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Magnetodielectric coupling in a Ru-based 6H-perovskite, $Ba_3NdRu_2O_9$

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A large spin-orbit coupling is a way to control strong magnetodielectric (MD) coupling in higher *d*-orbital materials. However, reports are rare on such compounds due to often leaky conductive behavior. Here, we demonstrate MD coupling in a Ru-based 6H-perovskite system, $Ba_3NdRu_2O_9$. The rare-earth ion in a 6H-perovskite makes the system insulating enough to carry out MD investigation. The compound is ferromagnetically ordered below 24 K (T_C), followed by another magnetic feature at T ~17 K (T_2). The dielectric constant clearly traces the magnetic ordering, manifesting a peak at the onset of T_C, which is suppressed by the application of an external magnetic field (H). The results indicate the presence of MD coupling in this compound, which is further confirmed by the H-dependence of the dielectric constant. Interestingly, a cross-over of the sign of MD coupling is observed at T ~ T_2. We conclude that two different mechanisms control the MD coupling, which yields positive and negative coupling, respectively. Both mechanisms are competing as a function of temperature and magnetic field. This brings us a step closer to design and control the magnetodielectric effect in 6H-perovskites containing higher *d*-orbital elements. *Published by AIP Publishing.* https://doi.org/10.1063/1.5034449

The research on higher *d*-orbital materials (4*d*/5*d*) has attracted significant attention due to their exotic magnetic behavior as a result of various competing effects like extended orbitals, pronounced crystal-field effects, and strong spin-orbit coupling.[1] The higher *d*-orbital is also of interest in the field of magneto(di)electric coupling (MD) and multiferroicity due to the possibility of large MD coupling originating from the enhanced spin-orbit coupling. Systems, crystallizing in the perovskite structure exhibiting magnetic frustration and containing rare-earth (R) and 3d metal ions (such as $RMnO_3$, R = Ho, Tb, etc.), are vastly explored in the field of multiferroicity and MD coupling.[2] It has been theoretically predicted that 3*d*–5*d* double perovskite systems, such as Bi_2MReO_6 (M = 3d metal) and Zn_2FeOsO_6, could serve as good multiferroic and magnetoelectric systems as a result of a higher *d*-orbital.[3,4] The investigation through the non-contact spectroscopic method also supports this framework.[5] However, experimental reports of MD effects in magnetic 4*d* or 5*d* materials are rare in the literature,[6] because these systems are less insulating due to the large extended *d*-orbitals leading to leaky conductive behavior. This hampers the investigation of bulk dielectric/ferroelectric properties experimentally. The compound $Co_4Nb_2O_9$ exhibits giant MD coupling[7] though Nb is non-magnetic here and it may not have any direct role in MD coupling. However, no attention has been paid to multiferroicity and/or MD coupling in the 6H-perovskite system containing higher *d*-orbital (magnetic) ions, such as $Ba_3MRu_2O_9$ (M = 3d-ion, Bi, Y, La, R, etc.).[8–12] It is experimentally observed that the introduction of rare-earth ion (in place of Bi/La/Y) makes the system more insulating due to the localized atomic nature of rare-earth. For example, Haldane spin-chain system Y_2BaNiO_5 is not highly insulating and does not show any MD coupling or multiferroicity,[13] but R_2BaNiO_5 reveals highly insulating behavior and exhibits multiferroicity with strong MD coupling.[14] Also, *d*-*f* magnetic correlation plays a significant role in the complex magnetism and thus could have an interesting effect on MD coupling.

We show that $Ba_3NdRu_2O_9$ is a promising system revealing highly insulating behavior even at temperature below magnetic ordering, allowing the investigation of its dielectric properties. We have performed dc and ac magnetization and dielectric and magnetodielectric measurements as a function of temperature and magnetic field. There is no earlier report of multiferroicity/magneto(di)electric coupling in this $Ba_3RRu_2O_9$ system or even in a Ru-based system.

The system with general formula $Ba_3MRu_2O_9$ crystallizes in the 6H-perovskite structure and consists of Ru_2O_9 dimer (face-sharing distorted RuO_6 octahedral) and regular corner sharing the MO_6 octahedral.[8,9] Depending on the M-ion, the system $Ba_3MRu_2O_9$ exhibits different magnetic behaviors; for M= non-magnetic ion, it behaves like a dimer (Ru_2O_9) system,[15] and for the magnetic ion (M= Ni^{+2}/Co^{+2}), another magnetic super-exchange path Ru-O-M is established and long-range magnetic ordering is developed.[6] If M is replaced by lanthanides (Y^{+3}, La^{+3}, and R^{+3}), the system can crystallize in the 6H-perovskite structure and charge neutrality is balanced by changing the effective valence state of Ru (+4.5, one Ru-site is Ru^{4+} and the other site is Ru^{+5} in the Ru_2O_9 dimer).[9,10] This mechanism is confirmed by neutron diffraction as well.[11] Exceptionally, only in the case of $Ba_3Bi(Ru/Ir)_2O_9$, Bi possesses an unusual valence state of +2, rather than +3, and Ru has a usual +5 state.[16] Interestingly, the compound containing M = 3d-metal ion/Bi is semiconducting in nature, whereas the introduction of R-ions makes the system a rather good insulator. The title compound $Ba_3NdRu_2O_9$ is ferromagnetically ordered below 24 K (T_C), followed by another complex

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magnetic feature at ~17 K (T_2), which is attributed to a canted antiferromagnetic (AFM) ordering.[10,11] Magnetic frustration was already predicted in this compound.[11] Hence, this is a good system to explore MD coupling.

The compound $Ba_3NdRu_2O_9$ is synthesized by solid-state-reaction and forms a single phase with the expected P63/mmc space group, as reported earlier.[10] The temperature (T) and magnetic field (H) dependent dc and ac magnetic susceptibility (χ) measurements are performed using a Superconducting Quantum Interference Device (SQUID, Quantum Design). The complex dielectric measurement has been carried out as a function of temperature and magnetic field with a 1 V ac bias for various frequencies using an LCR meter (Agilent 4284A) with a home-made sample insert for a Physical Properties Measurement System (PPMS, Quantum Design). Silver paint is used to make the parallel plate capacitor of a pressed disc-like polycrystalline sample (5 mm diameter and 0.9 mm thickness).

The preliminary dc magnetic results were already reported by Doi *et al.*;[10] however, here we document detailed dc magnetization elucidating the magnetic behavior and for comparison to check the quality of the sample (see supplementary material for detailed results and discussion). In the present manuscript, we focus on the magnetic behavior in the presence of high magnetic fields and far above the ordering temperature T_C, which was not reported so far. Further ac susceptibility measurements in the presence of different dc magnetic fields are performed. The real (χ') and imaginary (χ'') parts of the ac susceptibility are shown in Fig. 1, as a function of temperature (T) for a fixed frequency of 111 Hz and in the presence of different dc magnetic fields (H_{dc}). The distinct peak anomaly in zero dc magnetic field (H_{dc}) at T_C in both $\chi'(T)$ and $\chi''(T)$ confirms the magnetic ordering [Figs. 1(a) and 1(d)]. The application of an external dc field, H_{dc}, decreases the magnitude of $\chi'(T)$ and the peak becomes broader and shifts towards higher temperature with increasing H_{dc} [Figs. 1(a), 1(b), and 1(c) for $H_{dc} = 0$, 10, and 50 kOe, respectively]. Obviously, $\chi''(T)$ is almost zero for high H_{dc} [Figs. 1(e) and 1(f)]. Generally, Tc increases slightly with the increasing dc field in a ferromagnetic system. However, such a large shift of peak temperature in susceptibility in the presence of H_{dc} [that is, 24 K to nearly 30 K from $H = 0$ to 50 kOe in $\chi'(T)$] is not at all typical for a ferromagnetic system. These results are in good agreement with dc magnetization, where the inverse susceptibility ($\chi^{-1}(T)$) does not superimpose for different H far above T_C [see supplementary material for detailed discussions]. In addition, the isothermal magnetization exhibits a non-linear behavior even at 30 K (see supplementary material for details). The reported results suggest the presence of magnetic correlation far above T_C and point towards complex magnetic interactions in this compound. Similar complex magnetic behavior is demonstrated in a geometrically frustrated spin-chain system $Ca_3Co_2O_6$.[17]

The temperature dependent dielectric constant is depicted in Figure 2 for 51 kHz, for two different orientations of E and H, that is, $E//H$ [Fig. 2(a)] and $E \perp H$ [Fig. 2(b)]. No frequency dependence dielectric feature is observed at low temperatures in the ordered magnetic state (see supplementary material). The value of the loss angle ($\tan\delta$) as a

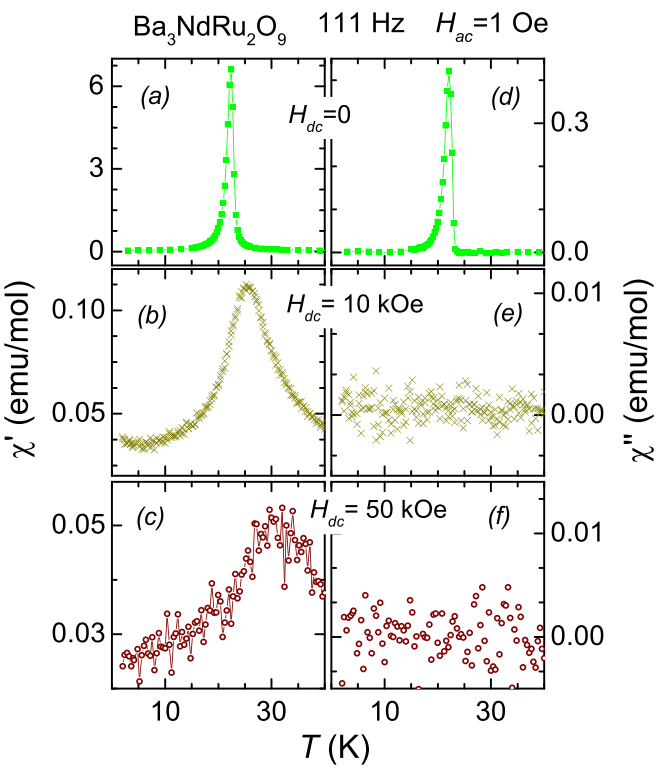

FIG. 1. Real (left panel) and imaginary (right panel) parts of ac magnetic susceptibility as a function of temperature for zero dc magnetic field (a) as well as H_{dc}= 10 kOe (b) and 50 kOe (c).

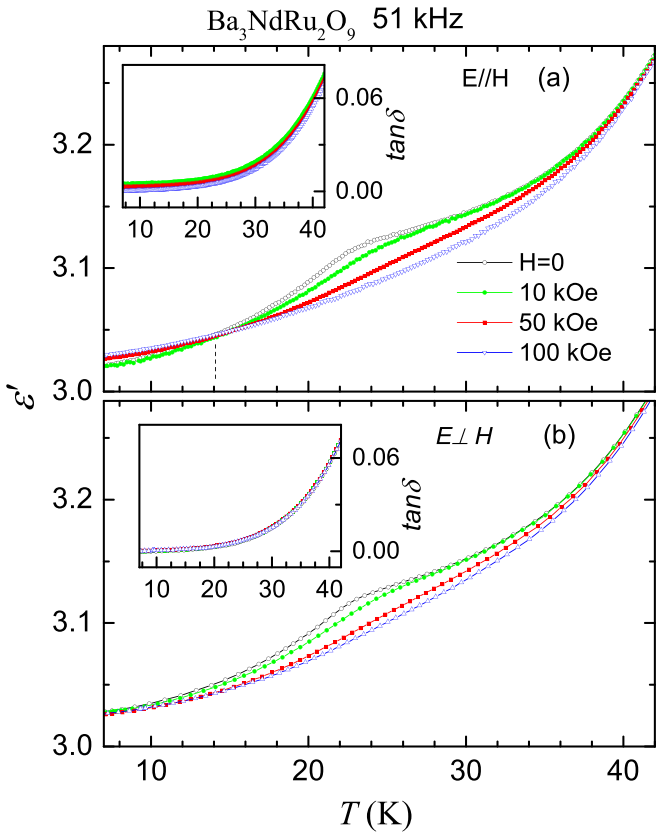

FIG. 2. Real part of the dielectric constant as a function of temperature for different magnetic fields for (a) $E//H$ and (b) $E \perp H$ orientation. Insets show the loss tangents, respectively.

measure of the dielectric loss is rather low (<0.1), as shown in the inset of Figs. 2(a) and 2(b) confirming the insulating nature of the sample. These dielectric properties allow investigating MD coupling in the magnetic ordered state. The real part of the dielectric constant (ε') clearly exhibits a peak at the onset of T_C in the absence of a magnetic field. Interestingly, the applications of a magnetic field strongly influence the dielectric constant in the magnetic regime. The dielectric peak at the ordering temperature T_C gradually suppresses with the gradual increase in magnetic fields for both the orientations, $E//H$ [Fig. 2(a)] and $E\perp H$ [Fig. 2(b)]. For $E\perp H$, the dielectric constant decreases with decreasing temperature and its value is lower for higher magnetic fields throughout the temperatures investigated. However, for $E//H$ orientation, a cross-over of the dielectric constant for different magnetic fields (with respect to the zero magnetic field) is observed at T ~13–16 K. The cross-over point [denoted by the dashed line in Fig. 2(a)] slightly varies with different magnetic fields. The value of ε' gradually becomes reduced with the increase in H for $T > 16$ K for all H ≥ 10 kOe and the value of ε' increases with increasing H for $T < 13$ K for H > 10 kOe, as depicted in Fig. 2(a). A negligible change in the value of ε' is observed for H ≤ 10 kOe.

We have further confirmed the MD behavior through measurements of the H-dependent dielectric constant, as shown in Figs. 3(a) ($E//H$) and 3(b) ($E\perp H$) for a fixed frequency of 51 kHz. The features are very similar for other frequencies (not shown here). For $E//H$ orientation [see Fig. 3(a)], a quadratic positive MD is observed at low T (for $T \leq 12$ K); however, at $T = 15$ K, the MD initially decreases with increasing H up to 25 kOe and becomes nearly flat before it increases again with increasing H above 40 kOe. Therefore, there is a cross-over of MD sign from negative to positive with increasing H. At $T = 20$ K, negative MD is observed [Fig. 3(a)]. Therefore, two components must exist, which vary differently as a function of T and H and probably are quite complex. The positive MD component completely dominates at low T (and high H), but the negative MD component competes with increasing T and dominates at further increasing temperatures (say, 20 K). For the $E\perp H$ orientation, negative MD nearly dominates at all temperatures [Fig. 3(b)], consistent with the T-dependent results in the presence of magnetic fields. However, the positive MD term exists at low temperatures as well for $E\perp H$ [see inset of Fig. 3(b) for 8 K]. Therefore, both positive and negative components of MD are present for the orientations of E and H, but only the absolute values of MD differ for these two orientations. This signals anisotropy effects within experimental uncertainties. Anisotropic MD behavior for different orientations of E and H ($E//H$ and $E\perp H$) in the polycrystalline form is rare but earlier reported in a hybrid metal-organic system.[18] Appreciable negative MD is observed above T_C for both the orientations of $E//H$ and $E\perp H$ (e.g., 30 K and 40 K) as shown in Figs. 3(a) and 3(b), which infers the existence of MD even above long-range magnetic ordering due to the presence of magnetic correlation. The small MD far above long-range ordering (even from short-range magnetic interaction) is observed in many oxide compounds exhibiting complex magnetic ground states (e.g., Refs. 17 and 19–21). However, the sample becomes less insulating with increasing temperature T ≥ 25 K [loss part tan δ increases with T above 25 K, inset of Fig. 2]; hence, the presence of a very small leakage current above 25 K cannot be ruled out. The magnitude of small negative MD at T ≥ 25 K may be amplified further by the leakage contribution of Maxwell–Wagner-like origin,[22] even in the case that the dielectric feature is of intrinsic origin. However, the sample is highly insulating below T_C (very low tan δ value which is nearly constant throughout the T-range below T_C). The strong positive MD below 15 K and the change of the nature (i.e., sign) of MD with H and T cannot originate from leakage currents.

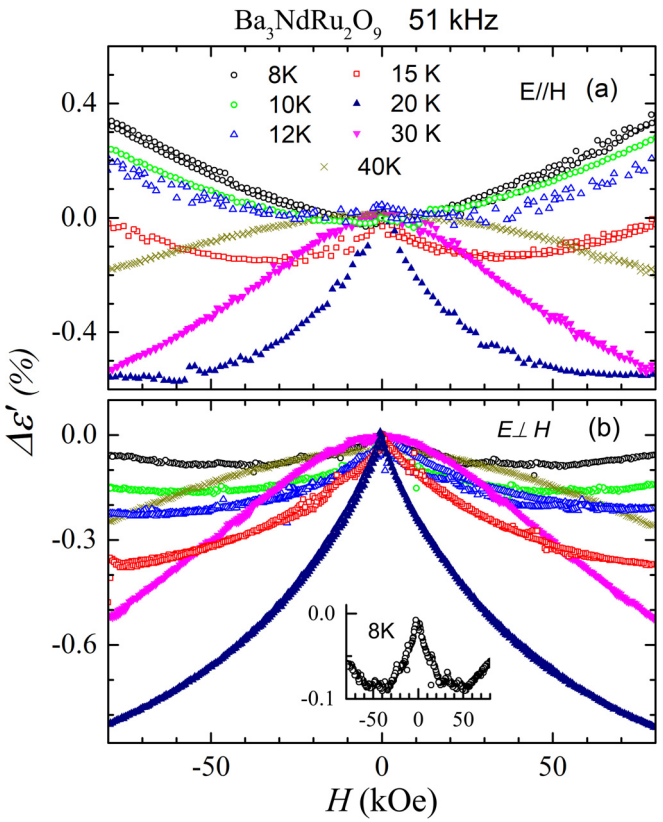

FIG. 3. Fractional change in ε' ($\Delta\varepsilon' = [\varepsilon'(H) - \varepsilon'(0)]/\varepsilon'(0)$) as a function of magnetic field for a fixed frequency of 51 kHz at selected temperatures for (a) $E//H$ and (b) $E\perp H$. The inset of (b) shows $\Delta\varepsilon'$ (H) at 8 K for $E\perp H$.

Systems containing dimers often exhibit magnetoelastic coupling, which is reported in this series as well.[12] Therefore, the small negative MD component may arise via magnetoelastic coupling. The presence of MD (absolute value could be amplified by leakage current) above T_c is also consistent with this conclusion. Strong positive MD shows up below ~T_2. The canted antiferromagnetic ordering of Nd moments and AFM ordering of ferromagnetic dimer are established below T_2.[11] Often, the Dzyaloshinskii-Moriya (D-M) interaction establishes in weak ferromagnetic or canted AFM systems, which may govern MD coupling. The magnetic frustration could be minimized via lattice distortion to a stabilized ground state at low temperature and/or high magnetic fields, which in turn may favor positive MD coupling. Hence, magnetic fields have a pronounced effect on the magnetic ordering as well as the MD coupling in the present sample. Neutron diffraction investigation by Senn *et al.*[11] demonstrates the thermal evolution of the magnetic moment of this compound, revealing that Nd-moment (z-component m_z) starts to order below 25 K (along

"*c*"-direction) and the moment (m_z) sharply increases with decreasing temperature nearly saturating below 15 K. In contrast, the other two magnetic components (m_x and m_y) in the "ab"-plane from both Nd and Ru moments emerge below 18 K and slowly saturate below the same temperature of about 15 K. Interestingly, the MD cross-over (i.e., emergence/dominance of the positive MD component) as a result of competing positive and negative MD occurs nearly at the same temperature (~15 K). The feature and change in the value of isothermal MD are nearly the same below 15 K [see Fig. 3(a) for T = 8, 10, and 12 K], where the magnetic ground state is fully stabilized and moments are almost saturated. The clear lattice anomalies at the onset of two magnetic transitions at T_C and T_2 are documented in Ref. 11, which supports MD anomaly around these two temperatures. A change in the O-Nd-O bond angle and distortion of NdO_6 octahedron is documented below ~18 K.[11] A sharp change is observed in lattice parameters ("a," "b," and "c") and for the O-Nd-O angle below 25 K, followed by a distinct decrease/increase in the value of these parameters with decreasing temperature. Below 15 K, they become nearly constant.[11] The variation of magnetic ordering in the system $Ba_3MRu_2O_9$ (for *M*= Y, Nd, and La) is also attributed to the lattice effect. Therefore, the lattice anomaly and magnetic interactions are correlated, i.e., the change in magnetism yields MD coupling via large spin-orbit coupling. However, the change in the lattice parameter and magnetic interaction turns out to be more complex in applied external magnetic fields *H*. This is documented through the *H*-dependent dielectric analyses for different orientations of *E* and *H*. Note that the system behaves like a soft ferromagnet, for which large hysteresis loop and strong MD effects can be observed at low temperatures, both could be related to magnetic domain wall motion as well.

The MD is quite complex as a function of *T*, *E*, and *H*. Probably, two or more different mechanisms are controlling the coupling, which are discussed in this manuscript. The negative MD could arise from magnetoelastic coupling below and above magnetic ordering, whereas strong positive MD coupling may arise from the D-M interaction below T_2 and dominates over negative MD coupling. The well-known multiferroic system RMn_2O_5 exhibits complex MD coupling, where both the D-M interaction and magnetostriction are responsible;[23] eventually, possible magnetic-field-induced ferroelectricity far above magnetic ordering is recently reported through another mechanism.[24] The complex MD behavior is observed in some other rare-earth based compounds as well, where dielectric behavior is quite intriguing in the presence of *H*.[25] Our bulk investigation on this interesting system warrants more detailed microscopic and theoretical studies to explore the exact mechanism of MD coupling, especially in external magnetic fields.

In summary, we have evaluated detailed magnetic and magnetodielectric coupling properties of a 6H-perovskite system containing Ru (4*d*-orbital), which is a good insulator at low temperature due to the presence of the rare-earth ion. Magnetodielectric coupling is demonstrated in this Ru-based system. Interestingly, there is a cross-over of magnetodielectric coupling below ~T_2, which is more profound for a particular orientation of *E*//*H*. Magnetic correlations and weak magnetodielectric coupling are present even above T_C. The positive and negative magnetodielectric coupling is governed via two different mechanisms which are competing as a function of *T*, *E*, and *H*. Although the magnetodielectric coupling is rather weak, our primary results yield a path designing insulating systems which contain both rare-earth and higher *d*-orbitals to achieve strong magnetodielectric coupling.

See supplementary material for detailed DC magnetization in the presence of different magnetic fields. The temperature dependent ac magnetic susceptibility and dielectric constant for different frequencies are plotted.

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