

High-energy X-ray Diffraction Study of Orbital Physics in $LnVO_3$ ($Ln = La, Ce, \text{ and } Y$)

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Introduction

In correlated electronic systems, the interplay between charge, spin, orbital degrees of freedom, and lattice distortion gives rise to a large variety of interesting physical properties (e.g., high- T_c superconductivity, colossal magnetoresistance [CMR] effect, ferroelectrics). It has been recently realized that the orbital degree of freedom plays an important role in these phenomena. Because of its strong coupling with charge, spin, and lattice dynamics, it often manifests itself in various structural responses (e.g., charge stripes, diffuse scattering, charge/orbital-ordering induced-phase transition). Among these responses, the orbital physics in transition metal oxides is of particular interest [1]. High-energy x-ray scattering is a powerful technique for probing the subtle structural changes that contain information that is essential for understanding the fundamental physics in these highly correlated systems.

The transition metal oxides with partially filled t_{2g} orbitals exhibit interesting phenomena, because of the relative weak Jahn-Teller coupling, high degeneracy, and greater orbital freedom when compared with the e_g orbitals. Recently, we have found temperature-induced magnetization reversals and a novel phase transition in YVO_3 single crystals [2]. The transition involves competition between single-ion anisotropy and antisymmetric interaction and a change of two types of spin and orbital ordering in a noncubic symmetry [3], where the G-type spin-ordering (G-SO) is associated with the C-type orbital-ordering (C-OO), and the C-type spin-ordering (C-SO) is associated with the G-type orbital-ordering (G-OO) (Fig. 1) [4]. However, in $LaVO_3$ and $CeVO_3$, the situation is different because they form in a pseudo-cubic structure with almost equal V-V bonds above T_N , in which quantum fluctuations among orbitals are expected to play an important role in the C-type magnetic ordering where both FM and AF coupling coexist. In addition to the relatively simple electronic configurations, it is a challenging task to understand the complex interactions between different degrees of freedom. It is thus of particular interest to perform detailed and systematic theoretical and experimental studies of the orbital physics in these systems.

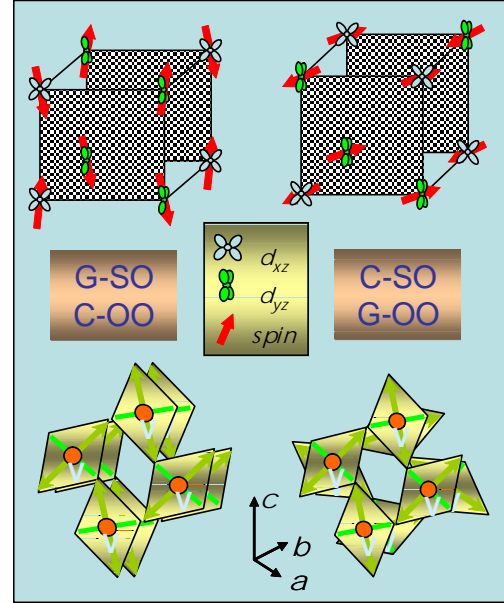


FIG. 1. Two types of spin- and orbital-ordering and associated lattice distortion in $LnVO_3$.

Methods, Materials, and Results

The structural phase transition in the orthovanadates $LaVO_3$ and $CeVO_3$ has been studied with high-resolution, high-energy synchrotron x-ray diffraction at beamline station 11-ID-C at the APS. $LaVO_3$ undergoes a second-order phase transition at $T_N = 143K$ and a first-order transition at $T_I = 141K$ (Fig. 2). In $CeVO_3$, however, there are second-order phase transitions occurring at $T_0 = 154K$ and first-order phase transitions occurring at $T_N = 134K$. These phase transitions are confirmed by specific heat measurements. The phase transition at T_I in $LaVO_3$ or T_0 in $CeVO_3$ is due to a G-type orbital ordering, which lowers the structure symmetry from orthorhombic $Pbnm$ to monoclinic $P2_1/b11$. The structure change at T_N in $CeVO_3$ is ascribed to an orbital-ordering-enhanced magnetostrictive distortion. The change at T_N in $LaVO_3$, however, is most probably due to an ordered occupation of the vanadium $3d t_{2g}$ orbitals associated with antiferromagnetic ordering. We propose that the

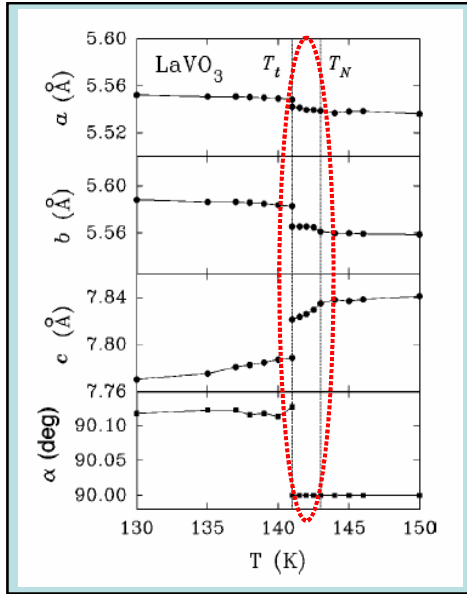


FIG. 2. Lattice parameters versus temperature of LaVO_3 .

first-order phase transition at T_t in LaVO_3 should be associated with a sudden change of both the spin and orbital configurations, similar to the phase transition at $T_s = 77\text{K}$ in YVO_3 [2-4], causing a reversal of the net magnetization. However, the ordered state above T_t in LaVO_3 is identical to that below T_s in YVO_3 . It has been found that with a decreasing lanthanide ionic radius from La, the Neel temperature T_N decreases while the orbital ordering onset temperature increases in these orthovanadates (Fig. 3). However, when the Ln^{3+} size further decreases, the system shows much more complex behavior, which is the subject of our current investigations.

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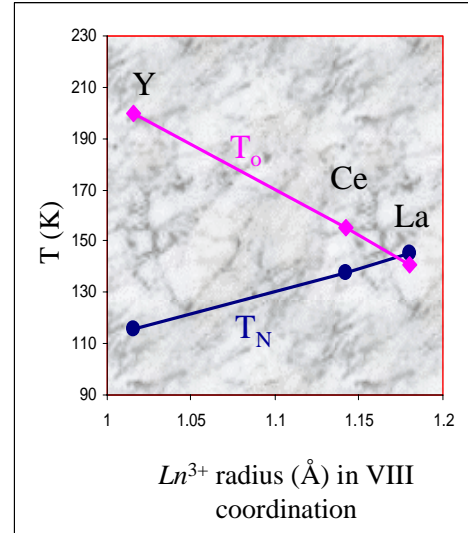


FIG. 3. Magnetic- (T_N) and orbital-ordering temperature (T_o) versus Ln ionic radius.

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References

- [1] Y. Tokura and N. Nagaosa, Science **288**, 462-468 (2000).
- [2] Y. Ren, T.T.M. Palstra, D.I. Khomskii, E. Pellegrin, A.A. Nugroho, A.A. Menovsky, and G.A. Sawatzky, Nature **396**, 441-444 (1998).
- [3] Y. Ren, T.T.M. Palstra, D.I. Khomskii, A.A. Nugroho, A.A. Menovsky, and G.A. Sawatzky, Phys. Rev. B **62**, 6577-6586 (2000).
- [4] G.R. Blake, T.T.M. Palstra, Y. Ren, A.A. Nugroho, A.A. Menovsky, Phys. Rev. Lett. **87**, 245501 (2002).