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BRIXEL, Wolf Dietrich, et al.

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MAGNETIC FIELD INDUCED MAGNETOELECTRIC EFFECTS, \((\text{ME})_H\), IN THE
PEROVSKITES \(\text{Pb}_2\text{CoWO}_6\) and \(\text{Pb}_2\text{FeTaO}_6\)

WOLF BRIXEL, JEAN-PIERRE RIVERA, ANTON STEINER AND HANS SCHMID
Department of Mineral, Analytical and Applied Chemistry,
University of Geneva, CH-1211 Geneva 4, Switzerland

Abstract On \(\text{Pb}_2\text{CoWO}_6\) the linear \((\text{ME})_H\) effect has been
measured in the ferromagnetic/ferroelectric phase \((T < 7.6K)\)
and the quadratic one in its paramagnetic/ferroelectric phase
\((T > 7.6K)\). On \(\text{Pb}_2\text{FeTaO}_6\) the quadratic \((\text{ME})_H\)-effect has been
measured \((4-17K)\), a new magnetic transition evidenced at \(\approx 9K\)
and the magnetoelectric coefficients \(\beta_{ijk}\) evaluated at 4.4 K

INTRODUCTION

Linear and quadratic \((\text{ME})_H\)-effects \(^{1,2}\) are based upon the following
two terms of the density of stored free enthalpy \(g\):

\[-g = \alpha_{ij}E_iH_j + (1/2)\beta_{ijk}E_iH_jH_k + \ldots\]

where \(\alpha_{ij}\), \(\beta_{ijk}\) are the linear and non-linear ME-susceptibilities,
\(E_i, H\) the electric and magnetic fields, respectively. The magnetic
field induced polarisation, \(P_i\), is defined by

\[-\partial g/\partial E_i = P_i = \alpha_{ij}H_j + (1/2)\beta_{ijk}H_jH_k + \ldots\]

This study reports measurements of \((\text{ME})_H\)-effects in the
orthorhombic phases III (ferroelectric/paramagnetic) and IV
(ferroelectric/ferromagnetic) of B-site ordered \(\text{Pb}_2\text{CoWO}_6\) (PCW)\(^{3,4}\)
and in the trigonal ferroelectric/antiferromagnetic phase of B-site
disordered \(\text{Pb}_2\text{FeTaO}_6\) (PFT)\(^{5,6}\). The magnetic field-induced charges
were measured by charge integration using a Keithley-642 electrometer\((10^{-10}-10^{-16}C)\). An electromagnet, equipped with a
time-linear regulation provided a field between 0 and 1 T. The
sample platelets were cut from flux grown single crystals\(^7\),
polished and gold-electroded.
THE FERROMAGNETIC TRANSITION OF Pb$_2$CoWO$_6$ AT 7.6K

The onset of ferromagnetism in the ferroelectric phase of PCW was found by means of magnetization measurements on ceramics ($T_c = 9$K)$^8$, corroborated by neutron powder diffraction$^3$ and in the present study by an anomaly of the (ME)$_H$-effect (Fig. 1). The effect was measured on a ferroelastic polydomain platelet previously cooled in a field of 50 kVcm$^{-1}$. Below $T_c$ the effect is linear in agreement with ferromagnetism and it becomes quadratic above. This seems the first time that a linear (ME)-effect has been measured in a ferroelectric perovskite, but the intricate domain pattern hindered the deduction of the ferromagnetic point group. The quadratic effect above 7.6K proves a ferroelectrically poled state (space group Amm21', predicted on semi-theoretical grounds$^4$) because an equi-weight distribution of polar domains would cancel the effect$^9$.

A NEW MAGNETIC TRANSITION OF Pb$_2$FeTaO$_6$ AT $\approx 9$K

In the trigonal ferroelectric phase of PFT$^5$ a quadratic (ME)$_H$-effect was measured between 4 and 17 K (Fig. 2). The anomaly at 9K is probably caused by a magnetic transition analogous to that of Pb$_2$FeNbO$_6$ at $\approx 9$K.$^{10}$

Determination of the coefficients $\mathbf{\beta}_{ijk}$ of Pb$_2$FeTaO$_6$. For one of the 8 possible trigonal domains (Fig. 3a), $P_s$, the tensor $\mathbf{\beta}_{ijk}$ (identical for 3m, 3m' and 3m''') leads to the three components of induced polarization, $P_1 = \mathbf{\beta}_{111}(H_1^2 - H_2^2) + 2\mathbf{\beta}_{113}H_1H_3$, $P_2 = -2\mathbf{\beta}_{111}H_1H_2 + 2\mathbf{\beta}_{113}H_2H_3$, $P_3 = \mathbf{\beta}_{311}(H_1^2 + H_2^2) + \mathbf{\beta}_{333}H_3^2$ along the axes of the coordinate system (Fig. 3c). Only their projections are measurable on (110)$_c$. The four equations needed for the resolution of the system are obtained from two series of measurements of $P_i$ versus the angle formed between the magnetic field $H_0$ and a sample reference direction. The magnet was rotated around a different crystallographic axis for each set of measurements (Fig. 4).

By poling along [110]$_c$ mainly domains $P^1_s$ and $P^2_s$ occur (Fig. 3b), but with varying domain weight ratios $X/Y$ depending on history as seen...
FIGURE 1 Induced polarization on PCW versus temperature at magnetic field $H_o = 0.5/T/[110]$, (100) _c -cut; 0.063 mm x 4.58 mm$^2$

FIGURE 2 Induced polarization in PFT versus temperature at maximum field $H_o = 1/T/[110]$, (100) _c -cut; 0.023 mm x 5.53 mm$^2$

FIGURE 3 Possible domain states of $P_s$ in a (110) _c section of trigonal ferroelectric Pb$_2$FeTaO$_6$; a) zero electric field, b) electric field $// [110]$, c) coordinate system for $P_s$

FIGURE 4 Induced polarization of PFT versus the field directions; $H_o$ rotating around a) [110] _c; b) [100] _c, resp.; same sample as in Fig. 1
under polarized light. Table 1 gives $\beta_{ijk}$ values for some observed $X/Y$ ratios. Their order of magnitude comes close to that found for $\text{Pb}_2\text{NbFe}_0\text{O}_6$.

Table 1 Values of $\beta_{ijk} \times 10^{-17}$ s/A of PFT at 4.4K ($X + Y < 1$ because of the presence of some domain states other than $P_1^s$ and $P_1^p$)

<table>
<thead>
<tr>
<th>$X/Y$</th>
<th>$\beta_{111}$</th>
<th>$\beta_{113}$</th>
<th>$\beta_{311}$</th>
<th>$\beta_{333}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.7/0.1</td>
<td>1.67</td>
<td>-9.25</td>
<td>-1.09</td>
<td>1.72</td>
</tr>
<tr>
<td>0.6/0.1</td>
<td>1.81</td>
<td>-10.67</td>
<td>-1.35</td>
<td>2.16</td>
</tr>
<tr>
<td>0.5/0.1</td>
<td>1.93</td>
<td>-12.61</td>
<td>-1.73</td>
<td>2.83</td>
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