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Reference
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CHARACTERISATION OF SOME MAGNETIC AND MAGNETOELECTRIC PROPERTIES OF FERROELECTRIC Pb(Fe$_{1/2}$Nb$_{1/2}$)$_3$

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Abstract The magnetic properties of Pb(Fe$_{1/2}$Nb$_{1/2}$)$_3$ have been studied by neutron diffraction on a polydomain single crystal and susceptibility measurements on a single ferroelectric domain. A Néel temperature of ~160K was found but no further anomalies were indicated by the susceptibility data. No evidence of a spontaneous magnetic moment. Preliminary magnetoelectric measurements on single ferroelectric domains are presented.

INTRODUCTION

Recent birefringence and precision X-ray measurements on single crystals of the perovskite PbFe$_{1/2}$Nb$_{1/2}$O$_3$ - one of the first ferroelectrics with simultaneous antiferromagnetic ordering reported$^{[1]}$ - have shown$^{[2,3]}$ that the known transition at 383K is of second order and from cubic to tetragonal, whereas a hitherto unknown first order transition occurs at 354K, below which the crystal stays rhombohedral down to at least 4K.$^{[2]}$ On powder samples antiferromagnetic G-type spin ordering was found below 143-158K$^{[4,5]}$. However, magnetic susceptibility data of polydomain single crystals showed further anomalies around 125K and 63K and an antiferromagnetic transition at 9K. In addition, this transition was accompanied by the appearance of a spontaneous magnetic moment after cooling in simultaneous electric and magnetic fields, although the electric field did not appear to be essential$^{[6]}$. Using neutron diffraction on a polydomain single crystal and susceptibility and magnetic field induced magnetoelectric measurements, (ME)$_H$, on visually controlled poled ferroelectric single domains, the present work aimed at clarifying the previously reported susceptibility data$^{[6]}$ and the magnetic point group below $T_N$.  

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NEUTRON DIFFRACTION

A measured set of magnetic reflections \((h,k,1/2)\) confirms the \(G\) type ordering. The temperature dependance of the integrated intensity of the antiferromagnetic reflection \((1/2,1/2,1/2)\), indexed in terms of the chemical unit cell, is illustrated in Fig. 1. These measurement performed on the double axis spectrometer of the Saphir reactor, Würenlingen using neutrons of wavelength 2.338 Å (pyrolytic graphite monochromator), indicated \(T_N \sim 160K\) but considerable critical magnetic scattering is apparent above this temperature. Such scattering could result from the presence of small regions in which the Fe to Nb ion ratio is greater than average. These regions of more complete antiferromagnetic ordering would be expected to have a higher than average transition temperature.

MAGNETIC SUSCEPTIBILITY

The susceptibility measured in a field of \(1kG\), facilitated by a SQUID magnetometer, was determined as a function of temperature (Fig. 2) after first cooling the crystal in a magnetic field. The data shows only one anomaly at \(\sim 170K\) which is associated with the antiferromagnetic ordering observed by neutron diffraction. The absence of any indication of other anomalies and no evidence of a spontaneous magnetic moment below \(9K\) can only be understood in terms of differences of crystal homogeneity and stoichiometry. Optical studies\(^2\) and chemical analysis revealed that our crystals were of good homogeneity and high stoichiometry\(^7\). However, the anomaly at \(\sim 170K\) is not characteristic of an antiferromagnetic transition but rather has the form of a small antiferromagnetic effect superimposed on a paramagnetic background. One can thus suggest that not all the magnetic moments participate in the ordering. Such behaviour was also concluded by Bokov\(^4\).

MAGNETOELECTRIC EFFECT

The magnetic field induced magnetoelectric measurements were made on \((110)_c\) and \((111)_c\) platelets polished to \(\sim 60\mu\) such that optical control of the domain state etc. could also be carried out. To ensure the formation of a single magnetoelectric domain the crystal was cooled in an electric and magnetic field (\(\vec{E} \perp\) crystal plane, \(\vec{H} // <110>_c\)). The behaviour or the magnetoelectric signal\(^*\), \((\text{ME})_H\), as a function of rotation of the magnetic field about given crystal axes at \(15K\) (Fig. 3) is qualitatively consistent with the quadratic magnetoelectric effect expected for the trigonal magnetic point groups 3m or 3m\(^1\) - the only possibilities allowed by the presently available data on PFN. Possible origins of the quantitative discrepancies apparent from Fig. 3 are slight sample misorientation, inaccuracies due to temperature instability, the influence of other phenomena (e.g. magnetoelastic

\(^*\)The induced charge was recorded by a Keithley 642 electrometer
FIGURE 1 Temperature dependence of the integrated neutron intensity of the magnetic reflection (1/2, 1/2, 1/2).

FIGURE 2 Gram magnetic susceptibility as a function of temperature, measured in a field of 1 kG.

FIGURE 3 Induced polarisation as a function of magnetic field orientation for rotation about the axes indicated on the crystal sections shown at 15K. Signal measured by a field change of 5 kG (6 + 1 kG). At 0° H// <110>.

FIGURE 4 The axis convention employed. Orientation of axes within the pseudo-cube (a) and the (111) platelet (b).
effects) or the magnetic symmetry could be lower than trigonal. Tentative estimates of the non-zero coefficients (in SI units - sec/amp) of the quadratic magnetoelectric tensor at 15K (Fig. 4 shows the axis convention employed) are

\[ \beta_{111} = -\beta_{122} = -0.5 \beta_{212} \sim 8.7 \times 10^{-18}, \]
\[ \beta_{223} = \beta_{113} \sim 0.8 \times 10^{-18}, \]
\[ \beta_{322} = \beta_{311} \sim 7.4 \times 10^{-18}, \]
\[ \beta_{333} \sim 13.5 \times 10^{-18}. \]

To clarify all the possibilities raised by these preliminary results a detailed analysis of the (ME)_H effect in PFN, together with a full account of the derivation of the quadratic magnetoelectric tensor for 3m or 3m' symmetries, will be published later.

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