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The pressure effect on the magnetic commensurability and ferroelectricity in multiferroic $HoMn_2O_5$

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Abstract

The multiferroic $HoMn_2O_5$ is a frustrated magnetic system with a non-collinear commensurate structure in its ferroelectric state. This work presents polarization measurements under isotropic pressure showing the stabilization of the high ferroelectric polarization state. This is associated with the pressure induced phase transition from an incommensurate magnetic structure to a commensurate one once the critical pressure is passed as shown by elastic powder neutron diffraction measurements. In addition, the small increase in the polarization upon spin reorientation in the commensurate magnetic state of $HoMn_2O_5$ is associated with a change in the magnetic structure without change in commensurability.

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Multiferroic RMn_2O_5 (R = rare earth,Y) manganites have recently attracted much attention due to the strong correlation between the magnetic order of the Mn spin system and the ferroelectricity [1]. The sizable magneto-electric coupling in these systems has been an object of intense investigations for fundamental reasons as well as for their prospective practical applications. The coupling of the frustrated magnetic system to the lattice, that drives the ferroelectricity along the *b*-axis results to a highly degenerate ground state. The system undergoes a cascade of phase transitions at $T < 45$ K where the phases are highly susceptible to perturbations such as an applied magnetic field [2,3]. This has been shown through strong magnetic field effects on the magnetic structure, dielectric constant, ferroelectric polarization as well as the lattice parameters [4–6]. In this work, as opposed to coupling directly to the spin structure via application of a magnetic field, isotropic pressure was applied to the system to tune the inter-atomic distances and consequently the magnetic exchange interactions between the Mn spins. The pressure effect on the commensurability of the magnetic order and on the ferroelectric polarization (**P**) was determined.

High quality single crystals of $HoMn_2O_5$ were made via the high temperature $PbO-PbF_2$ flux method as discussed elsewhere [2]. The ferroelectric **P** was determined from pyroelectric current measurements where the crystal was polished to an area of $13\,mm^2$ in the $a-c$ plane with a thickness of 0.3 mm along the *b*-axis. Au electrodes were then evaporated on the surfaces such that measurements were done parallel to the *b*-axis. The pressure was applied hydrostatically up to 18 kbar using the clamp cell method [7]. The sample in the pressure cell was field-cooled to 5 K under an external electric field of 330 kV/m. The pyroelectric current was then measured upon zero field warming at the rate of ~1 K/min.

To investigate the pressure effect on the commensurability of the magnetic structure across the phase transitions, high pressure neutron diffraction was done on

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polycrystalline $HoMn_2O_5$ in the BT1 high-resolution neutron powder diffractometer at the NIST Center for Neutron Research. The measurements were done using a helium gas loaded aluminum cell at ambient pressure and at 6 kbar.

Fig. 1 shows the effect of isotropic pressure on the measured $\mathbf{P}(T)$. At ambient pressure, the data shows the onset of ferroelectricity at T_{C1} (CM/FE1), coinciding with the lock-in transition of the magnetic order from incommensurate to commensurate. The temperature profile of $\mathbf{P}$ also shows a slight increase at T_{N2}, as well as the sudden destabilization to a lower $\mathbf{P}$ at T_{C2} (LT-ICM/FE2) which is consistent with previous reports [4]. Upon application of isotropic pressure, the phase transitions associated with the largest lattice strain (T_{C1} and T_{C2}) exhibit the expected shift in critical temperatures showing the tunability of these phase transitions by pressure. On the other hand, the most striking pressure effect was observed in the LT-ICM/FE2 phase where the measured magnitude of $\mathbf{P}$ increased dramatically with increasing pressure which effectively induces a phase transition into the high ferroelectric $\mathbf{P}$ CM/FE1 phase. Full quenching of the phase transition was observed at about 5 kbar, consistent with the pressure range where the associated dielectric anomaly was quenched as well [8].

It is suggested that the origin of these observed changes in the ferroelectric order is magnetic in nature. That is, the high ferroelectric $\mathbf{P}$ state results from the stabilization of a commensurate magnetic structure which appears to be favored under isotropic pressure. In this work, this was confirmed by measuring the magnetic ordering wave vector ($\mathbf{q}_m$) under isotropic pressure. Fig. 2 shows the pressure induced change in the magnetic structure, from the incommensurate (LT-ICM/FE2) phase at ambient pressure to the commensurate (CM/FE1) phase once the applied pressure was above the critical pressure at 6 kbar. Furthermore, the measurement of the integrated intensity of two orthogonal magnetic peaks (Fig. 2 inset) shows opposing behaviors with decreasing temperature. This signifies that the phase transitions at T_{N2} and T_{C2} correspond to Mn spin reorientations with the commensurability of the magnetic structure changing only at T_{C2} at ambient pressure.

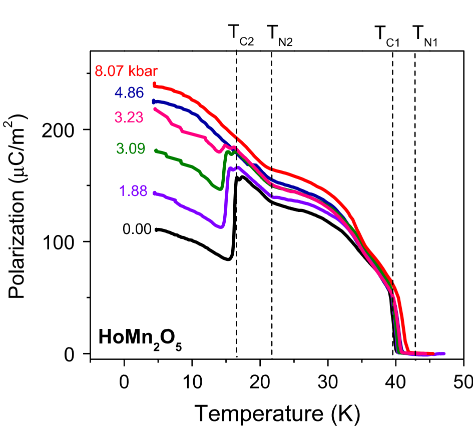

Fig. 1. Pressure effect on the temperature profile of the ferroelectric polarization in $HoMn_2O_5$ showing the quenching of the phase transition into the LT-ICM/FE2 phase above 5 kbar.

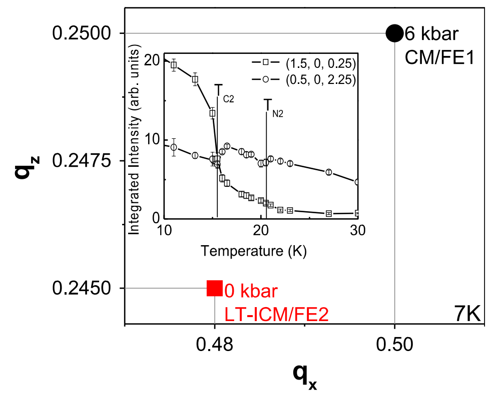

Fig. 2. Effect of pressure on the magnetic structure commensurability characterized by the magnetic wave vector $\mathbf{q}_m = (q_x, 0, q_z)$. A pressure induced phase transition from the incommensurate LT-ICM/FE2(0 kbar) to the commensurate CM/FE1 (6 kbar) phase was observed at 7 K. Inset shows evidence for the Mn spin reorientation at T_{N2} and T_{C2}.

The observed correspondence between the pressure effect in the $\mathbf{P}(T)$ and $\mathbf{q}_m$ exemplifies the strength of the magnetodielectric coupling in $HoMn_2O_5$. The determination of changes in the specific magnetic spin structure under pressure is needed for a microscopic picture of this correlation. In addition, the low temperature phase LT-ICM/FE2 in other RMn_2O_5's has been recently associated with a new elementary excitation, the electromagnon [9], such that the observed suppression of this low temperature phase may signify an inhibition of the said excitations as well.

The stabilization of the commensurate magnetic structure was also shown recently by the application of magnetic fields along the crystal's easy axis [4]. Furthermore, a critical magnetic field of around 14 T for $HoMn_2O_5$ was able to increase the ferroelectric $\mathbf{P}$ in the LT-ICM/FE2 such that no destabilization from the high $\mathbf{P}$ phase was observed. These observations under applied magnetic field, coupled with our measurements with applied isotropic pressure show the complementing effect of both perturbations in stabilizing a commensurate magnetic structure with a high ferroelectric $\mathbf{P}$ in $HoMn_2O_5$.

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