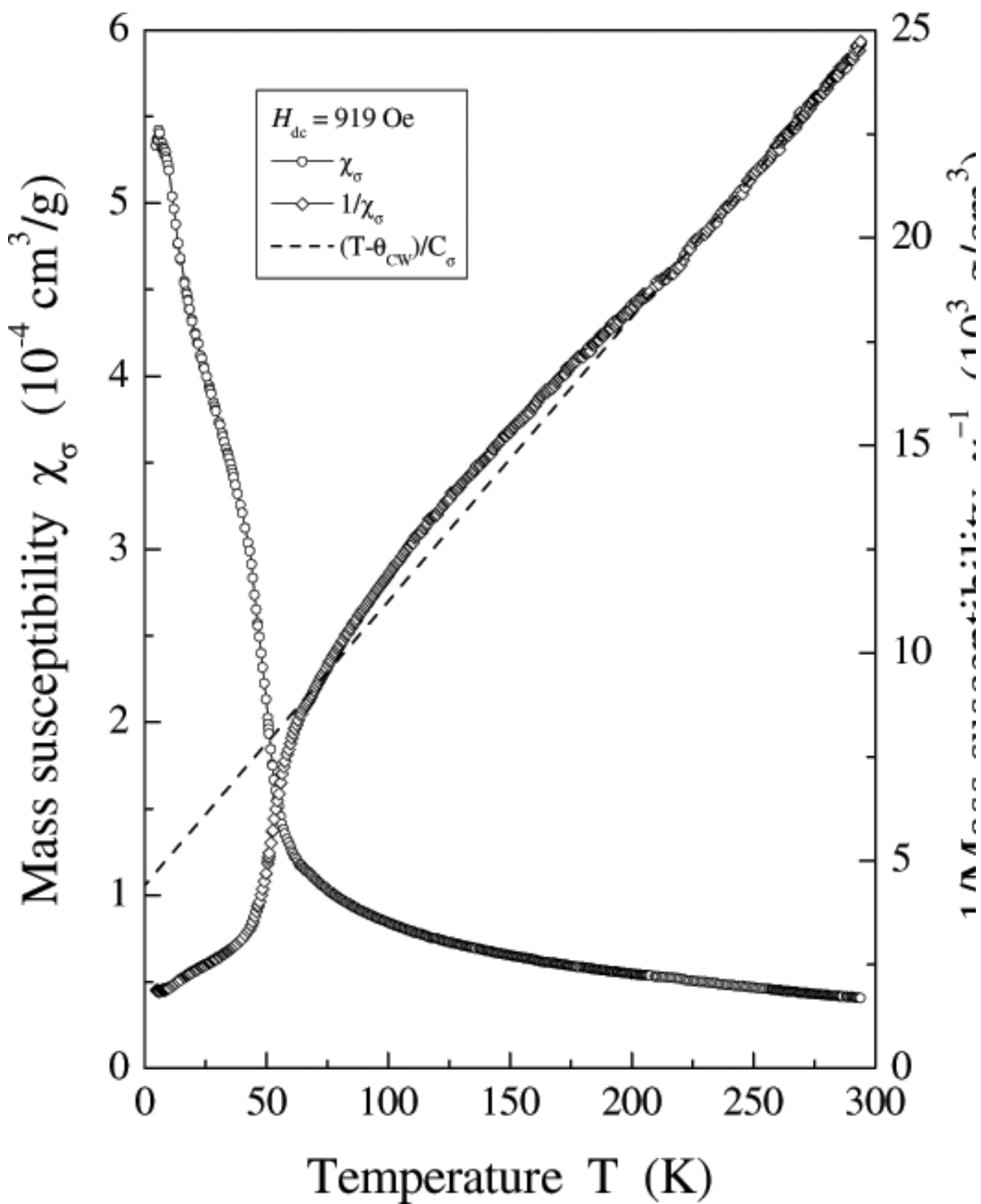


Fig 3

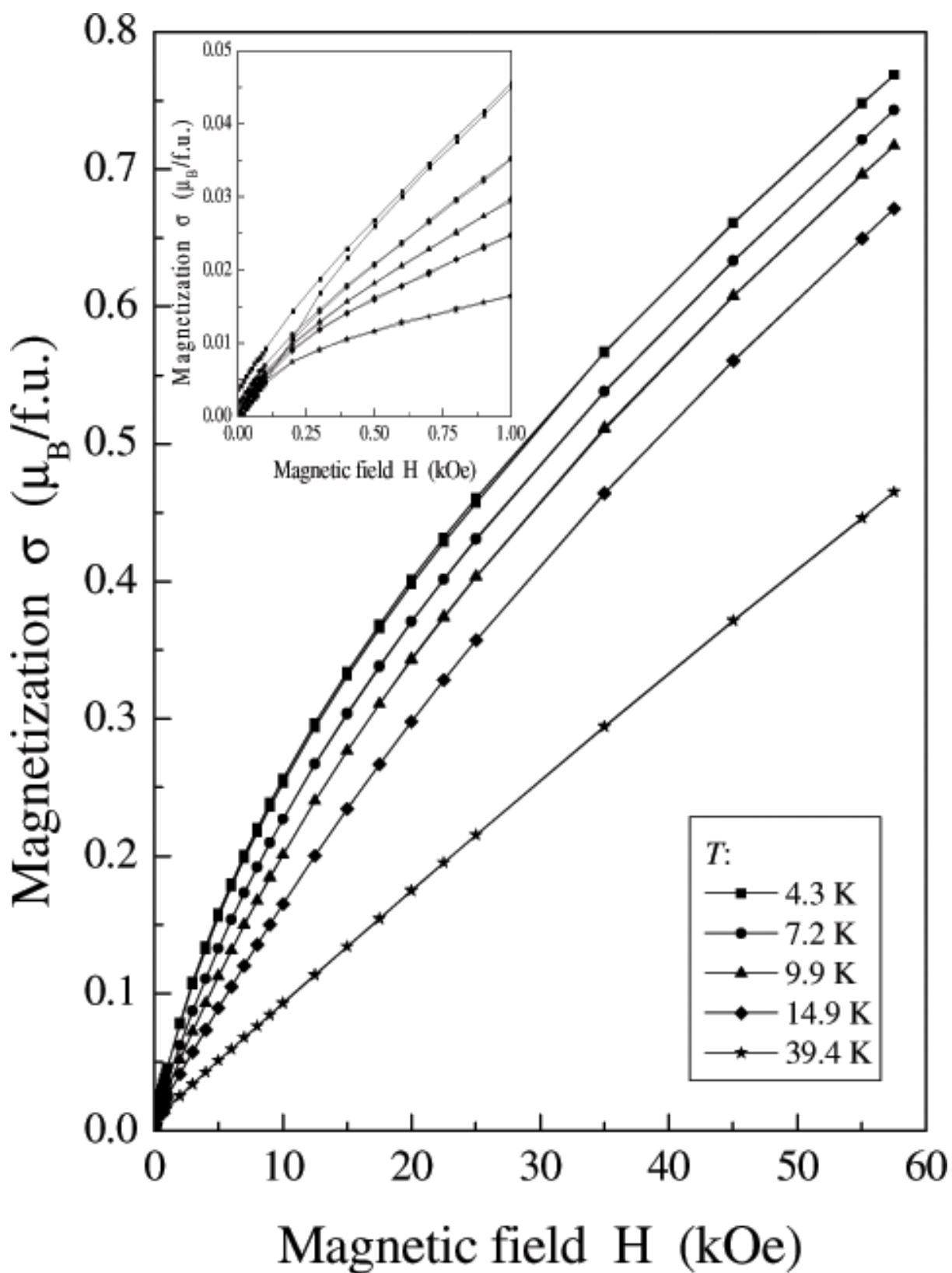


Fig 4

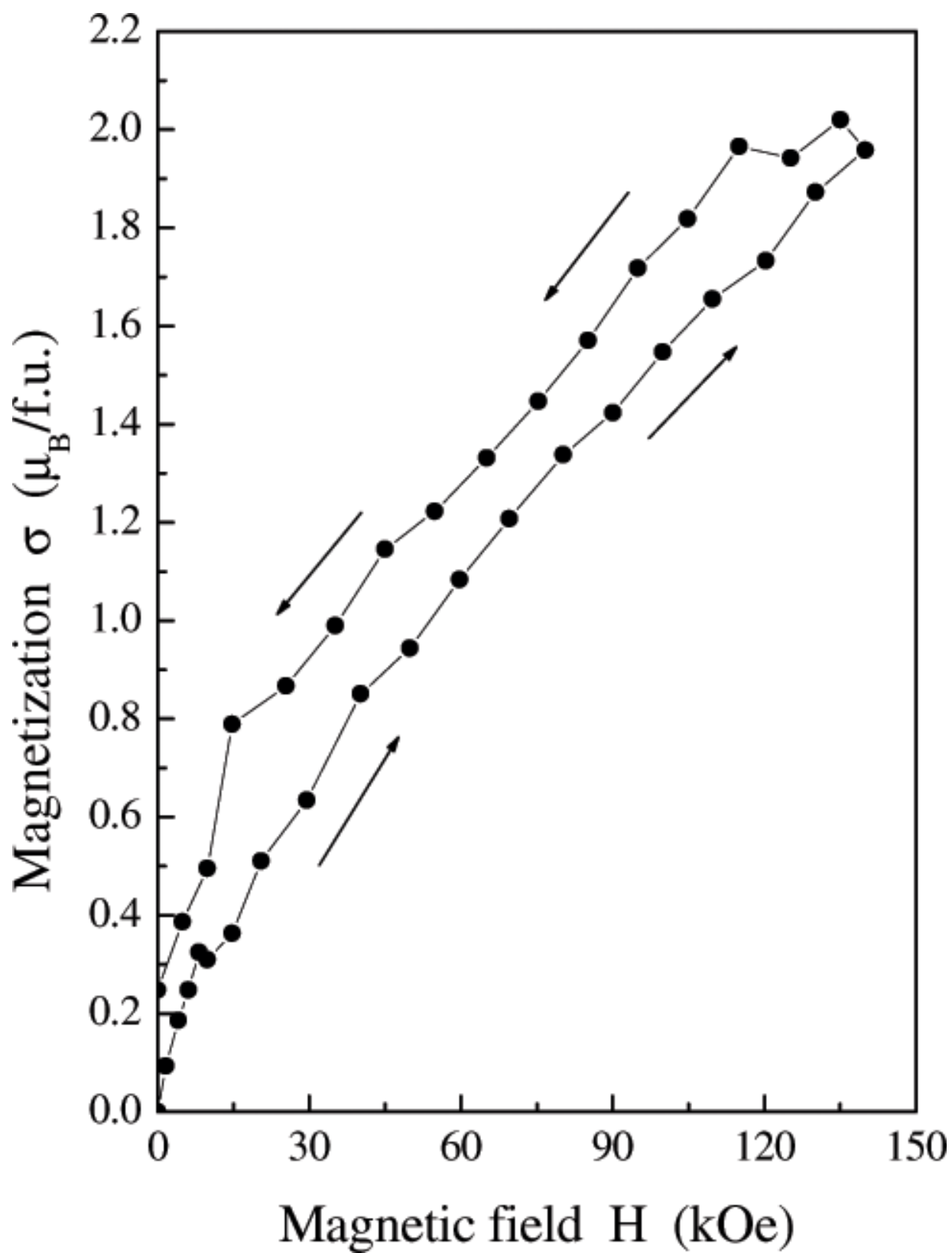


Fig 5

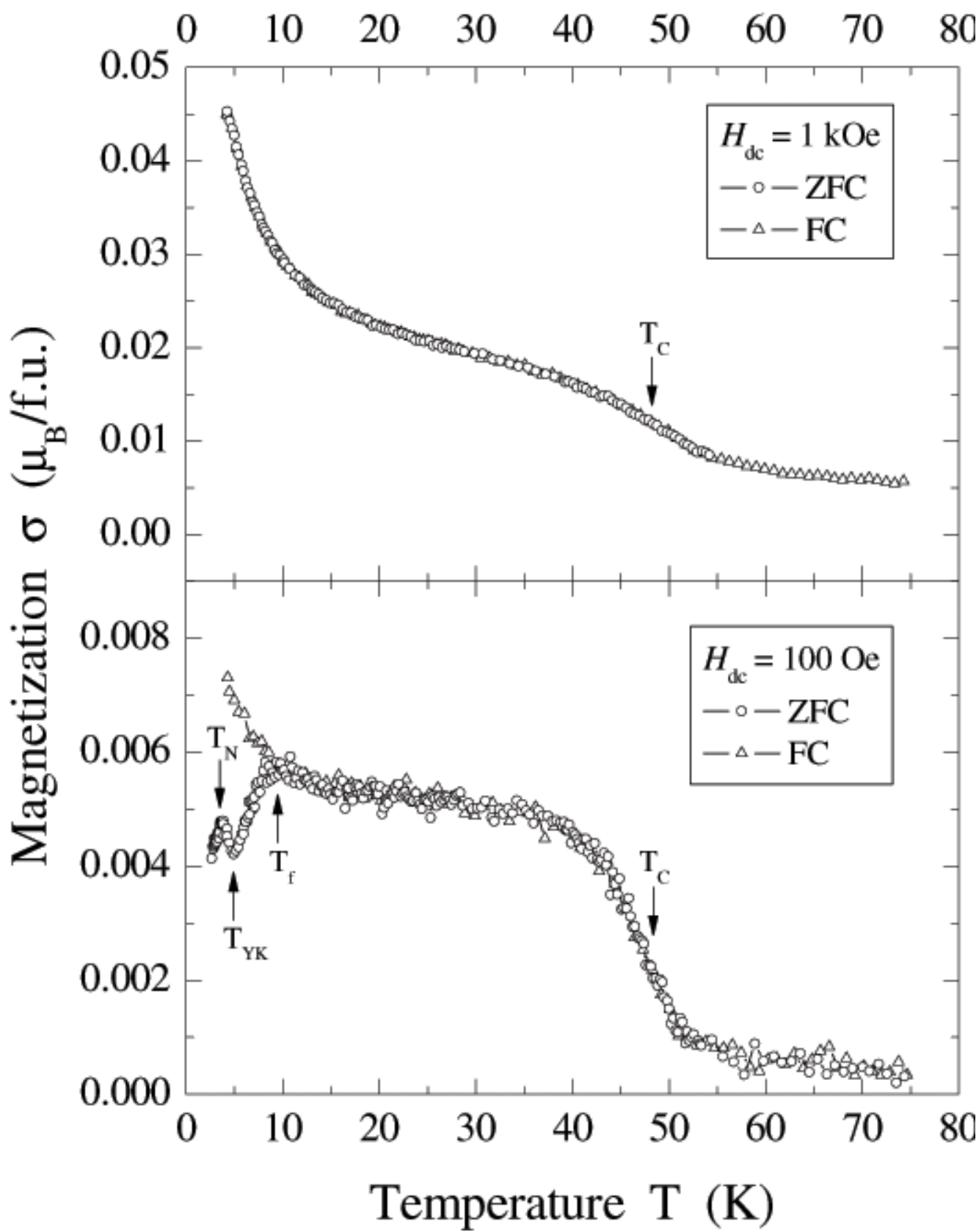


Fig 6

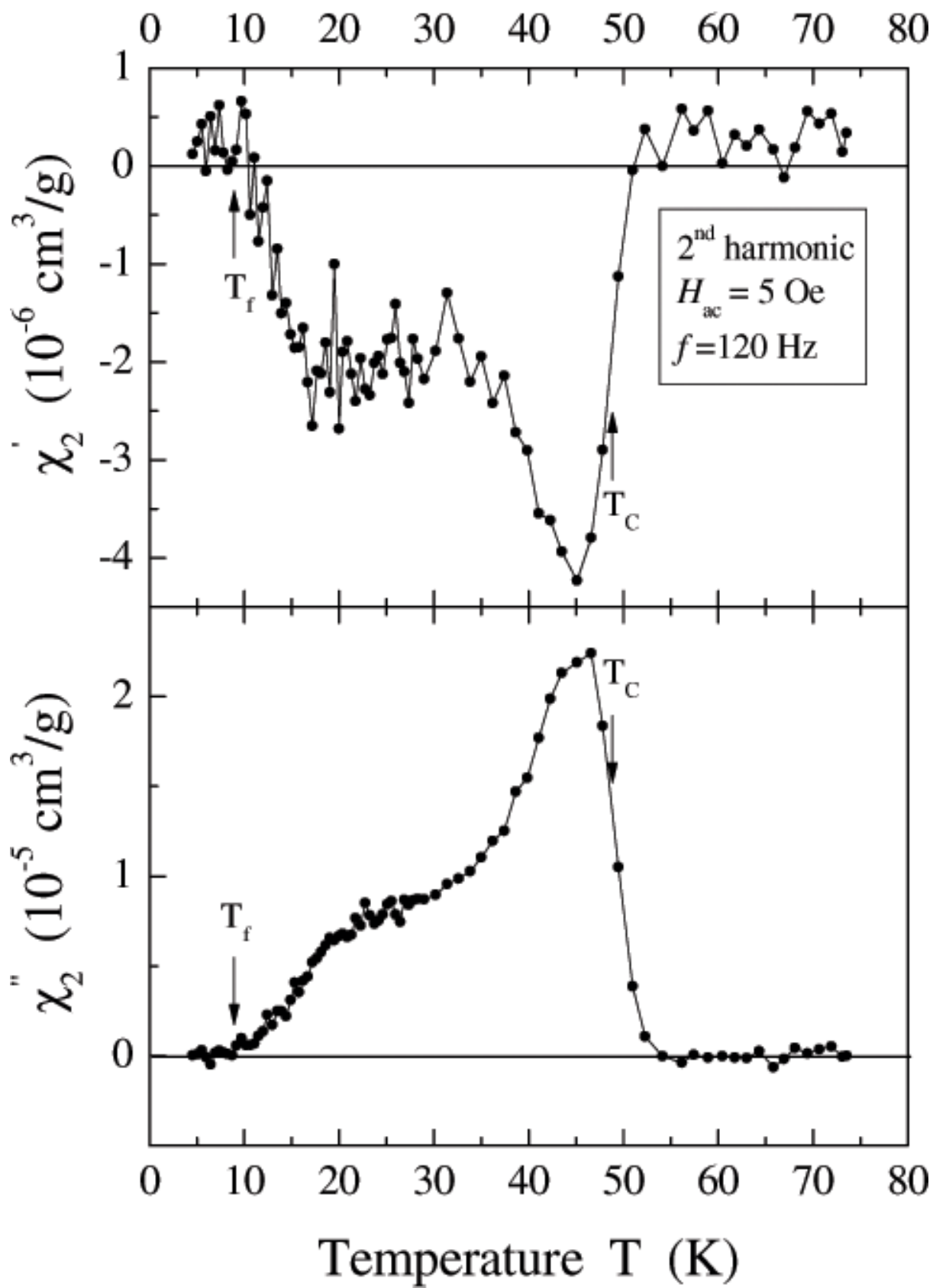


Fig 7

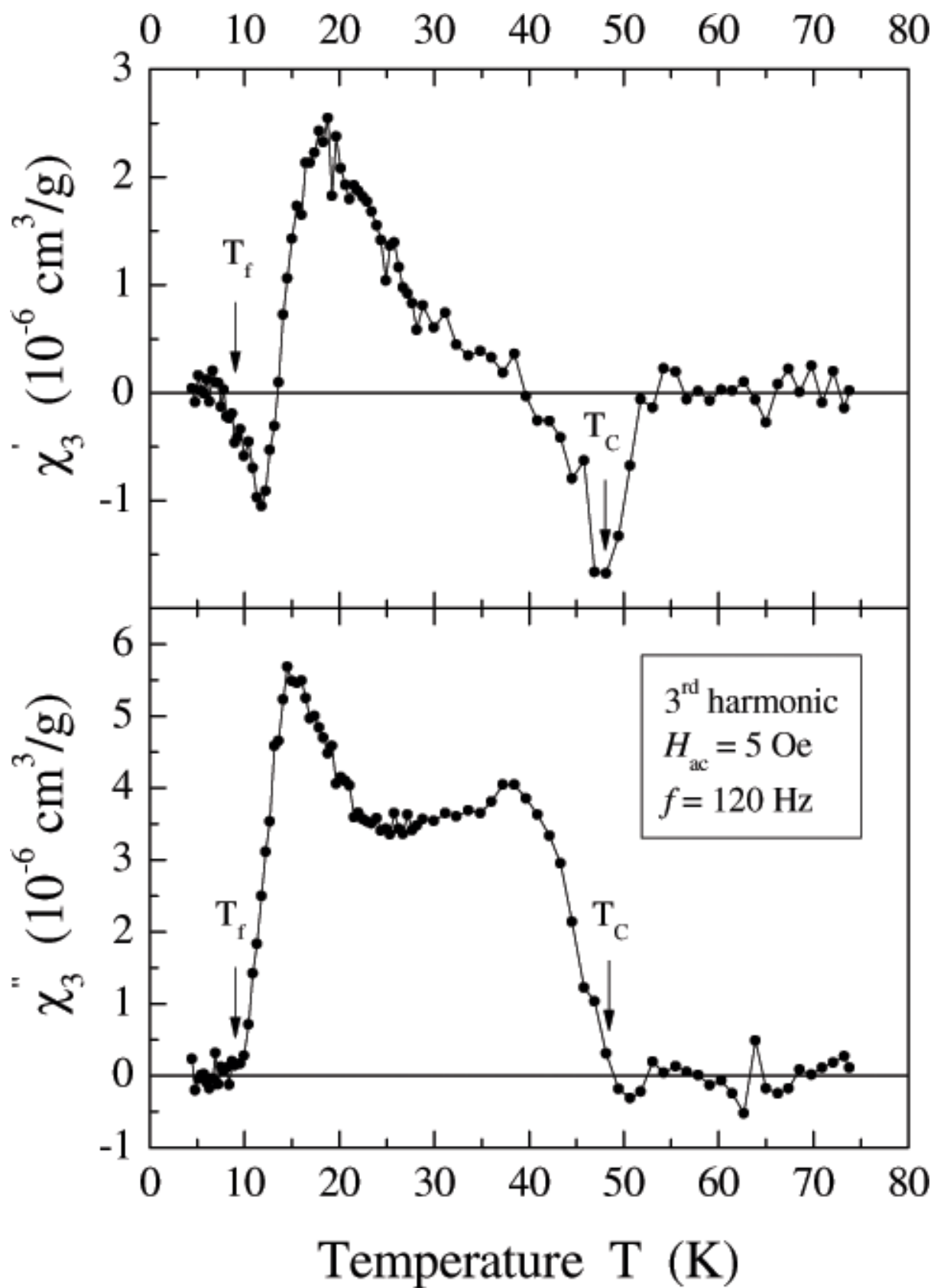


Fig 8

