The "devil's staircase" type phase transition in NaV_2O_5 under high pressure

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Introduction

It has been recognized that the inorganic compound NaV_2O_5 shows the spin-gap formation, lattice dimerization (2a×2b×4c) and charge ordering simultaneously at its transition temperature $T_c = 35$ K under ambient pressure. In 1999, Sekine[1)] measured the dielectric constant under high pressure and discovered that an intermediate phase exists at 0.5 GPa < P < 1.2 GPa and 18 K < T < 28 K. In this paper we report the detailed x-ray diffraction study on the intermediate phase from a microscopic point of view.

Experimental

X-ray study of NaV_2O_5 under high pressure and low temperature was carried out at BL-1B MPD (Micro Powder Diffractometer) and BL-4C. An Imaging plate (IP), an x-ray CCD system and a scintillation counter were used as a detector upon experimental requirements. Wavelength was tuned to 0.7 Å with a Si(111) double-crystal monochromator. For the low-temperature and high pressure experiment, a diamond anvil cell (DAC) was mounted on a closed-cycle He-gas refrigerator. Pressure was generated in the DAC using a 4:1 mixture of methanol/ethanol as pressure media and determined from the lattice constant of NaCl. The specimen temperature was monitored with a Au-0.07%Fe chromel thermocouple mounted on the DAC surface. We used as-grown high quality single crystals of NaV_2O_5 with the size of 150 μm × 400 μm × 80 μm ($a \times b \times c$).

Results and Discussions

The "devil's staircase" type phase transition has been discovered at low temperature and high pressure. Figure 1 shows the Temperature-Pressure (T-P) phase diagram. A large number of transitions are found to successively take place among higher-order commensurate phases with $2a \times 2b \times zc$ type superstructures. The observed temperature and pressure dependence of modulation wave number q_c, defined by $1/z$, is well reproduced by the Axial Next Nearest Neighbor Ising (ANNNI) model[2)] . The q_c is speculated to reflect atomic displacements coupled with charge ordering in this system. The experimental fact implies that two competitive interlayer interactions between the Ising spins, i.e., the nearest neighbor $J_1>0$ (ferro) and the next nearest neighbor $J_2<0$ (antiferro) along the c-axis, are intrinsic in this compound. Though a microscopic origin of the interlayer interaction is not yet known, the nearest neighbor interaction $J_1>0$ between the V_2O_5 layers is especially interesting. It is very surprising that the phase transition of such a complicated charge-lattice-spin coupled system NaV_2O_5 can be interpreted by the simple ANNNI model.

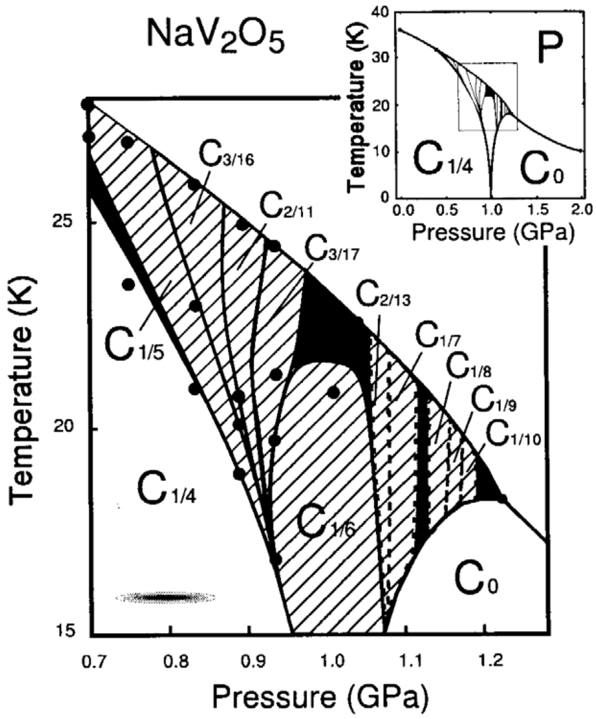

Fig. 1. Experimentally observed T-P phase diagram of NaV_2O_5. The hutched area shows commensurate phases unambiguously identified while the shaded area indicates more complicated higher-order commensurate or incommensurate phases unresolved by the present T-P resolution, which is represented with an ellipse at the left bottom corner. The phase boundary experimentally not clearly confirmed is drawn with dotted lines.

1) Y. Sekine, Doctoral thesis, (The University of Tokyo, 1998).
2) Per Bak and J. von Boehm, Phys. Rev. **B21**, 5297 (1980).

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