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Control of ferroelectric polarization in multiferroic YMn_2O_5 by external pressure

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Abstract. We show that the application of external hydrostatic pressure can reverse the ferroelectric polarization of multiferroic YMn_2O_5 at low temperatures. This unique effect is explained by the peculiarities of the highly frustrated magnetic order that is typical for all RMn_2O_5 (R=rare earth) in the commensurate and incommensurate phases. Our results indicate that the pressure changes the relative phase of adjacent antiferromagnetic spin chains propagating along the orthorhombic a-axis. With further compression the incommensurate phase transforms into the commensurate phase with the highest ferroelectric polarization.

1. Introduction

Multiferroic magnetoelectric materials in which various magnetic orders coexist with ferroelectricity have recently attracted considerable attention because of a wealth of new physical phenomena that originate from the strong correlation and mutual interaction of both fundamental orders and the coupling with the lattice degrees of freedom. Ferroelectricity in these materials can be induced by frustrated magnetic structures either through strong coupling of a helical, inversion symmetry breaking spin density wave with the lattice or through the displacement of ordered charges lifting the frustration of the magnetic system [1]. All multiferroic materials are highly frustrated either due to geometric constraints or to competing magnetic exchange interactions. This makes them extremely susceptible with respect to perturbations (e.g. magnetic field, pressure) since different magnetic states are close in energy and compete for the ground state. Accordingly, most multiferroics exhibit complex magnetic and ferroelectric (FE) phase diagrams with involving commensurate (CM) and incommensurate (IC) magnetic modulations as well as collinear and noncollinear spin structures. A typical example are the RMn_2O_5 compounds (R=rare earth, Y, Bi) which show at least three magnetic/ferroelectric phase transitions: At $T_{N1} \simeq 44$ K the Mn-spins order with an IC modulation along the a- and c-axes. This order locks into a commensurate modulation at $T_{C1} \simeq 39$ K with the onset of ferroelectricity along the b-axis. At lower temperatures, $T_{C2} \simeq 15$ to 22 K, the magnetic order becomes IC again accompanied by a sizable drop of the FE polarization [2, 3]. The dielectric constant, $\epsilon(T)$, exhibits a sharp peak at T_{C1} and a pronounced step at T_{C2} .

YMn_2O_5 is unique among all RMn_2O_5 since it was reported that the FE polarization reverses sign at T_{C2} , the low-temperature transition into an IC magnetic structure [2]. However, this is difficult to prove since the FE domains have to be aligned by an external electric field in order to measure the intrinsic value of the polarization. In passing through a first order phase transition at T_{C2} in YMn_2O_5 , the domain alignment may get lost, FE domains may flip and the global FE polarization will follow the external field, making it impossible to experimentally verify an intrinsic polarization reversal. Another obscure property of the FE state in RMn_2O_5 makes this task even more difficult. It was shown for R=Ho, Er, and Y [2, 4, 5] that the FE polarization in the commensurate phase follows a preferred orientation along one of the two possible directions along the b-axis. The origin of this asymmetry is not yet understood but it may be related to extrinsic effects such as structural defects that cannot easily be removed by annealing. In order to overcome these difficulties and to measure the effects of external pressure on the FE polarization $P(T)$ in YMn_2O_5 we have developed a procedure that guarantees the accurate measurement of $P(T)$ in the commensurate as well as the IC low-temperature phases.

2. Polarization at ambient pressure

A well established procedure of measuring the FE polarization in multiferroics is the pyroelectric current method. After cooling in a poling electric field to align the FE domains the current arising from a change of the polarization with temperature or magnetic field is detected by a sensitive ammeter in zero electric field. This method works well for most ferroelectrics if the poling field is high enough and the polarization is large at the lowest temperature. However, problems arise if the polarization at the temperature where the electric field is to be released is actually zero like in DyMn_2O_5 . In this case, pyroelectric measurements taken with a small poling voltage applied can still provide a meaningful result for the intrinsic polarization [6]. If the polarization changes sign as suggested for YMn_2O_5 the pyroelectric measurements can be difficult. In the measurement by Inomata et al. [2] the poling voltage was released at 4.2 K, i.e. below the reversal temperature, T_{C2} , and the current was recorded upon heating. The low-temperature polarization changed the sign with the sign of the poling voltage as expected, however, the polarization in the commensurate phase ($T_{C2} < T < T_{C1}$) appeared to be of the same sign independent of the sign of the poling voltage. We have observed similar results, shown in Fig. 1. The poling voltage obviously determines the sign of P below T_{C2} , but the domain alignment gets lost in warming through the $\text{ICM} \rightarrow \text{CM}$ phase transition and P in the CM phase is determined by the structural preference, as discussed above. From the previous data [2] and those of Fig. 1 it is not possible to conclude about a sign reversal of the FE polarization in YMn_2O_5 . In order to determine the intrinsic value of P in the CM phase the poling voltage has to be released at a temperature T_p above T_{C2} and the pyroelectric current can then be measured upon warming in zero bias voltage. We have chosen $T_p=18.7$ K which is close to the maximum of P near the transition at T_{C2} . The polarization in the low-T ICM phase is determined in a similar manner: The sample was cooled in electric field to T_p at which the poling voltage was released. The pyroelectric current was then measured in zero bias upon further cooling to 5 K. The polarization thus obtained for the whole temperature range is shown in Fig. 2. The open and solid symbols represent data upon cooling and warming, respectively, measured in zero bias from 18.7 K. Obviously, the intrinsic polarization in the CM phase is three times as large as obtained from the 5 K poling procedure ([2] and Fig. 1). Our results also prove unambiguously the sign reversal of the FE polarization at T_{C2} . In order to verify that the above method of poling (to 18.7 K) and pyroelectric measurement also reflects the true magnitude of the polarization in the low-T ICM phase we have checked the value of $P(T)$ at 5 K by employing an even more sophisticated poling procedure. The FE domains in the CM phase were aligned through cooling in a positive bias electric field right into the phase transition at T_{C2} . At T_{C2} , the poling voltage was reversed to support the domain alignment at lower temperatures after

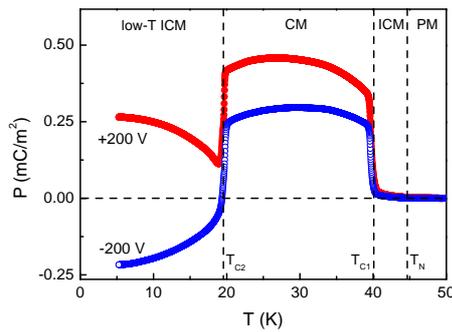


Figure 1. Polarization when poling in ± 200 V at 5 K.

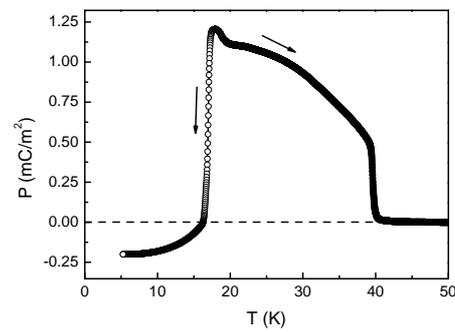


Figure 2. Polarization when poling in 200 V at 18.7 K.

$P(T)$ has switched sign. The negative bias was released to zero at 5 K and the polarization below T_{C2} was measured in zero bias upon warming. The obtained $P(5\text{ K})$ was only slightly larger than that of Fig. 2 which shows that the FE domain alignment was mainly preserved in cooling through T_{C2} . In all following polarization measurements at ambient and high pressure the poling method described above (i.e. poling to 18.7 K) and the procedure used to acquire the pyroelectric current resulting in the polarization data of Fig. 2 was employed.

3. Pressure effect on the ferroelectric polarization

Hydrostatic pressure was applied to the YMn_2O_5 single crystal up to 20 kbar. The FE polarization was measured as described in the previous section. The results shown in Fig. 3 reveal an interesting pressure effect on $P(T)$ in the low-T IC phase: The polarization reverses sign at a critical pressure of about 10 kbar. Between 10 and 14 kbar $P(T)$ still exhibits a step-like decrease at T_{C2} , however, at higher pressure there is no anomaly of $P(T)$ in this temperature range. The pressure dependence of $P(T)$ at a fixed temperature (5 K) is shown in Fig. 4. It is clear that the sign reversal of $P(5\text{ K})$ happens in a very narrow pressure range close to 10 kbar. This unique effect of pressure on the FE polarization has not been observed before and it is intimately related to the peculiarities of the magnetic orders in RMn_2O_5 .

The FE polarization in RMn_2O_5 is mainly determined by the degree of magnetic frustration between neighboring zigzag chains of Mn^{3+} - Mn^{3+} - Mn^{4+} ions propagating along the orthorhombic a -axis [7]. In the CM phase between T_{C1} and T_{C2} all spins within one chain are antiferromagnetically ordered and in the low-T ICM phase ($T < T_{C2}$) the spins form a perfect spin density wave along the a -axis with an IC modulation. Two adjacent chains are coupled through the antiferromagnetic superexchange interactions of two pairs of Mn^{3+} - Mn^{4+} spins within each unit cell [8]. Due to the specifics of the lattice structure and the magnetic order one pair of spins is frustrated whereas the second pair is not [8, 9]. The magnetic frustration can be reduced in moving the frustrated spins away from each other resulting in local electrical dipolar momenta that add up to a macroscopic polarization along the b -axis [9, 7]. This is the physical origin of the improper ferroelectricity induced by magnetic order/frustration in RMn_2O_5 .

The magnitude of the FE polarization depends on the angle of the frustrated magnetic moments and the relative phase of the spin density wave on adjacent chains. Neutron scattering measurements of the magnetic orders in YMn_2O_5 have revealed that the angle between spins of neighboring chains increases from 15° to 40° in passing into the low-T IC phase which reduces the magnitude of frustration and explains the drop of the FE polarization at T_{C2} that is typical for all RMn_2O_5 compounds [7]. In addition there is a phase shift of the spin density waves of every second chain that results in a shift of the major frustration from one Mn^{3+} - Mn^{4+} bond in the unit cell to the second bond which reverses the sign of the polarization. In view of these

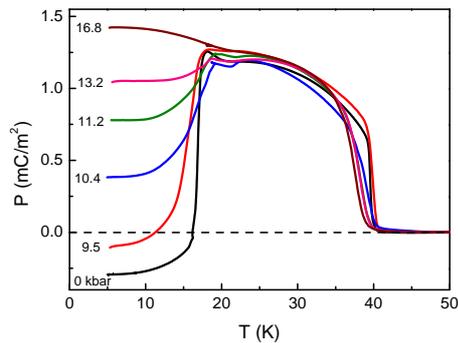


Figure 3. Ferroelectric polarization of YMn_2O_5 at various pressures.

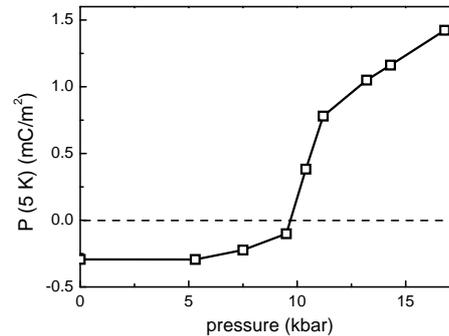


Figure 4. Sign reversal of the polarization at 5 K as function of pressure.

results we conclude that the effect of external pressure as observed by us is twofold:

(i) With increasing pressure up to about 10 kbar the relative phase of the spin order on two adjacent zigzag chains is decreased in the low-T IC phase. This results in a sudden reversal of the polarization at the critical pressure. It appears that the $P=0$ state is not thermodynamically stable since the jump from $P<0$ to $P>0$ (Fig. 4) is very fast.

(ii) With further increasing pressure the commensurability changes and above about 14 kbar the transition from the CM to the low-T IC phase is completely suppressed. The pressure stabilizes the commensurate magnetic order in RMn_2O_5 multiferroics as was recently confirmed for HoMn_2O_5 by high-pressure neutron scattering experiments [10, 11]. The polarization at high pressure increases smoothly upon decreasing temperature with no detectable change near T_{C2} .

4. Summary

We have shown that external hydrostatic pressure has a unique and remarkable effect on the low-temperature ferroelectric polarization of multiferroic YMn_2O_5 . At a critical pressure of 10 kbar the FE polarization changes sign and increases with further increasing pressure. The phase transition from the commensurate to low-T incommensurate magnetic phase is suppressed and, at high pressures, the low-T phase is proposed to be commensurate at all temperatures below T_{C1} . This needs to be confirmed by neutron scattering experiments.

Acknowledgments

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