Large off-diagonal magnetoelectric coupling in the quantum paraelectric antiferromagnet $EuTiO_3$

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(Received 9 October 2009; revised manuscript received 21 December 2009; published 23 February 2010)

The third-order E^2H^2-type magnetoelectric (ME) response of polycrystalline $EuTiO_3$ changes sign under magnetic bias and shows a large anomaly at the antiferromagnetic (AF)-paramagnetic phase boundary below $T_N \approx 5.3$ K. It is attributed to critical fluctuations of the AF order parameter reinforced by quantum paraelectric polar correlations. The underlying biquadratic spin-lattice coupling involves electric field induced Dzyaloshinskii-Moriya interaction as described within mean-field approximation. Single domaining by ME annealing (or cooling) significantly enhances the response by additional EH and EH^2 effects.

DOI: 10.1103/PhysRevB.81.064426 PACS number(s): 77.22.−d, 75.30.Et, 75.50.Ee, 77.84.Cg

I. INTRODUCTION

Recent years have seen a revival of the magnetoelectric (ME) effect,[1] which stands for the coupling between electric and magnetic properties of matter as conjectured by Curie[2] and first observed by Astrov[3] after predictions of Dzyaloshinksii[4] on crystalline Cr_2O_3. In the case of linear ME coupling an electric field $\boldsymbol{E}$ induces a magnetic polarization, $\delta(\mu_0 \boldsymbol{M}) = \boldsymbol{\alpha E}$ ($\boldsymbol{M}$=magnetization), while an electric polarization, $\delta \boldsymbol{P} = \boldsymbol{\alpha H}$, arises in a magnetic field $\boldsymbol{H}$ ($\boldsymbol{\alpha}$=linear ME susceptibility tensor). The maximum coupling, $\alpha_{zz} \approx 4 \times 10^{-12}$ s/m,[3] occurs at $T \approx 260$ K and denotes an extremely small average spin-flip rate of $\approx 5 \times 10^{-7}$ spins/(kV/cm).[5]

It was hoped[1] that *multiferroics*,[6] i.e. materials revealing simultaneously (anti)ferroelectric and (anti)ferromagnetic long-range order, might give rise to much larger ME susceptibility. Since $(\alpha_{ij})^2 \leq \mu_0 \chi^m_{ii} \varepsilon_0 \chi^e_{jj}$,[7] it promises to maximize at large magnetic and electric susceptibilities, χ^m_{ii} and χ^e_{jj}, respectively. "Giant" ME coupling is in reach, if joint ferromagnetic (FM) and (proper) ferroelectric criticality occurs in the vicinity of nearby magnetic and electric Curie temperatures, $T^m_C \approx T^e_C$.[8] However, no existing material even roughly fulfills this condition. In this paper we propose instead to choose a quantum paraelectric material coming close to FM instability (large χ^m_{ii}) in its fluctuation regime (large χ^e_{ii}).

This opportunity is offered by $EuTiO_3$, which attains G-type antiferromagnetic (AF) ordering with two interpenetrating fcc FM sublattices below $T_N \approx 5.35$ K. Large magnetic susceptibilities are warranted owing to strong FM next-nearest-neighbor exchange interaction ($J_2 > 0$). It competes with the AF nearest-neighbor one ($J_1 < 0$) and gives rise to a positive *paramagnetic* (PM) Curie temperature, $\theta = +3.8$ K.[9] Indeed, the magnetic perpendicular susceptibility measured on our polycrystalline sample of $EuTiO_3$ (Ref. 10) reaches $\chi^m_\parallel = \frac{\delta m}{V dH} \approx 40$ at $T \approx 5.3$ K [Fig. 1(a)], and the dielectric susceptibility yields $\chi^e_\parallel \approx 400$ at the same temperature.[11] As will be reported below, the ME coupling of $EuTiO_3$ turns out to be pretty large as expected from the spontaneous decrease in the dielectric permittivity by ≈7% below T_N following the simple magnetocapacitance formula:[11]

$$\varepsilon(H) = \varepsilon(H=0)(1 + \bar{\delta}\langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j \rangle), \tag{1}$$

where $\langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j \rangle$ is the nearest-neighbor spin-pair-correlation function and $\bar{\delta}$ a coupling constant. While Eq. (1) is equivalent to a symmetry-allowed third-order ME [or E^2H^2; see Eq. (3) below] effect[12] we have found after proper ME field treatment additional large bilinear (EH) and quadratic (EH^2) couplings, which reflect electric field induced symmetry breaking as in yttrium iron garnet.[13] Most unexpectedly, however, nondiagonal spin-spin coupling is encountered at magnetic fields close to the AF-to-PM transition. Electric field induced Dzyaloshinskii-Moriya (DM) interaction becomes active in $EuTiO_3$ analogously to that predicted[14] and

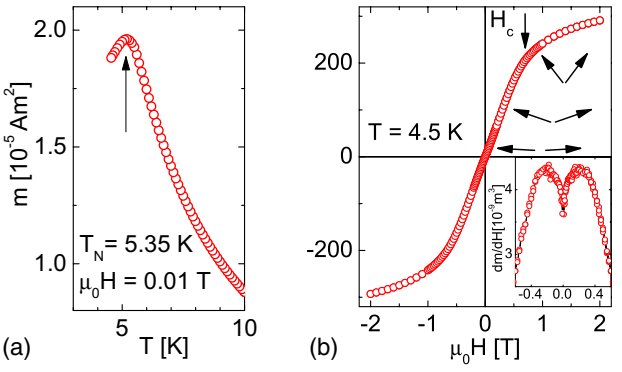

FIG. 1. (Color online) Magnetic moment of a polycrystalline sample of $EuTiO_3$ obtained by SQUID [MPMS 5S, Quantum Design] (a) in $\mu_0 H = 0.01$ T at temperatures $4.5 \leq T \leq 10$ K, and (b) at $T = 4.5$ K in magnetic fields $|\mu_0 H| \leq 2$ T. Arrows in (b) indicate the field evolution of the spin-flop phase and the AF-to-PM transition, respectively. Inset to (b): $\mu_0 H$ dependence of the susceptibility dm/dH.

observed in ferroelectric $FeTiO_3$,[15] and gives rise to giant ME peaks with inverted sign.

II. EXPERIMENTAL RESULTS AND DISCUSSION

A. Magnetoelectricity in $EuTiO_3$

To begin with, the lack of a polar instability down to $T=0$ prevents $EuTiO_3$ to emerge immediately victorious from the race for "giant" ME coupling. Inversion symmetry forbids both the bilinear EH (or α)[16] and the second-order electrobimagnetic $EH^2(\beta)$ effects.[17] Without ME cooling or annealing[5] only the third-order bielectrobimagnetic $E^2H^2(\delta)$ effect occurs, as expected for ferroelectromagnets of all symmetries.[18]

By assuming distance dependence of the exchange interactions, Jiang and Wu[19] derived a biquadratic spin-pseudospin Hamiltonian corresponding to the $E^2H^2(\delta)$ effect,

$$H^{me} = -\delta \sum_{l} \sum_{(i,j)} \sigma_l^2 \boldsymbol{S}_i \cdot \boldsymbol{S}_j, \tag{2}$$

where σ_l are the soft-mode displacements of the Ti^{4+} ions,[20] $\boldsymbol{S}_{i,j}$ the Heisenberg spin vectors of the Eu^{2+} ions ($S=7/2$), and δ a common coupling coefficient. Implementation of Eq. (2) into conventional soft-mode theory[19] finally yields the *magnetocapacitance* effect, Eq. (1), where $\bar{\delta} \propto \delta$. The same issue was addressed by Fennie and Rabe[12] within density-functional theory.

The biquadratic coupling constant δ in Eq. (2) is related to the third-order susceptibility tensor (δ_{ijkl}) entering the Landau free-energy density expansion[8] under Einstein summation

$$F(\boldsymbol{E},\boldsymbol{H}) = F_0 - \frac{1}{2}\varepsilon_0\varepsilon_{ij}E_iE_j - \frac{1}{2}\mu_0\mu_{ij}H_iH_j - \alpha_{ij}H_iE_j - \frac{\beta_{ijk}}{2}E_iH_jH_k - \frac{\gamma_{ijk}}{2}H_iE_jE_k - \frac{\delta_{ijkl}}{2}E_iE_jH_kH_l, \tag{3}$$

in addition to other possible ME coupling terms. In our experiments the electric field induced magnetization components of

$$\mu_0 M_i = -\partial F/\partial H_i = \mu_0\mu_{ij}H_j + \alpha_{ij}E_j + \frac{\gamma_{ijk}}{2}E_jE_k + \beta_{jki}E_jH_k + \delta_{jkli}H_jE_kE_l \tag{4}$$

are measured using ME superconducting quantum interference device susceptometry.[21] It involves ac and dc electric and magnetic external fields, $E=E_{ac}\cos\omega t + E_{dc}$ and H_{dc}, and records the first harmonic complex ac magnetic moment, $m(t)=(m'-im'')\cos\omega t$, where

$$m' = (\alpha E_{ac} + \beta E_{ac}H_{dc} + \gamma E_{ac}E_{dc} + 2\delta E_{ac}E_{dc}H_{dc})(V/\mu_0) \tag{5}$$

(V=sample volume) and $|m''| \ll |m'|$ at the measurement frequency $f=\omega/2\pi=1$ Hz. Orientation averaged coupling parameters α, β, and δ have been measured after different cooling protocols at $T<10$ K, hence, close to T_N.

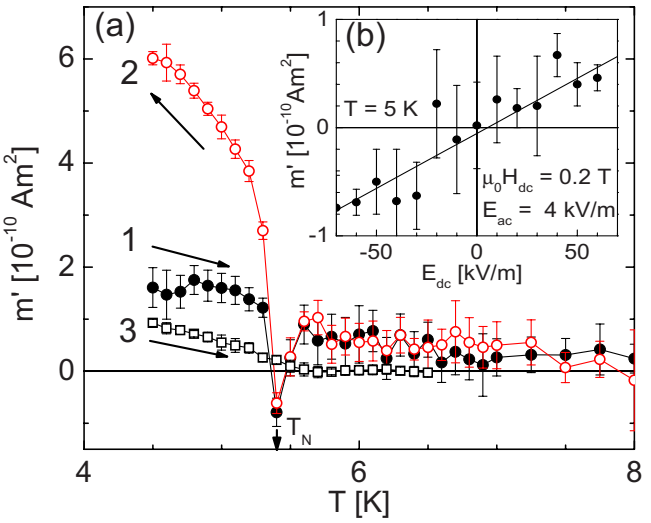

FIG. 2. (Color online) (a) Magnetic moment m' vs T induced by $E_{ac}=8$ kV/m within $4.5 \le T \le 8$ K after ZFC from 70 to 4.5 K measured (1) on FH and (2) on subsequent FC in $E_{dc}=80$ kV/m and $\mu_0 H_{dc}=0.2$ T, and (3) on subsequent FH in $E_{dc}=80$ kV/m and $\mu_0 H_{dc}=0$. (b) m' vs E_{dc} within ± 70 kV/m induced by $E_{ac}=4$ kV/m and $\mu_0 H_{dc}=0.2$ T as a function of E_{dc}.

B. Experimental results

Curve 1 in Fig. 2(a) shows the temperature dependence of the ME moment, m', driven by $E_{ac}=8$ kV/m after zero-field cooling (ZFC) in $\mu_0 H_{dc}=0=E_{dc}$ on field heating (FH) in $\mu_0 H_{dc}=0.2$ T and $E_{dc}=80$ kV/m (all fields in z direction perpendicularly to the sample face). Starting from $m'(4.5\ \text{K}) \approx 2\times 10^{-10}$ A m^2 it gradually decreases and vanishes above T_N only at $T>8$ K. According to Eq. (5) and in view of the three fields involved we obviously encounter a third-order ME signal, which is allowed to persist above the AF ordering temperature, where it senses AF correlations in the fluctuation regime. We have checked the linear dependences of m' on all three fields, as shown in Fig. 2(b) for E_{dc} at constant $E_{ac}=4$ kV/m, $\mu_0 H_{dc}=0.2$ T, and $T=5$ K. Within errors $m'(E_{dc}=0)=0$, hence, $EH^2(\beta)$ coupling can be excluded. Note that the slope of the best-fitted line, $\delta=(4.8\pm 0.8)\times 10^{-22}$ s m/V A, matches the magnetocapacitive effect[11] quantitatively.

Just below T_N at $T=5.3$ K a spikelike excursion of m' toward negative values is observed. As will be discussed below, this is a signature of the AF-to-PM transition in the applied magnetic field, $\mu_0 H_c(5.3\ \text{K})=0.2$ T. Here we notice that the Heisenberg antiferromagnet $EuTiO_3$ (Ref. 9) is easily magnetized after entering a spin-flop (SF)-like phase in finite magnetic field [Fig. 1(b): m vs H with gradually aligning spin symbols]. Peaks of the susceptibility data, $dm/dH(4.5\ \text{K})$ vs H [Fig. 1(b): inset], indicate the SF transition at $\mu_0 H_{SF}(4.5\ \text{K}) \approx \pm 0.2$ T, while the AF-to-PM transition occurs at $\mu_0 H_c(4.5\ \text{K}) \approx 0.7$ T [Fig. 1(b): vertical arrow].

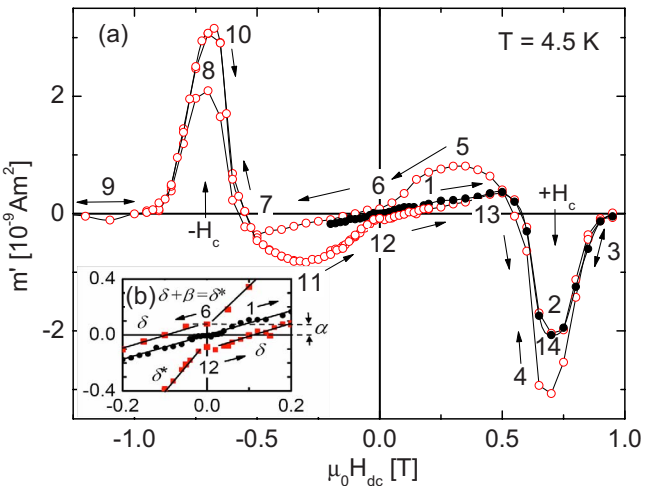

FIG. 3. (Color online) (a) Magnetic moment m' vs $\mu_0 H_{dc}$ induced by $E_{ac}=8$ kV/m measured after ZFC from 70 to 4.5 K at $T=4.5$ K under $E_{dc}=80$ kV/m and $\mu_0 H_{dc}$ from −0.2 to 1.0 T (virgin curve), back to −1.2 and +1.0 T. Arrows and numbers 1…14 denote the sequence of the hysteresis curve. (b) Expanded view of the hysteresis cycle of (a) close to $H_{dc}=0$, where α, δ, and $\delta^*=\delta+\beta$ indicate different intercepts and slopes, respectively.

When returning to low T under field-cooling (FC) in $\mu_0 H_{dc}=0.2$ T and $E_{dc}=80$ kV/m [Fig. 2(a): curve 2] the ME signal becomes tripled in the AF regime, while it remains roughly unchanged above T_N. This enormous increase is due to the ME *cooling* from above T_N, where both applied fields favor the growth of AF single domains.[5] Electric poling now allows for the bilinear $EH(\alpha)$ and the quadratic $EH^2(\beta)$ effects in addition to the bare $E^2H^2(\delta)$ effect of curve 1.

The bare α effect remains after switching off the magnetic field at $T<T_N$, where $m'(4.5\ \mathrm{K})\approx 0.9\times 10^{-10}$ A m^2 [Fig. 2(a): curve 3] corresponds to $\alpha=(6.3\pm 0.2)\times 10^{-12}$ s/m, which is of the same order of magnitude as the peak value of Cr_2O_3 at $T\approx 260$ K.[3] It is definitely due to the AF order, since it vanishes upon heating without any spiky anomaly (curves 1 and 2) at $T\approx T_N$. Its independence of weak E_{dc} (not shown) proves the absence of an E^2H (γ) effect, see Eq. (5). Its invariance at $E_{dc}=0$ evidences electric ordering due to the intense E_{dc} cooling into the quantum paraelectric regime. Obviously polycrystalline $EuTiO_3$ behaves like a soft ferroelectric owing to intrinsic impurities and lattice defects as known from quantum paraelectric $SrTiO_3$ ceramics.[22]

The enhancement of the ME effect for $H_{dc}>0$ [Fig. 2(a): curve 2] compared to the sum of the $E^2H^2(\delta)$ and $EH(\alpha)$ effects (curves 1+3) is attributed to the $EH^2(\beta)$ effect arising at finite magnetic fields, see Eq. (5). This is confirmed by the magnetic field dependence of the ME signal, m' vs H_{dc}, as shown in Fig. 3 for field cycles within $|\mu_0 H_{dc}|\leq 1.2$ T at $T=4.5$ K and $E_{dc}=80$ kV/m. The sequence of data is labeled 1…14 as will be detailed below. Its hysteretic properties are understood when noticing that symmetry breaking takes place at $\mu_0 H_{dc}\approx\pm 0.7$ T, where the AF memory gets lost or refreshed, respectively. The obvious inversion symmetry with respect to $H_{dc}=0=m'$ indicates dominance of $E^2H^2(\delta)$ and $EH^2(\beta)$ contributions being odd functions of H_{dc} in Eq. (5). Further, the ME annealing at the phase boundaries, yields $EH(\alpha)$ contributions of either sign, which warrant inversion symmetry at $H_{dc}=0$ as seen in the exploded view of Fig. 3(b).

The data start after ZFC from 70 to 4.5 K with a linear virgin curve within $|\mu_0 H_{dc}|<0.3$ T under $E_{dc}=80$ kV/m and $E_{ac}=8$ kV/m [Figs. 3(a) and 3(b), section 1]. Its slope corresponds to δ (4.5 K)$=4.8\times 10^{-22}$ s m/V A, corroborating the bare $E^2H^2(\delta)$ data of Fig. 2(a) (curve 1). At $\mu_0 H_{dc}\approx 0.5$ T, however, the curve continuously bends to negative values and peaks with $m'_{min}\approx -2.1\times 10^{-9}$ A m^2 at the critical field $\mu_0 H_c\approx 0.7$ T (section 2), whereupon the signal ascends rapidly to zero.

When decreasing the field again from 0.95 T (section 3) to below H_c, i.e., performing isothermal ME *field annealing*,[5] the negative peak recovers to an even larger magnitude, $m'(\mu_0 H_c=0.7\ \mathrm{T})=-3.1\times 10^{-9}$ A m^2. The induced ME magnetization $|M|=|m'|/V=1.6$ A/m of the sample (volume $V=2\times 10^{-9}$ m^3) exceeds that of Cr_2O_3 at 260 K under the same electric field $E_{ac}=8$ kV/m, $M=\alpha E/\mu_0=0.025$ A/m, by 2 orders of magnitude. It comes even close to the value of the ME magnetization $|M|=2.9$ A/m of the single-phase record holder $TbPO_4$.[23]

At $\mu_0 H_{dc}\approx 0.6$ T, m' changes sign again and finally yields a considerably increased slope as $H_{dc}\to 0$, $\delta^*=2.1\times 10^{-21}$ s m/V A (section 5). As mentioned above this is attributed to the additional $EH^2(\beta)$ effect induced by ME *annealing*. Closer inspection of the magnified plot in Fig. 3(b) reveals a finite-ordinate intercept, $m'(H_{dc}=0)=0.9\times 10^{-10}$ A m^2 (section 6), which is due to the $EH(\alpha)$ effect in zero magnetic field, where $\alpha=6.3\times 10^{-12}$ s/m agrees with data from Fig. 2(a). When continuing the magnetic field scan to negative values [Fig. 3(a), section 7], the slope δ^* is abruptly reduced to the bare $E^2H^2(\delta)$ contribution, hence, the $EH^2(\beta)$ response gets discontinuously lost. This is ascribed to the formation of an AF multidomain state below H_{SF} as $H_{dc}\to 0$, which does not seem, however, to break the continuity of the $EH(\alpha)$ effect. On further field cycling (sections 8…14) the ME behavior is perfectly inverted to that of sections 2…8.

C. Off-diagonal magnetoelectric coupling

The unusual near-divergent behavior of the $\delta+\beta$ effect at the AF-to-PM transition is probably the most remarkable ME feature of $EuTiO_3$. It sheds some light on the field-induced change in magnetic symmetries and requires more than the simplistic one-parameter description of Eq. (1). In the scarce literature on third-order ME susceptibility we found only one remark on its internal symmetry, supposedly being that of the elasto-optic tensor.[8] In view of the magnetic character of ME interaction we prefer to consider the second-order magneto-optic tensor with the same symmetry.[24] In a cubic crystal it contains only a few nonzero elements related to spin-pair-correlation functions.[25] Likewise this also holds for the third-order ME effect, where, e.g., δ_{zzzz} and δ_{xzxz} are linked to diagonal and off-diagonal correlation functions, $\langle S_i^z S_j^z\rangle$ and $\langle S_i^x S_j^z\rangle$, respectively. The latter ones are related to DM-like

exchange interaction creating off-diagonal spin-spin coupling as was predicted[14] and shown[15] in the rhombohedrally distorted perovskite $FeTiO_3$, whose A sites are occupied by magnetic ions (as in $EuTiO_3$), and whose unit cells are doubled by antiferrodistortive rotations of the oxygen octahedra [as predicted for $EuTiO_3$ (Ref. 26]. The most important condition for DM-like interaction, the lack of inversion symmetry, is promoted by the external $\boldsymbol{E}$ field, which is—like $\boldsymbol{H}$—perpendicular to the AF vector $\boldsymbol{L}$ in the spin-flopped phase at intermediate magnetic fields [Fig. 1(b)]. Here we propose the off-diagonal coupling to be at the origin of the giant $E^2H^2+EH^2(\delta+\beta)$ peaks [Figs. 2(a) and 3(a)].

Indeed, we have been able to deduce the salient features of $E^2H^2(\delta)$ coupling by adding a DM-like off-diagonal term to the ME interaction Hamiltonian, Eq. (2), involving two coupling constants, δ^+ and δ^-,

$$H^{me} = -\sum_k \sum_{(i,j)} (\delta^+ S_i^z \sigma_k^2 S_j^z + \delta^- S_i^z \sigma_k^2 S_j^x). \quad (6)$$

The electric field induced mean-square soft-mode amplitude $\sigma_k^2 \propto \chi_e^2 E^2$ is obtained from a transverse Ising model describing quantum paraelectric $EuTiO_3$ (Ref. 27) and applies to the four Ti^{4+} ions next to the center of gravity of a given spin pair. Inserting Eq. (6) into a Heisenberg-type magnetic Hamiltonian with competing exchange[19] and calculating the molecular fields acting in the two magnetic sublattices within Néel approximation, following[19] we straightforwardly obtain the magnetization $\langle S^z \rangle$ in the direction of the external magnetic field H,

$$\langle S^z \rangle = (12J_1 + 24\chi_e^2 E^2 \delta^+)^{-1}(-H + 24\chi_e^2 E^2 \delta^- S^2/\langle S^x \rangle). \quad (7)$$

When neglecting the E dependence of the transverse magnetization $\langle S^x \rangle$, the ME susceptibility is

$$\chi_{me} = \partial\langle S^z \rangle/\partial E = 48\chi_m^2 \chi_e^2 E(\delta^+ H + 12J_1 \delta^- S^2/\langle S^x \rangle) \quad (8)$$

where $\chi_m \equiv -(12J_1+24\chi_e^2 E^2 \delta^+)^{-1}$ and $S^2 \equiv \langle S^x \rangle^2 + \langle S^z \rangle^2$. Due to $J_1 < 0$, $\chi_{me} \propto m'(H)$ has two contributions of different sign in case of like signs of δ^+ and δ^-. The first term describes the conventional δ term $\propto EH$ as shown in Fig. 3 (section 1). The second term scales with E (but not with H!) and marks the observed divergence at $H \to H_c$, where the AF component of the spin-flop state vanishes, $\langle S^x \rangle \to 0$. We further notice: (i) the result of Eq. (8) preserves its main features (except for a constant factor), if the E dependence of $\langle S^x \rangle$ is taken into account; (ii) the $EH^2(\beta)$ effect can be treated in the same approximation by replacing $\delta^\pm$ by $\beta^\pm$ and $\chi_e^2 E^2$ by the (induced) polarization $\chi_e E$ in Eqs. (6)–(8); and (iii) off-diagonal coupling, Eq. (6), will probably allow to improve fitting the magnetocapacitive effect[11] after properly modifying Eq. (1).

III. CONCLUSION

We have shown that the AF quantum paraelectric $EuTiO_3$ bears uncommon ME features. The magnetic field dependence of the ME third-order $E^2H^2(\delta)$ effect has required a tensorial coupling scheme, which hints at novel DM-like off-diagonal exchange interaction under ME bias. The ME moments induced at the AF-to-PM phase transition compare to those found in "strong" single-phase first-order magnetoelectrics such as $TbPO_4$. Single domaining by ME annealing (or cooling) significantly enhances the response by additional $EH(\alpha)$ and $EH^2(\beta)$ effects. Detailed symmetry analysis will need additional experiments on single crystals.

ACKNOWLEDGMENTS

Financial supports by EU (STREP Grant No. NMP3-CT-2006-032616 "MULTICERAL"), Deutsche Forschungsgemeinschaft (SFB 491), and Czech Science Foundation (Project No. 202/09/0682) are gratefully acknowledged.

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