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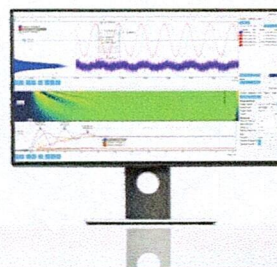
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Considerably Strong Magnetodielectric Coupling in Zircon-type DyVO₄

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Abstract. We investigate the magnetic and dielectric properties of zircon-type DyVO₄. The evidence of dielectric anomaly at the Jahn-Teller (JT) transition temperature (T_D) in DyVO₄ points a direct consequence of the onset of an electric polarization. We also report that the JT domain control gives rise to a remarkable magnetodielectric (MD) coupling, which is probably maximum in the RVO₄ series. MD coupling scales linearly with the squared magnetization as described by the Ginzburg-Landau theory.

INTRODUCTION

Recently, the RVO₄ (R = rare-earth) compounds have received renewed attention for their unusual magnetic properties [1, 2]. These materials usually crystallize in the zircon-type structure with space group $I4_1/amd$ for smaller ionic radius of R ion [3]. The crystal structure of this phase consists of VO₄ tetrahedra and RO₈ bisdisphenoid polyhedra. RO₈ units connect with one another along the a and b axes as depicted in Fig. 1. Along the c -axis, RO₈ alternately align with VO₄ units, as a result, the VO₄ units are spatially isolated by RO₈ polyhedra. The magnetic properties of RVO₄ are mainly determined by the 4f spin of R ion. Recent studies showed that some of the materials of RVO₄ family have a very high magnetocaloric effect in low temperature, which is technologically promising for cryogenic magnetic refrigeration [4, 5].

DyVO₄ is also a promising material with significant magnetic [5] and optical properties [6]. The zircon-type DyVO₄ is a typical example of showing the Jahn-Teller (JT) effect induced by the quadrupolar interaction. It undergoes the JT transition from the $I4_1/amd$ tetragonal to an orthorhombic structure at T_D (~15 K), which is associated with the ordering of 4f quadrupole moments of the Dy³⁺ ions [7]. DyVO₄ also shows high magnetocaloric effect near the antiferromagnetic (AFM) ordering of Dy³⁺ moments and the structural transition associated with 4f ferroquadrupolar ordering of Dy³⁺ ion [5]. The structure analysis showed that quadrupolar distortion in DyVO₄ also induces an antiferroelectric lattice distortion, and makes it promising for investigation of the multiferroic properties in details [7].

In this paper, we report magnetic and dielectric properties in zircon-type DyVO₄. We observe a strong magnetodielectric (MD) effect around the JT transition temperature of DyVO₄. MD coupling scales linearly to the squared magnetization indicating the presence of spontaneous electric polarization around T_D in DyVO₄.

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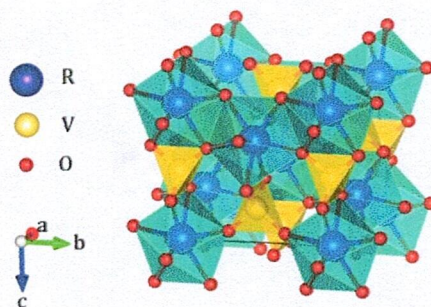


FIGURE 1. Zircon-type crystal structure of RVO_4 at room temperature.

EXPERIMENTAL DETAILS

The polycrystalline $DyVO_4$ is prepared using a solid-state reaction [4]. The single-phase chemical composition is confirmed by the x-ray diffraction studies at room temperature recorded in a Bruker D8 Advance powder diffractometer using the $Cu K_\alpha$ radiation. The powder sample pressed into a pellet is used for the dielectric measurements using a E4980A LCR meter (Agilent Technologies, USA) equipped with a commercial PPMS evercool-II system of Quantum Design. The electrical contacts are fabricated using an air-drying silver paint. Magnetization is measured in a commercial magnetometer of Quantum Design (MPMS, evercool).

EXPERIMENTAL RESULTS AND DISCUSSIONS

The Rietveld refinement of the x-ray diffraction data of $DyVO_4$ at room temperature are shown in Fig. 2. The refinement is done using tetragonal structure with $I4_1/amd$ space group with atomic positions Dy (0,0,0), Cr (0,0,0.5), and O [0,0, 0.1781(2), 0.3375(6)]. The reliability parameters $R_w(\%) \sim 3.92$, $R_{exp}(\%) \sim 3.71$, and $\chi^2 \sim 1.38$ are reasonably small. The refined lattice parameter of the unit cell are $a = 7.1461(2)$ and $c = 6.3071(8)$ Å, which are close to the previously reported values [8]. The difference plot shown at the bottom confirms the single phase without trace amount of impurity.

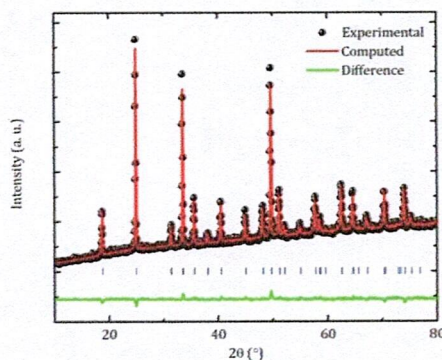


FIGURE 2. Rietveld refinement of x-ray powder diffraction patterns of $DyVO_4$ at room temperature.

The thermal variations of the zero-field cooled (ZFC) and field-cooled (FC) magnetization curves recorded at 100 Oe are depicted in Fig. 3(a). Both curves show a paramagnetic (PM) to AFM transition below $T_N \sim 3.5$ K due to the long-range ordering of the Dy^{3+} moments. No significant difference has been observed between ZFC and FC cycles. Analogous the previous reports, no signature of JT transition around 15 K is observed in the $M(T)$ curve [5]. Figure 3(b) shows the magnetization curve $[M(H)]$ at selected temperatures above and below T_N . The $M(H)$ at 2 K increases rapidly above a critical field $B_C \sim 2.5$ kOe due to the metamagnetic transition from AFM to ferromagnetic (FM) state. The $M(H)$ curve shows a saturating trend above 40 kOe indicating the FM nature above B_C . The nonlinear nature of



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$M(H)$ is observed at 12 K, which indicates the existence of short-range ordering due to the reported JT transition associated with 4f ferroquadrupolar ordering of Dy^{3+} ion. With further increasing the temperature the $M(H)$ curve becomes linear in the PM state.

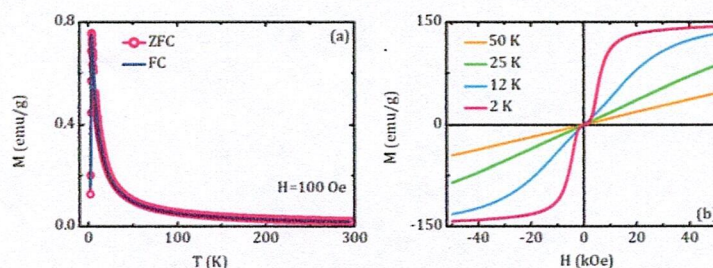


FIGURE 3. (a) T variation of ZFC-FC magnetization curves at 100 Oe. (b) Magnetization curves at selected temperatures.

The dielectric permittivity (ϵ) is recorded at different frequencies (f) by varying T for $DyVO_4$. Figure 4(a) depicts thermal variation of the real component (ϵ') of ϵ at different f . Inset of Fig. 4(a) clearly shows a Curie-type temperature dependence above T_D (~ 15 K) and a sudden drop below T_D , which is consistent with the results reported previously [9, 10]. The peak around T_D does not show any frequency dispersion. The dielectric anomaly observed at the JT transition temperature in $DyVO_4$ was understood as a direct consequence of the onset of an antiferroelectric ordering which is driven by the softening B_{1g} strain mode, i.e., as an optical-mode condensation without its softening [9]. No signature around T_N is observed in $\epsilon'(T)$.

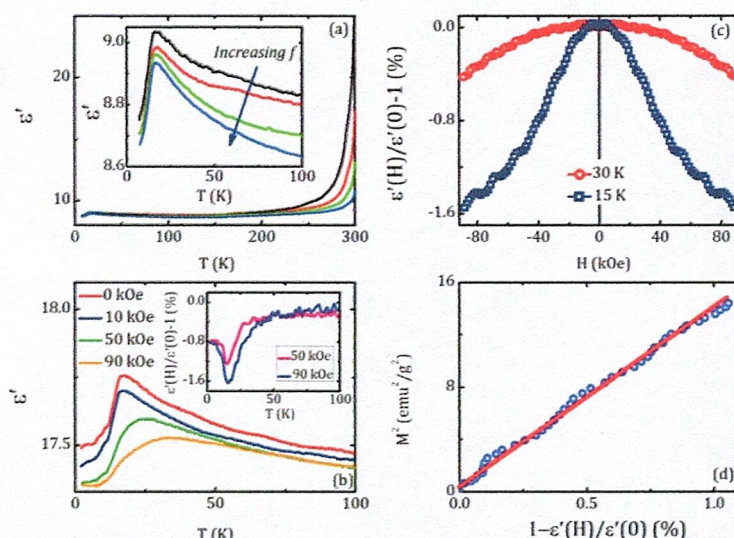


FIGURE 4. (a) T variation of ϵ' at different f . (b) T variation of ϵ' at different magnetic fields at $f=2$ kHz. (c) The H variation of $[\epsilon'(H)/\epsilon'(0)-1]$ at selected T . (d) The M^2 vs $-[[\epsilon'(H)/\epsilon'(0)-1](\%)]$ plots at 15 K up to 50 kOe magnetic field. Inset of (a) shows the $\epsilon'(T)$ in low temperature region and (b) shows the variation of $\epsilon'(H)/\epsilon'(0)-1$ with T at $H=50$ and 90 kOe.

The magnetization curve below T_D indicates the existence of short-range magnetic ordering above T_N . Figure 4(b) depicts the thermal variation of ϵ' at different applied magnetic fields. With increasing field the peak at T_D depressed rapidly. The magnetodielectric (MD) response, defined as $\epsilon'(H)/\epsilon'(0)-1$, is measured with T , as depicted in the inset of Fig. 4(b) at $H=50$ and 90 kOe. The MD% value shows maximum around T_D for both the magnetic fields and we get 1.6% MD at T_D for 90 kOe applied field. This high value of MD is probably maximum in the RVO_4 series of compounds. The MD effect is further investigated with H above and below T_D . The MD effects as a function of magnetic field are shown in Fig. 4(c) at $T=15$ and 30 K. The value of MD increases with H up to $\sim 1.6\%$ at 90 kOe at 15 K. The MD effect relates the ME coupling, which can be phenomenologically expressed using the Ginzburg-

Landau theory through the ME coupling term $\gamma P^2 M^2$ in the thermodynamic potential (Φ) defined as,

$$\Phi = \Phi_0 + \alpha P^2 + \frac{\beta}{2} P^4 - PE + \alpha' M^2 + \frac{\beta'}{2} M^4 - MH + \gamma P^2 M^2 \quad (1)$$

Where α , β , α' , β' , and γ are the constants and functions of temperature. In the magnetically ordered state, the role of magnetic order on MD in a field is followed by the linear curve of M^2 versus $[\epsilon'(H)/\epsilon'(0)-1](\%)$ in the low-field region. Here, the M^2 versus $-\epsilon'(H)/\epsilon'(0)-1](\%)$ plot at 15 K is depicted in Fig. 4(d). The result indicates that the ME coupling term $\gamma P^2 M^2$ of the Ginzburg-Landau theory [Eq. (1)] is significant up to 50 kOe magnetic field, which is similar as reported earlier for various magnetodielectric compounds [11, 12].

CONCLUSION

The above results demonstrate that the applied magnetic field rearranges the quadropolar moments of Dy^{3+} ions associated with the JT transition. As a result, the proposed antiferroelectric ordering near T_D is strongly affected by the applied magnetic field and gives a very promising MD effect. These results indicate that $DyVO_4$ may exhibit a good multiferroic nature around the JT transition temperature. Further studies in experimental and theoretical calculations are suggested for establishing a possible multiferroic order in $DyVO_4$.

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