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## Magnetic and transport properties of vanadate PrVO<sub>3</sub>

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**Abstract.** The magnetic and electric properties of the vanadate PrVO<sub>3</sub> with the GdFeO<sub>3</sub>-type orthorhombic structure are reported. The temperature-dependent magnetization displays an obvious irreversibility under zero-field cooling (ZFC) and field cooling (FC), and two magnetic phase-transition temperatures 20 K and 90 K, which may correspond to the spin reorientation and the magnetic ordering, respectively. A canted antiferromagnetic configuration is derived from the low spontaneous magnetization at 5 K. In the sintered bulk, a high coercivity 2.4 T is due to the pinning of the domain wall. The resistance measurement shows that the compound is an insulator at low temperatures.

### 1. Introduction

Transition-metal oxides with perovskite structure can have significant physical properties such as high-temperature superconductivity in cuprates, colossal magnetoresistance in manganites and very diverse magnetic properties. Recently, the perovskite vanadates RVO<sub>3</sub> (R = rare earth), especially with non-magnetic R elements La, Lu, Y, have attracted more attention because of the metal–insulator transition in the alkali-earth-doped compounds [1, 2] and the temperature-induced magnetization reversal in polycrystalline LaVO<sub>3</sub> [3, 4] and in the single-crystal YVO<sub>3</sub> [5]. All RVO<sub>3</sub> compounds crystallize in orthorhombic symmetry (space group: *Pbnm*) resulting from a cooperative distortion of the VO<sub>6</sub> octahedra along the [−110] direction of the ideal cubic unit cell [6]. Low-temperature neutron- and x-ray-diffraction investigations have verified the first-order character of the structural phase transition and the changes between the different types of magnetic structure while the detailed magnetic structure is still controversial [5, 7, 8]. The temperature-induced magnetization reversal has been attributed to a reversal of the ferromagnetic component of a canted-spin antiferromagnet on cooling through a first-order phase transition at  $T_i < T_N$  [4, 5]. However, few works [8, 9] have been performed on RVO<sub>3</sub> with magnetic rare-earth elements to investigate the effect of magnetic rare-earth elements on structure and magnetic properties of the perovskite vanadates. In the present paper, we report magnetic and electric properties of polycrystalline PrVO<sub>3</sub>. The results are interpreted in terms of a canted antiferromagnetic insulator with a very high low-temperature coercivity for the sintered bulk.

### 2. Experiment

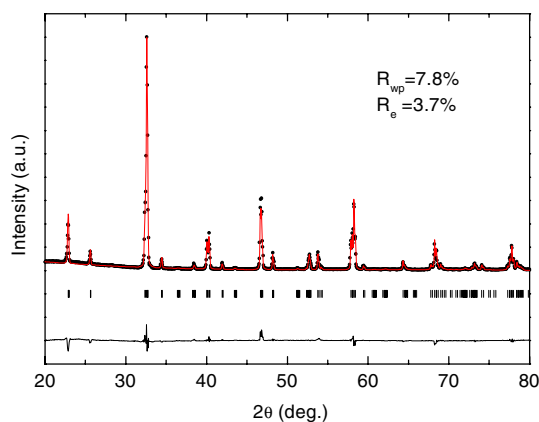
A powder sample of PrVO<sub>3</sub> was prepared by deoxidizing PrVO<sub>4</sub> powder in flowing hydrogen mixed with argon at the volume ratio of H<sub>2</sub>:Ar = 1:7 at 950 °C for 24 hours. The PrVO<sub>4</sub>

powder was obtained by standard solid-state reaction in air at 900 °C of stoichiometric mixtures of  $\text{Pr}_2\text{O}_3$  and  $\text{V}_2\text{O}_3$  powders of purity 99.99%. The single-phase perovskite  $\text{PrVO}_3$  was pressed into pellets, then sintered in flowing ultrapure argon at 1400 °C for 6 hours.

Step-scanning data of x-ray diffraction were collected on the powder and sintered  $\text{PrVO}_3$  at  $2\theta$  angles from 2° to 80° using a Rigaku rotating-anode x-ray diffractometer with  $\text{Cu K}\alpha$  radiation monochromatized by a single-crystal graphic monochromator. Magnetization as a function of temperature or applied field was recorded by a commercial superconducting quantum interference device (SQUID) magnetometer, and resistance was measured by the standard four-probe method in the SQUID.

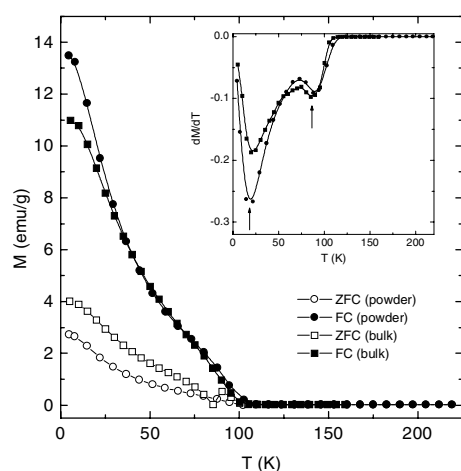
### 3. Results and discussion

Rietveld refinement of the step-scanning data (figure 1) via the program Fullprof [10] shows that the as-cast and the sintered samples of  $\text{PrVO}_3$  crystallize in the orthorhombic  $Pbnm$  structure, isostructural with  $\text{GdFeO}_3$ . The lattice constants  $a = 5.4747 \pm 0.0005 \text{ \AA}$ ,  $b = 5.5255 \pm 0.0004 \text{ \AA}$  and  $c = 7.7550 \pm 0.0006 \text{ \AA}$ , in agreement with the result from JCPDS (card No 25-590). No other phase could be detected in the specimens.

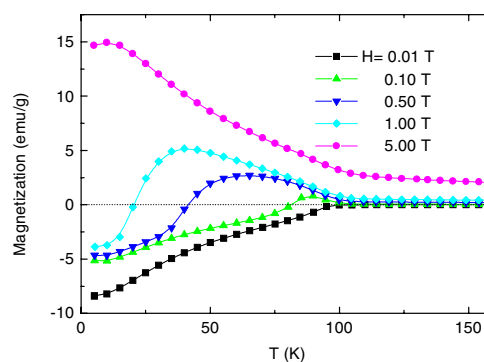


**Figure 1.** The measured (dot) and calculated (line) patterns of the powder x-ray diffraction of  $\text{PrVO}_3$ . The calculated positions are indicated at the bottom. The differences between the observed data and the calculated values are also given.  $R_{wp}$  is the weight pattern factor, and  $R_e$  the expectation factor.

Figure 2 shows the temperature dependence of magnetization of the powder and the sintered bulk  $\text{PrVO}_3$  as measured on warming in 0.05 T, after being cooled under zero field (ZFC) and 0.05 T field cooled (FC) from room temperature. An apparent irreversibility and a large ferromagnetic component at low temperature can be observed. Two different magnetic-ordering temperatures 90 K and 20 K can be derived from the differentiation of the  $M-T$  curves (the inset in figure 2). The former corresponds to the Néel temperature although it is almost 50 K lower than that in [9], and the latter is attributed to the spin reorientation detected by neutron diffraction [8]. The temperature dependence of the susceptibility  $\chi(T)$  in the paramagnetic state can well be described by the Curie–Weiss law  $\chi(T) = C/(T - \Theta_P)$ , where  $C \sim \mu_p^2$  is the Curie constant and  $\mu_p$  is the total paramagnetic moment of the magnetic ions per formula unit. The fitted value  $\mu_p = 5.6 \pm 0.2 \mu_B$  is very close to the sum of the theoretical moments of  $\text{Pr}^{3+}$  ( $3.58 \mu_B$ ) and  $\text{V}^{3+}$  ( $2.83 \mu_B$ ), and  $\Theta_P = 86 \text{ K}$  in agreement with the magnetic phase transition.



**Figure 2.** Temperature dependence of magnetization of the powder (circle) and the sintered bulk (square)  $\text{PrVO}_3$  in a field of 0.05 T, as measured on warming after samples are ZFC (open) and FC (solid). Inset is the differentiation of the temperature-dependent magnetization under FC.

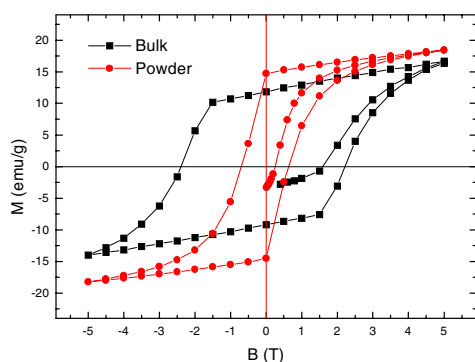


**Figure 3.** Temperature dependence of magnetization of the sintered bulk  $\text{PrVO}_3$  with ZFC in the case of frozen moment due to the remaining field of the SQUID magnetometer.

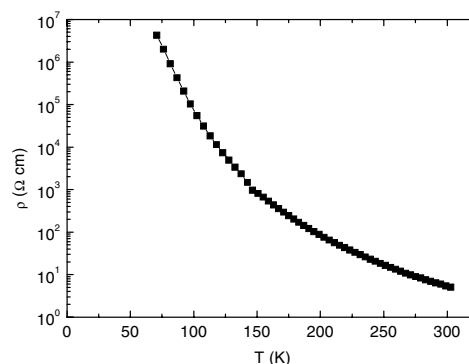
It is noted that an ‘anomalous’ diamagnetism is easily detected in the case of ZFC just like that in  $\text{CeVO}_3$  reported by Nguyen and Goodenough [11]. As seen in figure 3, the magnetization is negative before the paramagnetic state is reached in the field of 0.01 T, but always positive in the field of 5.0 T. The sign of the magnetization changes from negative to positive at a temperature decreasing with the applied field. This behaviour would result from the frozen moment owing to a remaining field, even though very small, antiparallel to the direction of the normal positive field and to the high coercivity at low temperatures. Therefore the applied field will be equal to the coercivity at the temperatures where the magnetization changes its sign. Namely, the coercivity reaches 0.1 T at 80 K, 0.5 T at 40 K and 1.0 T at 20 K, respectively. Derived from the hysteresis measurements at corresponding temperatures, the values of the coercivity, excepting about 10% increase due to a relatively high field of 6 T applied in order to magnetize the sample, are in agreement with the former ones.

The curves of initial magnetization and hysteresis of the powder and the sintered bulk of  $\text{PrVO}_3$  up to 6.0 T at 5 K are plotted in figure 4. The intrinsic coercivity is 2.4 T for the sintered bulk while it is only 0.7 T for the powder. The negative magnetization in the initial part of the magnetization curve is due to the frozen moment in the remaining field of the SQUID. From the magnetization curves, one can conclude that the mechanism of the high coercivity is the domain-wall pinning. Mostly, the high coercivity at low temperatures, detected in the rare-earth-transition-metal intermetallic compounds, results from the pinning of the narrow domain wall. But the great difference of the coercivity between the powder and the bulk of  $\text{PrVO}_3$  may imply that the coercivity is not determined by the narrow-domain-wall pinning. From the magnetization curve of the free powder, the spontaneous magnetization is estimated to be  $15 \text{ emu g}^{-1}$ , i.e.  $0.65 \mu_B$  per formula unit, far lower than the ionic theoretical moment, which indicates a canted antiferromagnetic configuration not only for the V sublattice [3–5, 7, 8] but also for the Pr sublattice.

Figure 5 shows the temperature dependence of the resistance of  $\text{PrVO}_3$  measured by the standard four-probe method. The resistance below the temperature of 70 K is out of the range



**Figure 4.** The curves of magnetization and hysteresis of the powder and the sintered bulk  $\text{PrVO}_3$  at 5 K.



**Figure 5.** Temperature dependence of resistivity of the sintered bulk  $\text{PrVO}_3$ .

which our equipment can detect. It can be found that the compound displays the behaviour of an insulator with  $\rho = 4.8 \Omega \text{ cm}$  at room temperature which is satisfactorily consistent with the result reported by Onoda and Napasawa [6]. Neither a magnetic nor structural phase transition is demonstrated in the temperature-dependent resistance. The external field has no effect on the resistivity up to 6 T.

In summary, the vanadate  $\text{PrVO}_3$  crystallizes in the  $\text{GdFeO}_3$ -type orthorhombic structure with  $a = 5.4747 \text{ \AA}$ ,  $b = 5.5255 \text{ \AA}$  and  $c = 7.7550 \text{ \AA}$ . The temperature-dependent magnetization shows an obvious irreversibility. Two magnetic phase changes are observed at 20 K and 90 K, which may be representative for the spin reorientation and the magnetic ordering, respectively. A canted antiferromagnetic spin configuration gives rise to a low spontaneous magnetization. In the sintered bulk, the coercivity due to the pinning of the domain walls (these may not be narrow domain walls) reaches 2.4 T. The temperature-dependent resistance shows the compound is an insulator.

### Acknowledgments

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