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Magnetic properties of a LuVO₃ single crystal studied by magnetometry, heat capacity and neutron diffraction

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ABSTRACT

We have studied the magnetic properties of a LuVO₃ single crystal. The compound shows an orbital ordering at T_{OO} = 179 K followed by the antiferromagnetic spin ordering at T_{SO} = 109 K. In the magnetically ordered regime, there appears an abrupt change at T₀ = 82.5 K in the magnetisation, indicating a first-order transition. The compound has very large negative Weiss temperature observed along all the main crystallographic axes, suggesting a strong antiferromagnetic correlations in the paramagnetic state. The observation of hysteresis curves in the collinear antiferromagnetic regime is discussed in terms of an inhomogeneity generating some spins with weak local fields in a strongly antiferromagnetic matrix.

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1. Introduction

The interplay between spin-orbital interaction and phase transitions has attracted much interest recently in strongly correlated electron systems, in particular the transition metal (TM) oxides. Coupling to the lattice further enriches the interplay 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 3d e_g bands at the Fermi level. Perovskite Vanadates RVO₃ (R = rare earth and Y) belong to the same type but with 3d t_{2g} bands and show very different behaviour.

The crystal structure of RVO₃ is distorted from cubic to orthorhombic *Pbnm* symmetry by a cooperative rotation of the VO₆ octahedra [2]. A long-range magnetic ordering of the V sublattice has been observed at low temperatures for different RVO₃ compounds with the magnetic structures being either being C-type with the spins parallel along the *c*-axis but antiparallel in the *ab*-plane or G-type with the spins antiparallel along all directions [3]. In RVO₃ it was found that, with decreasing Lanthanide ionic radii, the onset temperature for the orbital ordering (OO) T_{OO} increases

(137 K for LaVO₃ and 179 K for LuVO₃) while the spin ordering (SO) temperature T_{SO} decreases monotonically (139 K for LaVO₃ and 107 K for LuVO₃). The crossover of T_{OO} and T_{SO} is between R = La and Ce. Recent Hartree-Fock studies [4] have shown that the C- and G-phases are energetically close, consequently, the interplay between different factors such as JT distortions, orbital quantum fluctuations, and the Dzyaloshinsky–Moriya interaction have led to very interesting properties.

In the perovskite-type RVO₃ compounds, the OO phenomenon investigated for LaVO₃ and YVO₃ indicated an orbitally induced-structural phase transition from orthorhombic to monoclinic when cooling through T_{OO}. In LaVO₃, the SO temperature T_{SO} is 139 K which is slightly above T_{OO} of 137 K [5], whereas it is equal to 116 K and so well below the 200 K of T_{OO} for YVO₃. When cooling down further, interestingly, an additional first-order phase transition appears at transition at T_S = 77 K for YVO₃ below which the orthorhombic phase is recovered and the magnetic structure becomes G-type [6,7]. Concerning the high temperature phase at T_S < T < T_{SO} in YVO₃, a magnetic neutron scattering study [8] has revealed some unusual features: i) the magnetic structure is non-collinear, and just more complex than previously assumed for the simple C-type; ii) The magnon band width as derived from inelastic neutron scattering along the ferromagnetic *c*-axis is larger than that in the antiferromagnetic *ab*-plane. This violates the standard Goodenough–Kanamoru rules according to which ferromagnetic superexchange interactions are generally substantially weaker than

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the antiferromagnetic interactions; iii) The spectrum is split into optical and acoustic magnons with a gap of 5 meV. To explain the latter feature, C. Ulrich *et al.* [8] proposed two different ferromagnetic exchange bonds J_c along the c -axis (i.e. dimerisation) which can be made possible by an orbital Peierls state due to the formation of an orbital singlet. However, Z. Fang *et al.* [9] argued that the splitting should be accounted for by the two different exchange interaction J_{ab} of inequivalent VO_2 layers which have different amounts of JT distortion.

For LuVO_3 , an earlier powder neutron diffraction (PND) study by Zubkov *et al.* [3] indicated that the compound has a G-type magnetic structure at low temperature. Muñoz *et al.* [10], also using PND, studied the structural and magnetic structure in the temperature range from 2 to 300 K. They pointed out that LuVO_3 has G-type magnetic ordering below $T_{\text{SO}} = 107$ K and this magnetic structure remains stable down to 2 K. The material also has an OO temperature of 190 K, but without any structural phase transition at this temperature. The change in the crystallographic structure from orthorhombic to monoclinic symmetry occurs instead between ~ 82 and 94 K, which is below the SO temperature. Recently, we have studied this compound in detail using high quality single crystals combining a variety of experimental methods including neutron and synchrotron studies [11]. In this work, a canted C-type magnetic structure was observed that transforms to a collinear G-type at lower temperature. It has also been shown that the features of orbital-Peierls state (i.e. orbital-singlet similar to spin-singlet dimers) attributed previously in YVO_3 [8] are in fact a consequence of the static OO and corresponding JT distortion.

In this contribution, we report on the magnetic, heat capacity, and neutron diffraction studies of single crystal LuVO_3 . The compound appears to be an antiferromagnet and its observed magnetic properties are consistent with the inhomogeneous nature of the compound.

2. Experimental details

Single crystal LuVO_3 was grown by means of the floating zone technique using a high temperature Xenon arc-furnace. At first, LuVO_4 was prepared by mixing stoichiometric quantities of Lu_2O_3 and V_2O_5 (with purity of 99.9%), followed by annealing at 1100 °C for 48 h. The product was then reduced at 1000 °C in flowing H_2 for 10 h to produce the LuVO_3 powder phase. The LuVO_3 feed and seed rods used for the single crystal growth were made by pressing the powder under hydrostatic pressure and then annealing these rods at 1500 °C under a flow of Ar. A similar procedure for single crystal growth is described elsewhere [12].

Measurements of the zero-field-cooled (ZFC) [13] and field-cooled (FC) magnetisation and the magnetic isotherms were carried out in a Quantum Design SQUID magnetometer. Here we use *zero* and *ZFC* in italics to indicate that we neglect the small trapped field in the superconducting solenoid of the magnetometer. For the FC measurements, the sample was cooled from the paramagnetic region to 1.8 K in an applied field, e.g. 0.1 kOe, with the data collected (FCC), then it was warming during the measurements (FCW). For the ZFC measurements, the sample was cooled in *zero* field to 1.8 K before the magnetic field was applied. The data were then taken on warming. Heat capacity measurements of the sample were carried out in a Quantum Design Physical Property Measurement System (PPMS) with a heat capacity option using a relaxation technique. The magnetic structure of the compound was determined from single crystal neutron diffraction measurements on the TriCS instrument at the Paul Scherrer Institute, Switzerland using a wavelength of 1.1807 Å [13].

3. Results and discussion

In Fig. 1, we present the results of the heat capacity measurements on the LuVO_3 single crystal; C vs. T (left scale) and C/T vs. T (right scale). Three transitions are detected and defined as the orbital ordering (OO) $T_{\text{OO}} = 179$ K, the spin ordering (SO) $T_{\text{SO}} = 107$ K, and $T_0 = 82.5$ K manifested as a drop in the heat capacity with decreasing temperature. The values of these transition temperatures are in good agreement with those obtained for the polycrystalline sample [10].

To determine the magnetic structure, the results of the neutron diffraction studies with some selected reflections as a function of temperature are presented in Fig. 2. Below $T_0 = 82.5$ K, we observed the magnetic contribution on top of the $(h\ k\ l)$ reflections with h zero or even, k odd and vice versa, l odd (e.g. $(0\ 3\ 1)$ $(0\ 1\ 1)$ reflections as seen in Fig. 2) indicating the collinear G-type magnetic structure [8]. Between T_0 and T_{SO} , a magnetic contribution is seen to develop on a different set of $(h\ k\ l)$ with h zero or even (odd), k odd (zero or even) and l zero or even characteristic of the C-type magnetic structure (see Fig. 2 for (100) and (012) reflections). The magnetic structure is, however, canted since the magnetic contribution due to a G-type magnetic structure is seen not to diminish completely which is also in accordance with that reported in Ref. [11].

To explore further, the results of the FCC and FCW $M(T)$ measured in two different magnetic fields of 0.1 kOe and 0.4 kOe along the principal axes are displayed in Fig. 3. There is an upturn in the magnetisation with decreasing the temperature at $T_{\text{SO}} = 107$ K. In the SO regime, there is an additional first order transition at T_0 of about 82.5 K, consistent with the observation of magnetic hysteresis between FCC and FCW data.

Earlier, we reported $M(T)$ measurements for some different RVO_3 compounds [12], and showed that the form of the ZFC curves are very much dependent on the very small value of the trapped field (TF) in the superconducting magnet of the SQUID magnetometer. We have examined this TF carefully. Before each measurement, we ran a degauss sequence to minimise the TF; its absolute value was estimated to be less than 2 Oe. We can “generate” a TF with opposite sign by reversing the sign of the magnetic fields in the degauss sequence [12]. In Fig. 4, it can be seen that the ZFC magnetisation measured in an applied field of 0.1 kOe observed after cooling in a positive TF (ZFC_PTF) is mirrored with that of the negative TF (ZFC_NTF) even though the TF is about two orders of magnitude smaller than the applied field used for the measurement. It is well known that for conventional magnetic materials, domain translation is reversible at (very) low magnetic

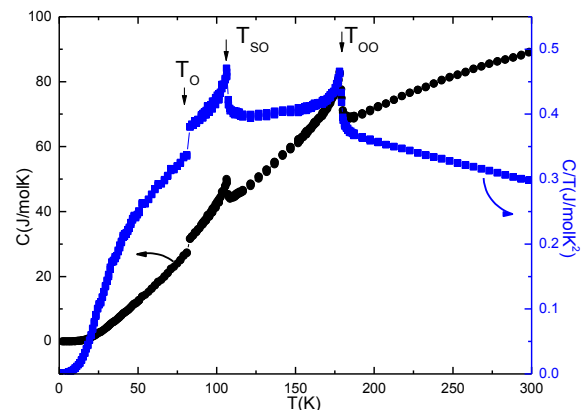


Fig. 1. Heat capacity C and C/T as a function of temperature for a LuVO_3 single crystal.

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