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$PrVO_3$: An inhomogeneous antiferromagnetic material with random fields

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We have studied the zero-field-cooled, field-cooled, magnetization and the hysteresis loops of a $PrVO_3$ single crystal. The compound has shown some unusual features: (i) below the antiferromagnetic spin ordering (SO) temperature $T_{SO}=132$ K, we observed another transition with a glassylike behavior; (ii) the hysteresis loops of the compound exhibit staircaselike behavior at a temperature below 3 K; (iii) although the high negative Weiss temperatures indicate that the compound is a (very) strong antiferromagnet, we observe relatively high values of the remanent magnetization (M_r) along all of the axes. Based on the results obtained, we argue that $PrVO_3$ should be considered as a disorder (or inhomogeneous) antiferromagnet with random fields. We discuss the possible sources of disorder in the compound in terms of orbital quantum fluctuations.

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I. INTRODUCTION

The transition metal (TM) oxides exhibit a rich variety of physical properties, many aspects of which continue to puzzle the research community. Generally speaking, in these systems the orbital degrees of freedom are a key issue because the orbital occupancy can affect the interaction between the spins. Coupling to the lattice further enriches the interplay between spin and orbital effects through lattice distortions, phonons, and cooperative effects such as Jahn-Teller (JT) distortions.[1] Cuprate superconductors and manganites with colossal magnetoresistance belong to the TM oxides with a $3d\ e_g$ bands at the Fermi level. In these systems the JT interaction is relatively strong and the orbital degeneracy is lifted well above the magnetic ordering temperature. The situation is different for the vanadates with the general formula RVO_3 (R=rare earth or Y) where the $3d\ t_{2g}$ bands are at the Fermi level but the JT interaction is much weaker. As a result, the intrinsic frustration between spin and orbital degrees of freedoms is believed to be crucial for understanding the interplay between the different ordering mechanisms in these compounds and their highly unusual magnetic properties.[2–4]

Early neutron powder diffraction studies on RVO_3 suggested that these materials order with one of two different types of magnetic structures.[5] For the compounds with large rare earth radii (La–Dy), the magnetic structure is of a C type, i.e., the spins order antiferromagnetically in the ab plane and ferromagnetically along the c axis, whereas compounds with small rare earth ionic radii (Ho–Lu) have a G-type magnetic structure, i.e., the spins order antiferromagnetically along all the directions. According to the Goodenough-Kanamori rules,[6] the C-type and G-type spin configuration will lead to G-type and C-type orbital ordering (OO), respectively. More recent neutron diffraction studies on YVO_3 (see Ref. 3) have indicated that the spins in the intermediate temperature range with C-type structure are canted by an angle $\theta=16.5(1.8)^\circ$ out of the ab plane. On the basis of magnetization data collected on polycrystalline material, a canted antiferromagnetic scheme was also speculated for $PrVO_3$.[7] Our preliminary recent single crystal neutron diffraction studies on $PrVO_3$ have indicated that both ferromagnetic and antiferromagnetic interactions are present within the system, although the exact nature of the exchange pathways remains unclear at this time.[8]

In this paper, we study in detail the magnetic properties of a $PrVO_3$ single crystal. The compound has shown an OO transition at $T_{OO}=155$ K and then followed by the antiferromagnetic spin ordering (SO) transition at $T_{SO}=132$ K. In the ordering region, there is an additional transition with the glassylike behavior. In addition, we have observed a staircaselike behavior of the hysteresis loops at low temperature, a feature which is usually expected only for disorder systems.

II. EXPERIMENT

Single crystal $PrVO_3$ was grown by means of the floating zone technique using a high temperature xenon arc furnace. At first, $PrVO_4$ was prepared by mixing the nominal composition of Pr_6O_{11}, and V_2O_5 (with purity of 99.9%) followed by annealing at 1100 °C in 48 h. The product is then reduced at 1000 °C in H_2 flow in 10 h to create $PrVO_3$ powder phase. The $PrVO_3$ feed and seed used for single crystal growth were made by pressing the powder under hydraulic pressure and then annealed at 1500 °C under a flow of Ar.

The zero-field-cooled (ZFC), field-cooled (FC) measurements of the magnetization were carried out using a commercial Quantum Design superconducting quantum interference device (SQUID) magnetometer in the temperature range 2–300 K. For the field dependence of the magnetization, we used an Oxford Instruments vibrating sample magnetometer (VSM), in which the magnetic field sweep rate can be easily controlled. The heat capacity measurements were performed in the temperature range from 0.45 to 300 K using a Quantum Design physical property measurement system (PPMS).

III. RESULTS

In Fig. 1, we plot the heat capacity (C_p) as a function of temperature. Two peaks at 128 and 155 K correspond to the SO and OO transitions, respectively. These peaks are weakly

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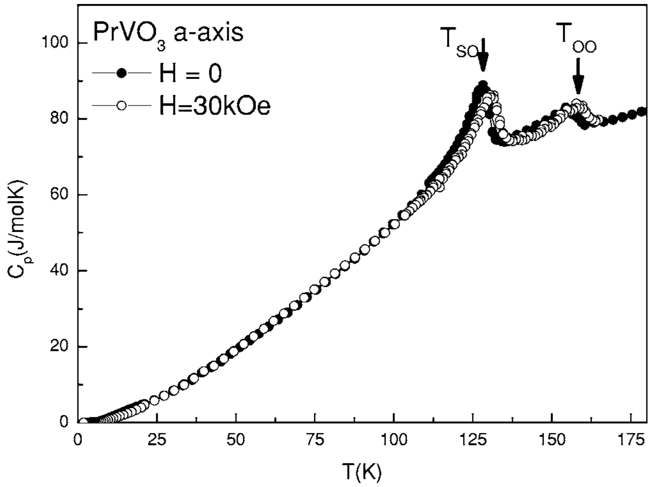

FIG. 1. Heat capacity C_p as a function of temperature of PrVO$_3$ crystal. The SO and OO temperatures are indicated by the arrows.

field dependent; in an applied field of 30 kOe the peaks are slightly shifted to higher temperature.

The results of the ZFC, FC magnetization versus temperatures sweeps collected in applied fields of 0.1 and 0.4 kOe along different crystallographic axes are presented in Fig. 2.[9] In this figure, we have divided the FC data by a factor of 10 to better visualize the ZFC curve. In the figure, we can see that all the ZFC curves exhibit a diamagnetic response at low temperatures. Our careful investigation indicates that this feature is simply due to the fact that the sample is very sensitive to the presence of a small (negative) field during cooling. This negative field either comes from the trapped field of the superconducting magnet of the SQUID magnetometer or the earth magnetic field at our location. We will report latter this issue elsewhere. We observe that at (low) applied field of 0.1 kOe, the ZFC curve has a single peak at 128 K which is at the same position of the SO transition peak observed in the heat capacity of the compound. The splitting of the ZFC and FC, however, (noticeably) starts at 132 K. We therefore take 132 K to be the value of T_{SO} in this compound. At applied field of 0.4 kOe, the ZFC curve starts to show an additional broad peak at lower temperature. The position of this broad peak shifts towards lower temperature with increasing applied fields (see Fig. 3). The FC magnetization, on the other hand, changes the slope at T_{SO} and then it increases rather smoothly with decreasing temperature. Interestingly, we see that at small applied fields ($H \leqslant 1$ kOe), the splitting between the ZFC and FC curves occurs right at T_{SO}, but it moves towards the low temperature broad peak with increasing applied field. The latter behavior of the ZFC, FC is very similar with that of spin glass or nano magnetic particle systems.

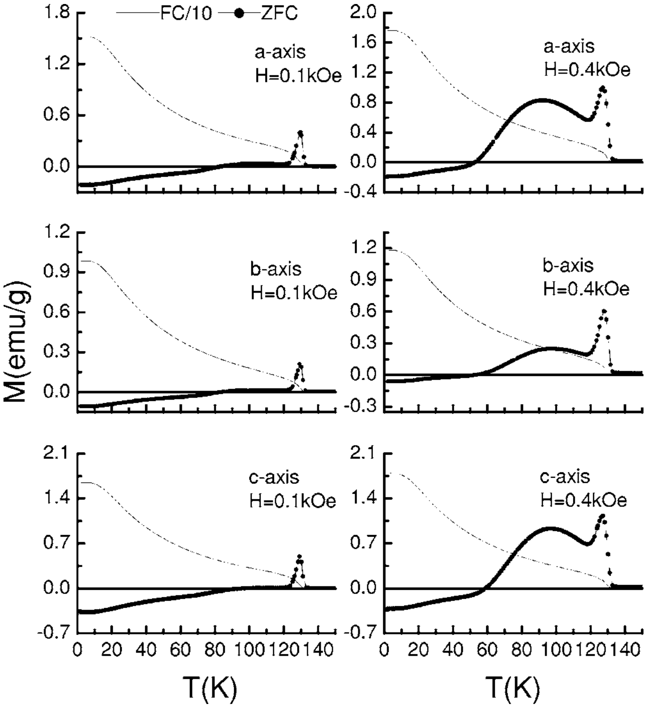

FIG. 2. ZFC, FC magnetization of PrVO$_3$ crystal along the main axes at applied fields of 0.1 kOe (left) and 0.4 kOe (right). The data for the FC are divided by a factor of 10 to better visualize the ZFC curve.

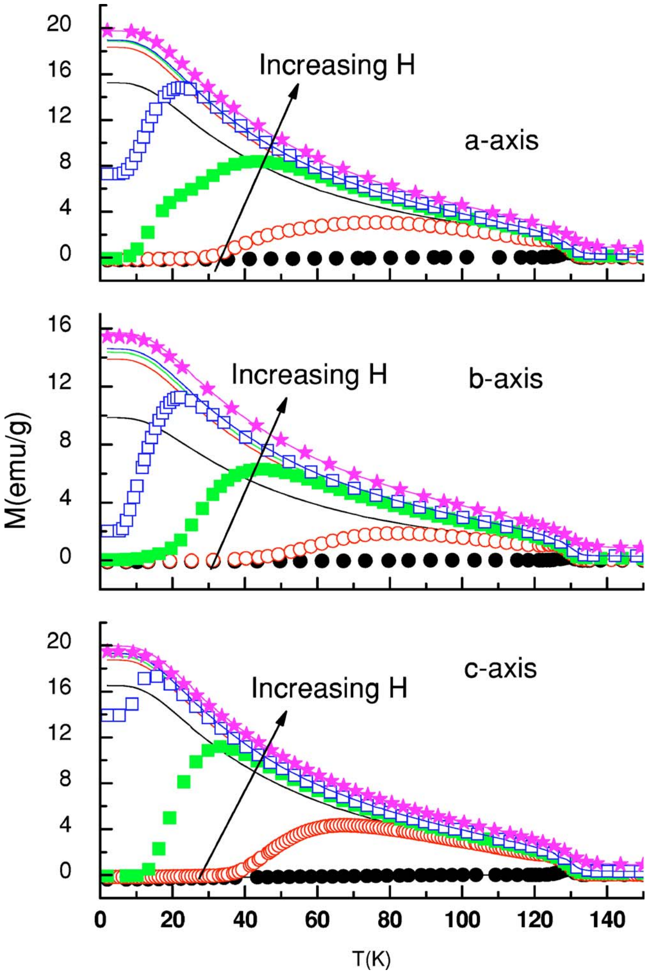

FIG. 3. (Color online) ZFC (symbols) and FC (lines) magnetization of PrVO$_3$ crystal measured at different applied fields of 0.1 kOe (black), 1 kOe (red), 3 kOe (green), 7 kOe (blue), and 20 kOe (magenta) along the main axes.

Well above T_{SO} (Fig. 4), the susceptibility follows the Curie-Weiss behavior $\chi = C/(T-\theta_P)$ (where C is the Curie

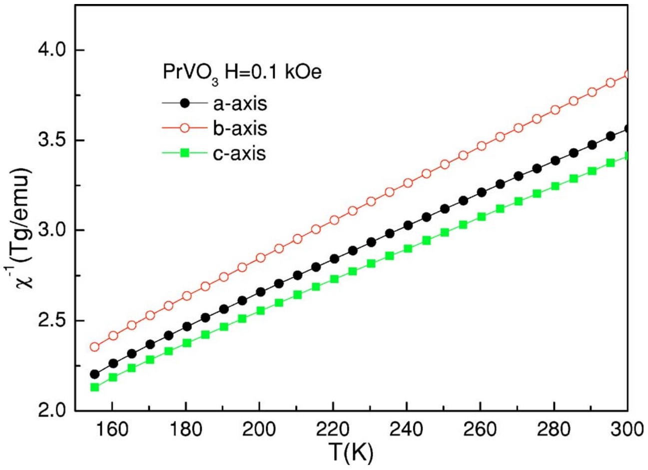

FIG. 4. (Color online) Temperature dependence of the inverse susceptibility in the paramagnetic region of PrVO$_3$ crystal.

constant and θ_P is the Weiss temperature). The change in slope along different axes seen in the $\chi^{-1}(T)$ in the paramagnetic region arises from the influence of the crystal fields on the Pr^{3+} and V^{3+} ions. Fitting using the Curie-Weiss expression gives the effective moments $\mu_b^{\mathrm{eff}}=4.3\ \mu_B$/f.u., $\mu_a^{\mathrm{eff}}=4.55\ \mu_B$/f.u., $\mu_c^{\mathrm{eff}}=4.68\ \mu_B$/f.u., and Weiss temperatures $\theta_P^b=-74.3$ K, $\theta_P^a=-85.6$ K, $\theta_P^c=-91.2$ K, along the *b*, *a*, and *c* axes, respectively. Since, in the paramagnetic region, the system consists of the two different non-interacting spins V^{3+} and Pr^{3+}, the effective moments can be estimated through the relationship $\mu_{\mathrm{eff}}=\sqrt{\mu_{\mathrm{eff}}^2(\mathrm{V}^{3+})+\mu_{\mathrm{eff}}^2(\mathrm{Pr}^{3+})}$.[10,11] Assuming that the spins of the V^{3+} and Pr^{3+} are in the ground state with $\mu_{\mathrm{eff}}(\mathrm{V}^{3+})=2.83\ \mu_B$ (spin only, $S=1$) and $\mu_{\mathrm{eff}}(\mathrm{Pr}^{3+})=3.58\ \mu_B$, an effective moment $\mu_{\mathrm{eff}}(\mathrm{PrVO_3})=4.56\ \mu_B$ is obtained, which is very close to the observed experimental value.

The hysteresis loops at 1.5 K for fields applied along different crystallographic axes are displayed in Fig. 5. It can be seen that for applied fields above 18 kOe, the magnetization is only weakly field dependent along all three directions measured. This remains true up to an applied field of 100 kOe (data not shown). The remanent magnetization corresponds to $M_r^b=0.58\ \mu_B$/f.u., $M_r^a=0.84\ \mu_B$/f.u., $M_r^c=0.82\ \mu_B$/f.u., along the *b*, *a*, and *c* axes, respectively. Remarkably, along all three axes, we observe a staircaselike behavior in the *M* vs *H* loops up to a temperature of 3 K (see inset of Fig. 5). Some of the transitions occur for field variations as small as 1 Oe. We have investigated in detail whether there are any training effects in the magnetic response, as well as the field sweep rate dependence of the loops. Several loops were collected under the same experimental conditions (i.e., cooling protocol, temperature, field sweep rate). The results are presented in the inset of Fig. 6 at temperature of 1.5 K. There do not appear to be any training effects present in these data. However, it is clear that features seen in the data along the *b* and *c* axes still depend to some extent on the field sweep rate. In Fig. 6, we can see that as the field is reduced after "*saturation*," the position of the first step in the data shift towards higher absolute values of magnetic field as the field sweep rate is reduced.

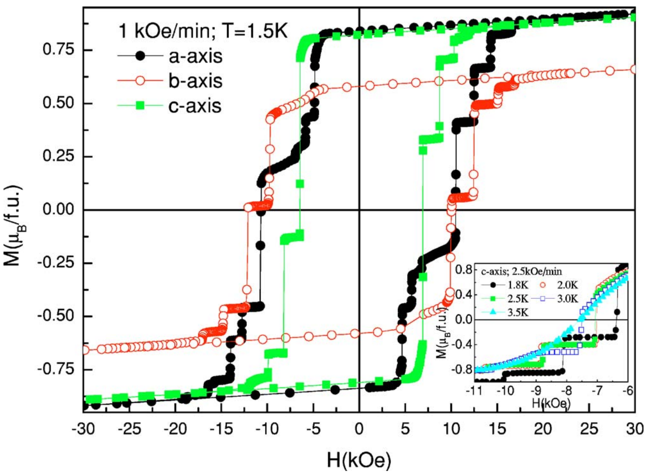

FIG. 5. (Color online) Hysteresis loops at 1.5 K along the main axes of PrVO$_3$ crystal. The inset shows the hysteresis loops at different temperatures along the *c* axis.

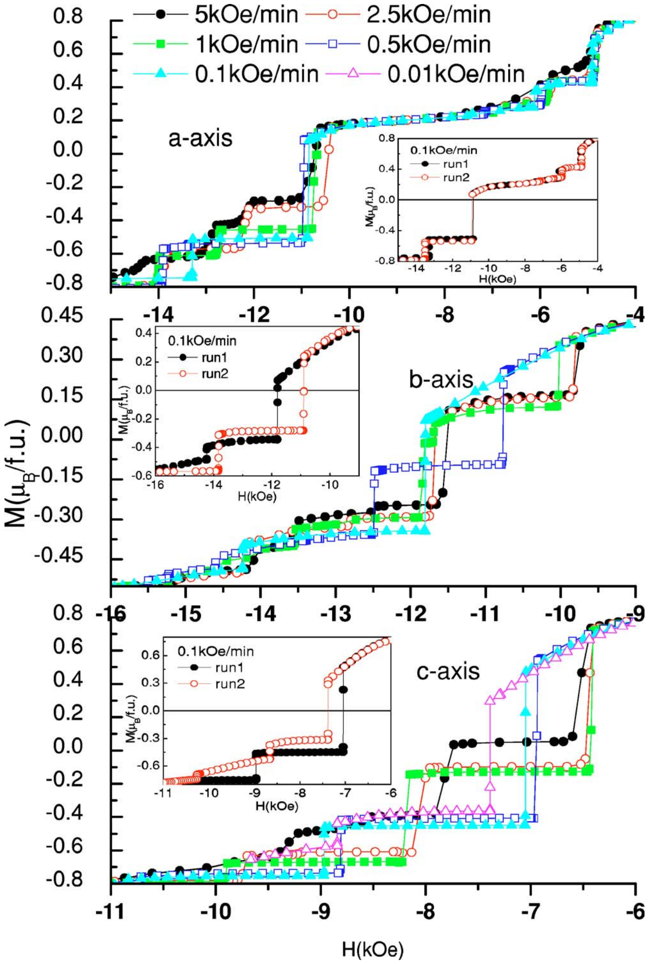

FIG. 6. (Color online) The decreasing field sections of hysteresis loops at 1.5 K along the main axes of PrVO$_3$ crystal with different field sweep rates as indicated. The inset shows the no training effect.

IV. DISCUSSIONS

In $PrVO_3$, one might expect that the magnetic contribution is due to both the Pr^{3+} and the V^{3+} ions. However, due to the low symmetry of the compound, i.e., orthorhombic at room temperature and supposedly monoclinic below T_{SO}, the J ground state multiplet splits into nine singlets for Pr^{3+} under the effect of the crystal fields.[12] Therefore, it is quite possible that in this case the Pr^{3+} will be in a nonmagnetic state, provided that the exchange interaction is not too strong. This scenario is further supported by the fact that we do not observe any additional transition associated with the ordering of Pr^{3+} below T_{SO} of 132 K either in the heat capacity or the ZFC, FC magnetization curves. It seems likely that the ordered magnetic properties of the compound are entirely due to the contribution from the V^{3+} ions.

The staircaselike behavior observed in the low temperature hysteresis loops of $PrVO_3$ is clearly related to avalanches of domain flips which are the analog of a Barkhausen jumps in real magnets (the Barkhausen effect).[13] Our preliminary studies of all the other RVO$_3$ vanadate single crystals down to a temperature of 1.5 K indicate that this feature is unique to $PrVO_3$. Furthermore, we note that while the staircaselike behavior is observed with the field applied along different crystallographic axes and in different crystals, it is completely absent in polycrystalline samples (data not shown). This fact suggests that either the grain boundaries, averaging effects along different axes or a different stoichiometric composition in polycrystalline samples influence the magnetic response of the material.

One possible mechanism of the staircaselike behavior of the hysteresis loops is based on domain propagation and growth involving the motion of small portions of domain wall that are pinned by defects. This mechanism has been invoked in the case of chemically disorder magnet $SmCo_{3.5}Cu_{1.5}$ (see Ref. 14) to account for the observed low temperature staircaselike behavior of the hysteresis loop as well as the field sweep rate dependence seen in this material.

For other disorder systems, the staircaselike behavior of the hysteresis loop is attributed to the presence of random fields (RF).[15] Under this mechanism, the flipping of domains at some external field may force the neighboring domains which are interacting with them to flip, thus leading to an avalanche of domain flips. In systems with no RF, the local fields at each crystallographic site are identical so the spins (or all the domains) will flip at the same time giving rise to a single step at the switching field.

However, the RF mechanism itself cannot explain for the observed field sweep rate dependence of the jumps in $PrVO_3$. Here, the thermal coupling of the spins to the heat bath (i.e., the magnetocaloric effect) plays an important role.[16] The shift in the position of the jumps in the hysteresis loops to higher applied magnetic field when the field sweep rate decreases simply reflects the adiabaticity and the dissipation mechanism of the spin reversal energy. When the microscopic system is in an adiabatic state, the energy generated by the spin reversal process HdM at higher field sweep rates cannot easily dissipate to the heat bath and can instigate the reversal of neighboring spins. As a result, the jumps in the hysteresis loops for higher field sweep rates occur at lower fields.

To date, in bulk materials, such a staircaselike behavior of the hysteresis loops is only expected to be observed for disorder systems. Exception is, however, the superconducting ferromagnetic UGe_2. Here, since the compound is crystalline and undiluted, it is hard to imagine any intrinsic source of disorder, provided that there is no site interchange between U and Ge. The jumps observed in UGe_2 appeared to occur at regular intervals of applied field and were attributed to a field tuned resonant tunneling of nanosized magnetic domains between quantum spin states.[17] Such arguments were later criticized by Lhotel *et al.*[18] who claimed that the steps observed in UGe_2 are dependent on the sample size and the field sweep rate and thus related to the pinning of domain walls by extrinsic defects. Anyhow, it appears difficult to understand the uniqueness of the staircaselike behavior of the hysteresis loop observed in UGe_2 among other ferromagnetic materials where extrinsic defects are also present.

Although at the time being, we do not completely rule out a mechanism that involves wall pinning by defects to account for the observed staircaselike behavior of the hysteresis loops in $PrVO_3$, in the following we would like to discuss the possibility that RF may be used to account for all of the intriguing results observed for $PrVO_3$.

To date, most of the theoretical work on the RF model has been done using Ising models in which the ordering involves the alignment of magnetic spins along a unique direction within the system. Many different models have been considered for disorder ferromagnets including the random anisotropy Ising model (RAIM),[19,20] the random-bond Ising model (RBIM),[21] and the RF Ising model (RFIM).[22] For antiferromagnets, it was realized that an effective RF can also be formed in diluted or mixed antiferromagnets (with a random distribution of the magnetic ions on the lattice positions) by the application of a uniform field along the uniaxial direction,[23] i.e., they can be mapped as the same universality class as the RFIM. $PrVO_3$ is not a diluted antiferromagnetic system. In addition, there is no mixed valence (as evidenced by the results of fitting to the Curie-Weiss law for Pr^{3+} and V^{3+}) and no site interchange between Pr and V in this compound. Here, we argue that the source of the RF in $PrVO_3$ may be an orbital quantum fluctuation (OQF). In RVO$_3$, we have already mentioned that the orbital degrees of freedom which come about as a result of partly filled degenerate d orbitals play an important role in the physics of these materials. Recently, Khaliullin *et al.*,[24] have pointed out that the quantum fluctuations between the degenerate d_{yz} and d_{zx} orbitals (note, the d_{xy} is occupied on every V site) may be quite large and tend to stabilize the ferromagnetic interaction along the c axis as realized in the spin C-type magnetic structure in $LaVO_3$. On the other hand, OQF can compete with long range magnetic ordering and might lead to the quantum melting of the long range magnetic order.[25] The latter effect of OQF is similar to that expected from the RF in diluted antiferromagnets.[26,27] A model for RF generated by OQF in $PrVO_3$ reflects the fact that the Jahn-Teller interaction in RVO$_3$ is relatively weak allowing OQF to come into play. We further note that this mechanism may also account for the lack of training effects observed in this system, a feature which differs from the diluted antiferromagnet case. Since OQF can occur at every site of crystal, the distribution of the sites with "*out of tune*" orbital is changed for different times of cooling. This results in a (slightly) different behavior in the $M(H)$ loops for different runs.

The RF mechanism can also be used to account for the intriguing behavior of the magnetization in the "*high*" field regime. We note that the aforementioned high negative values of the Weiss temperature of $PrVO_3$ clearly indicate that the antiferromagnetic interactions in the compound are very strong. A simple calculation based on the "*classical*" model for a homogeneous antiferromagnet would give the antiferromagnetic interaction strength in the order of 10^4-10^5 kOe. It means that one would then expect to observe a relatively low value of the magnetization, at least along one of the axis of crystal at an accessible applied field. However, in $PrVO_3$, above the applied field as low as 18 kOe we already observe more than 1/3 of the full moment of 2 μ_B expected for V^{3+} along all the axes of crystal. Above 18 kOe, the magnetization is only weakly field dependent and there is difference in the value of the high field magnetization obtained along the a, c axes and the b axis. These features look very similar to those of a weak antiferromagnet with strong crystal field effects. Here, the magnetic field is strong enough to break the antiferromagnetic couplings between the moments and create a forced ferromagnetic state, but it cannot entirely suppress the effect of the crystal fields which give rise to the reduced and anisotropic value of the "*saturation*" magnetization observed along different axes. In view of the RF, it appears to us that there is a fraction of spins having weak local fields and thus they can turn (or flip) to lie along the direction of the applied field. The remaining spins are strongly antiferromagnetically coupled (i.e., are hardly affected by the applied field) and are responsible for the observed SO temperature T_{SO}. In addition, we would like to note that the weak local fields of the former spins also imply that the crystal field effects can lead to the reduced magnetic moment as well as the anisotropy in the magnetization along different directions which are consistent with our observed results. In order to estimate the number of spins with weak local fields, we consider the ratio between M_r/M_s where M_s is the saturation magnetization which we assume smaller than 2 μ_B of the full moment expected for V^{3+}. At 1.5 K, the ratio M_r/M_s is then larger than 30% along the b axis; and 40% along the a and c axes. From these, we derive the number of spins with weak local fields in respect to the applied field of at least 20% which is determined as a half of the latter value.

Finally, we would like to use the RF mechanism to explain for the aforementioned unusual observed feature of the coexistence of antiferromagnetic and glassylike behavior in the ZFC, FC data. We note that a coexistence of spin glass and antiferromagnetic orders has been reported earlier for some (RF) diluted Ising system.[28,29] In $PrVO_3$, we can see that the fraction of spins with weak local field is responsible for the glassylike behavior. That is in the ZFC, the spins are randomly frozen resulting in the low value of the magnetization. With increasing temperature, the thermal energy allows the spin direction fluctuating and so let it easier be aligned along the direction of the applied field, thus the value of the magnetization increases. Above the glassy temperature, the thermal energy becomes too high and drives the spins away the direction of the applied field, and the magnetization decreases with increasing temperature. For the FC curve, at lowest temperature, the majority fraction of spins is already aligned along the direction of the applied field. The increase of temperature just releases these spins more away from the field's direction and so resulting in a monotonous decrease of the FC magnetization. A superimpose of the two different contributions, i.e., fraction of spins with weak local fields and the remaining strongly antiferromagnetically coupled spins, at different applied fields would result in the observed ZFC, FC of the compound. Furthermore, we would like to note that, under our proposed mechanism of the "*two phase*" behavior, i.e., with spin glasslike and long range antiferromagnetic orderings, the temperature where the low temperature peak occurs in the ZFC does not reflect the true glassy temperature T_g but it depends to some extent the contribution of each phase into the ZFC curve. The dominant contribution of the long range antiferromagnetic phase to the ZFC curve at low applied magnetic fields, leads to the shift of the anomaly to temperature higher than the actual T_g of the glassy phase or even the absence of the anomaly.

V. CONCLUSIONS

In summary, we have studied the magnetic properties of a single crystal $PrVO_3$. The compound reveals unusal behavior of the ZFC, FC, and hysteresis loops. Based on the observed results, we proposed that $PrVO_3$ should be considered as a disorder antiferromagnet with RF. We have argued that the origin of disorder (or RF) should come from the OQF of the compound. In order to confirm our proposed mechanism further experiments, e.g., neutron (inelastic) scattering, as well as theoretical calculations are required.

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[9] The crystal is detwinned in the *ac* and *bc* planes. Since the lattice parameter a (=5.4993 Å) is only slightly smaller than b (=5.5223 Å), it is not easy using the backscattering x-ray Laue technique to conclude whether or not the crystal is detwinned in the *ab* plane. Therefore the notation a and b should be understood as the two main axes in the *ab* plane which, as a result of twinning, might have an alternation of a and b axes.

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