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Details of Magnetic Properties in Pb(Fe$_{1/2}$Nb$_{1/2}$)O$_3$

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For Pb(Fe$_{0.5}$Nb$_{0.5}$)O$_3$ the $\chi(T)$ and $m(H)$ measurements up to 70 kOe were performed with the aim to estimate the antiferromagnetic (AFM) and superantiferromagnetic (SAF) contribution to $\chi(T)$. Below T_N the increase of $\chi(T)$ is attributed to a small part of Fe^{3+} ions in the SAF clusters. For high fields the suppression of the AFM and enhancement of the SAF susceptibility was observed. An inflection point of the $m(H)$ curves below the Néel point suggests the presence of a spin reorientation process connected with the AFM phase.

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1. Introduction

Lead iron niobate Pb(Fe$_{0.5}$Nb$_{0.5}$)O$_3$ (PFN) and the PFN based solid solutions have been studied in recent years with regard to their extreme multiferroic properties [1–7]. The basic magnetic properties of PFN were reported many years ago [8] but because of their complexity they were not up to now consistently interpreted. Above the Néel temperature $T_\mathrm{N} \approx 145$ K PFN is a paramagnet (PM) and below T_N this compound seems to represent a mixture of the long-range antiferromagnetic (AFM) and superantiferromagnetic (SAF) Fe^{3+} clusters [4]. At low temperatures a cluster glass (CG) state is observed with a freezing temperature near 10 K [1, 2, 4, 7]. In the present contribution, following the work on Ba and Ti doped PFN [7], we are concerned with the more detailed magnetometric study of PFN with the aim to clarify the increase of the susceptibility below T_N when going towards lower temperatures.

2. Experimental

The experiments were made on non-oriented single crystals of Pb (Fe$_{0.5}$Nb$_{0.5}$)O$_3$ grown by the spontaneous crystallization procedure from the PbO-B$_2$O$_3$ flux (for details we refer to the work [7]). The magnetic measurements were carried out using the SQUID magnetometers MPMS-5S, MPMS-XL (Quantum Design). The zero-field (ZFC) and field-cooled (FC) susceptibilities χ_ZFC, χ_FC were measured under different magnetic fields up to 70 kOe. The magnetization curves $m(H)$ with increasing field (virgin curves) and decreasing field were recorded in the wide range of temperatures (m is expressed in Bohr magnetons p.f.u.). The irreversibility has been found only below 10 K, which is in accord with the temperature dependence of the remanence [7].

Letting aside χ_ZFC and problems of the CG state we focus our attention on the χ_FC measured under different applied fields. The transition from a PM to long-range AFM state at the Néel temperature T_N can be well seen on the derivative of the $d\chi_\mathrm{FC}/dT$. This dependence for $H = 500$ Oe is shown in Fig. 1a. The amplitude A of $\mathrm{d}\chi_\mathrm{FC}/\mathrm{d}T$ depends on H (inset of Fig. 1a). For $H = 70$ kOe the transition is still visible but the amplitude A decreases to about 0.01 of the value at the low fields. In Fig. 1b we show $\mathrm{d}(1/\chi_\mathrm{FC})/\mathrm{d}T$ for $H = 500$ Oe and $H = 70$ kOe. The results of the study of the $m(H)$ curves are summarized in Fig. 2, in the form of the derivatives $\mathrm{d}m/\mathrm{d}H$. At $T = 2$ K and between 60 and 135 K we find a maximum of $\mathrm{d}m/\mathrm{d}H$ (inflection point of $m(H)$) at a critical field H_inf depending on temperature (inset of Fig. 2b).

3. Discussion

In the low temperature region (2–40 K) (Fig. 2b) we find an inflection point of the $m(H)$ at $T = 2$ K, which is a typical feature of the CG state (an S type of the $m(H)$). Near the freezing temperature, at $T = 10$ K this behaviour is absent.

Below T_N and reliably outside the region of the cluster growth ($T < T_{gs} \approx 50$ K [7]), between 60 and 120 K the $\chi_\mathrm{FC}(T)$ should contain both AFM and SAF contributions. The first χ_AFM has its origin in the long-range AFM (proved by the neutron diffraction in the work [3]) and the second SAF contribution $\chi_\mathrm{SAF}(T)$ corresponds to uncompensated spins. In our case, we may assume that $\chi_\mathrm{SAF}(T)$ follows the Curie-Weiss (CW) law (generally is given by the Brillouin function). An attempt to crudely estimate the value of these two contributions for $H = 500$ Oe will be made using a simplified assumption that between 60 and 120 K χ_AFM does not depend on temperature. Starting then from the relation $\chi_\mathrm{FC} = \chi_\mathrm{AFM} + C/(T+\theta)$ we get $C = 0.328$ emu K/(mol Oe), $\theta = 14.8$ K and $\chi_\mathrm{AFM} = 0.0019$ emu/(mol Oe). This AFM susceptibility representing about one half of the expected χ_AFM at the Néel point seems to be reasonable. (Taking $\theta = 520$ K [8] we obtain χ_AFM (145 K)= 0.0032 emu/(mol Oe)). The values of $d(1/\chi)/dT$ corresponding to this approximation are denoted in Fig. 1b by open circles. The interesting fact concerns the influence of the magnetic field. We see that for $H = 70$ kOe, between 100 and 140 K the value $\mathrm{d}(1/\chi_\mathrm{FC})/\mathrm{d}T$ remains approximately constant (Fig. 1b), which corresponds to the situation that $\chi_\mathrm{FC}(T)$ is given only by a CW term. In this case we obtain $C = 0.9$ emu K/(mol Oe) and $\theta = 90.4$ K.

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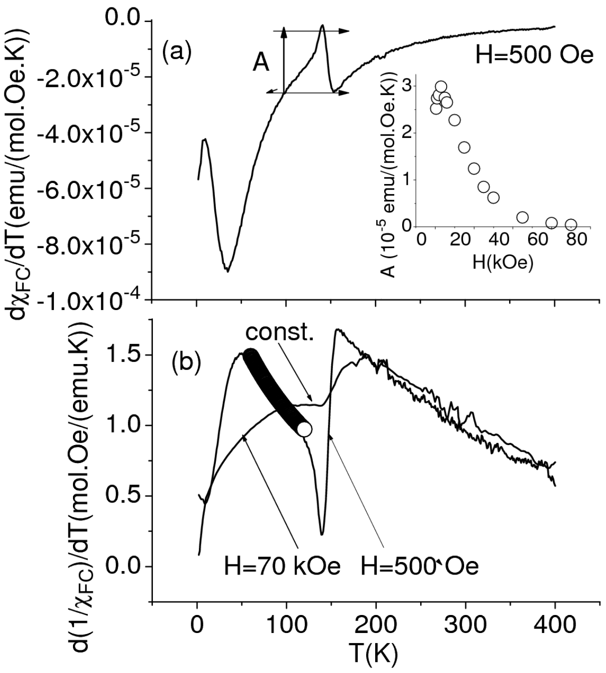

Fig. 1. (a) Evaluated values of $d\chi_{FC}/dT$ for $H = 500$ Oe, in the inset: amplitude A of $d\chi_{FC}/dT$ at $T = T_N$ as a function of the applied field, (b) evaluated values of $d(1/\chi_{FC})/dT$ for the applied fields 500 Oe and 70 kOe; open circles denote the approximation with χ_{AFM} independent on temperature.

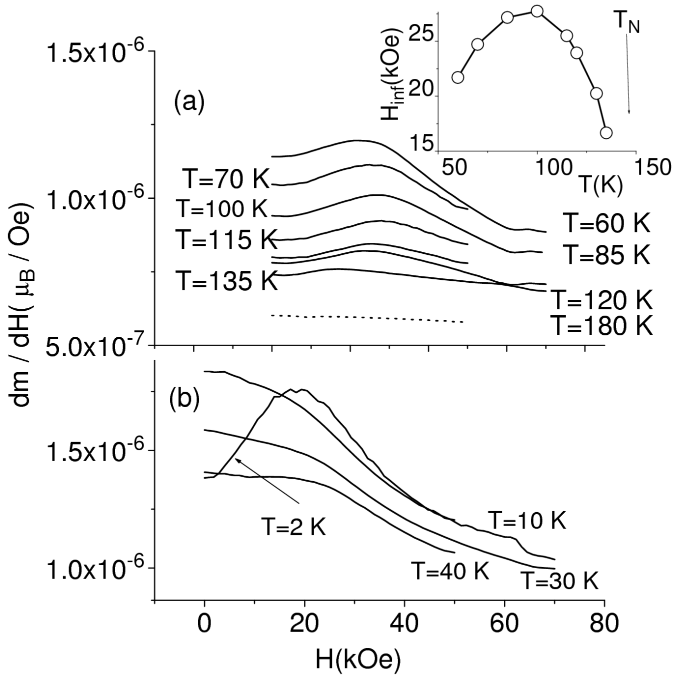

Fig. 2. Evaluated values of dm/dH, (a) the temperature region 60–180 K, in the inset: the field H_{inf} corresponding to the inflection point of $m(H)$ as a function of temperature, (b) temperature region 2–40 K.

This suggests that the magnetic field suppresses the AFM and enhances the SAF contribution. The same conclusion follows from the observed essential decrease of the amplitude A with increasing field. The reason for this behaviour remains unclear.

The portion p of the Fe^{3+} ions contributing to χ_{SAF} can be determined as $8.75C/(2.1875S_1(S_1+1))$ with S_1 denoting a spin of the uncompensated magnetic moment. (For 0.5Fe^{3+} the Curie constant $C = 2.1875$ emu K/(mol Oe)).

The behaviour of dm/dH (Fig. 2) suggests that χ_{SAF} is caused by small clusters with S_1 of the order of tens. In this case the increase of the χ_{FC} below T_N can be ascribed to only a small portion (several percents) of the Fe^{3+} ions. The field dependence of dm/dH has its origin in two contributions. The major contribution is due to the Brillouin behaviour of SAF clusters and second, with a maximum in dm/dH, can be ascribed to a spin reorientation in the AFM phase. With regard to the high value of the molecular field corresponding to T_N (10^6 Oe), this process seems not to be a metamagnetic transition in the long-range AFM phase, but rather is connected with a spin-flop transition.

4. Conclusions

At low fields ($H = 500$ Oe) the susceptibility below T_N (60–120 K), increasing with decreasing temperature, could be attributed to several percents of Fe^{3+} ions in the SAF clusters. For $H = 70$ kOe, below T_N the susceptibility seems to be formed only by a SAF term the AFM contribution being suppressed. In the temperature region 60–135 K the occurrence of an inflection point on the $m(H)$ curve indicates the presence of a spin-reorientation process connected with the AFM phase.

Acknowledgments

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