Materials Science

4C, 9C/2005G134

Orbital ordering in RVO_3 controlled by hydrostatic pressure

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Introduction

In addition to the charge and spin degrees of freedom, the orbital degree of freedom plays an important role in determining the electric and magnetic properties of the perovskite transition metal oxides. The orbital ordering is strongly coupled with the lattice distortion, i.e. Jahn-Teller distortion. Therefore, the purpose of the present study is to demonstrate the control of the orbital ordering phases in terms of the lattice distortion by applying pressure.

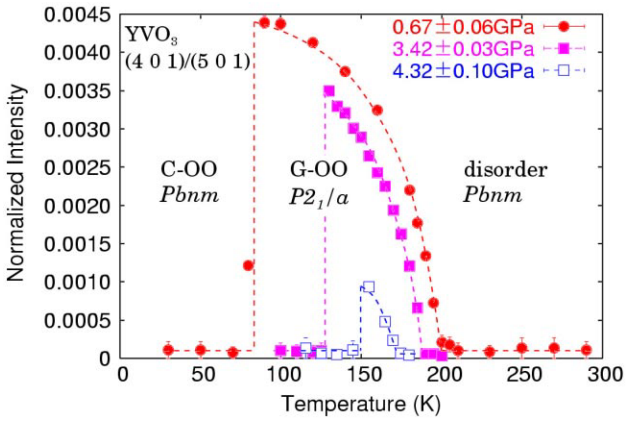
RVO_3 (R: rear earth or Y) shows various physical properties coupled with the spin and orbital states depending on R-ions [1]. RVO_3 (Tb-La) with large R-ionic radius undergoes a phase transition from an orbital disordered state to a G-type orbital ordering (G-OO) at T_{OO1}. The crystal structure also changes from orthorhombic ($Pbnm$) to monoclinic structure ($P2_1/a$) at T_{OO1} reflecting the orbital state. Moreover, the ground state of the magnetic structure becomes C-type spin ordering influenced by the orbital ordering. RVO_3 (Lu-Y) with smaller R-ionic radius has the G-OO phase at $T_{OO2} < T < T_{OO1}$, and a C-type orbital ordering (C-OO) with $Pbnm$ below T_{OO2}. In addition, the ground state of the magnetic structure becomes the G-type spin ordering. Consequently, the ground state of the orbital ordering in RVO_3 changes from G-OO to C-OO by R-ions substitution. In order to clarify the pressure effect on the orbital states, the pressure-temperature phase diagrams of the orbital ordering in RVO_3 (R = Y, Tb), which locates near the phase boundary between G-OO and C-OO, have been studied by x-ray scattering technique.

Experiments

The high-quality single crystals of RVO_3 were grown by a floating-zone method. X-ray scattering experiments were carried out at beam line 4C and 9C using a four-circle diffractometer. The x-ray energy was tuned to 18.0 keV with a Si(111) double-crystal monochromator, and the beam was focused by a bent cylindrical mirror. For the low-temperature and high-pressure experiments, a helium gas driven diamond-anvil cell (DAC) was mounted on a closed-cycle He cryostat, and the gas pressure and the temperature were controlled by a computer. Pressure was generated in a DAC using a 1:1 mixture of pentane and isopentane pressure medium, and was calibrated from a lattice constant of NaCl enclosed with the sample in the DAC. A typical sample size is 0.08x0.08x0.02 mm^3.

Results and discussions

The pressure-temperature phase diagram of the orbital state was studied in YVO_3, which has the C-OO phase as the ground state. The intensity of the forbidden reflection (401) of $Pbnm$ and the temperature dependences of lattice constants were measured. The forbidden reflection is only observed in the G-OO phase with the space group $P2_1/a$, and the intensity should be proportional to the square of the order parameter.



The figure shows the temperature dependences of the (401) intensity under high pressure. The intensities at ~ 0.67 GPa are observed below T_{OO1} ~ 200 K and disappears at T_{OO2} ~ 80 K as shown by closed circles. Namely, the space group becomes $P2_1/a$ of the G-OO at $T_{OO2} < T < T_{OO1}$, and the C-OO appears below T_{OO2}. The results are consistent with the previous experiment in ambient pressure [2]. With increasing pressure, T_{OO2} clearly increases and T_{OO1} decreases. As a result, applying pressure stabilizes the C-OO phase, and the intermediate phase, G-OO, is expected to disappear above 6 GPa. Namely, the phase transition from the orbital disordered state to the C-OO can be observed, and such a phase transition has never been observed in the study of R-ions substitution.

$TbVO_3$ has the G-type as the ground state, and is expected to locate near phase boundary between C-OO and G-OO. We also try to switch the ground state of orbital ordering by applying pressure in $TbVO_3$.

References

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