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Absence of the Itinerant Weak Antiferromagnetic Ordering and the Anomalous Suppression of the Low Energy Spin Excitation in Metallic V_2O_3 under Hydrostatic Pressure

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Neutron scattering experiments have been carried out in order to study the magnetic ordering and excitation in stoichiometric V_2O_3 under hydrostatic pressure up to 3.2 GPa. No itinerant weak antiferromagnetism has been observed in the metallic phase down to 1.5 K. The lattice distortion and the abrupt change in magnetic moment imply that the MI transition remains of first order up to the critical pressure. We found a remarkable suppression of the low energy spin excitation in paramagnetic metallic phase stabilized by pressure at low temperatures. This result is consistent with a recent high pressure NMR study which reported a clear suppress of the spin relaxation and Knight shift.

KEYWORDS: V_2O_3, Mott transition, high pressure

§1. Introduction

The nature of the ground state properties and the magnetic excitation in strongly correlated electron systems have attracted considerable recent interests. V_2O_3 is considered to be a prototype of the three-dimensional Mott-Hubbard system.[1)] Stoichiometric V_2O_3 exhibits a metal-insulator (MI) transition between high temperature paramagnetic metal (PM) and low temperature antiferromagnetic insulating (AFI) phases. The origin of the AFI phase is still under discussion. Recent synchrotron x-ray scattering study revealed the orbital ordering associated with the MI transition.[2)] The transition temperature decreases with applying pressure and the MI transition is suppressed above the critical pressure 2.5 GPa. So far no magnetically ordered phase has been reported in the metallic side of the MI transition in V_2O_3 .[3,4)] On the other hand, a recent neutron scattering study reported an itinerant SDW state in non-stoichiometric $V_{2-y}O_3$ below the Néel temperature T_N = 10 K.[5)] They found a helical magnetic structure of V moment with the size of 0.15 μ_B. From the theoretical point of view the existence of the itinerant weak antiferromagnetism is a key issue for the understanding of the MI transition. On the contrary a recent NMR study concluded the existence of the two distinct vanadium sites in $V_{2-y}O_3$, namely magnetic guest sites introduced by defects and host sites which have the same electronic state as in V_2O_3.[6,7)] Surprisingly the NMR signal associated with the host site shows a clear spin gap behavior with a strong suppression of the Knight shift below 80 K without any evidence for the magnetic ordering. Very recently a similar behavior is observed in relaxation and Knight shift in V_2O_3 stabilized under high pressure.[8)]

In this paper we report the results of the high pressure neutron scattering experiments on V_2O_3 in order to study the magnetic ordering and excitation near the MI transition.

§2. Experiments

Neutron scattering experiments were carried out using thermal neutron triple axis spectrometer TAS-1 installed at the research reactor JRR-3M in JAERI. We used a vertically bent (PG) monochrometer and a flat PG analyzer. A PG filter and Al_2O_3 single crystal were used to reduce higher order contamination. The incident energy was fixed at 14.7 meV or 30.5 meV. The horizontal collimation was 40'-80'-40'-80'. Hydrostatic pressure was applied with use of Macwhan-type cell.[9)] As a pressure transmitting medium Fluorinert was sealed with sample. The pressure was applied at room temperature and calibrated at 4.2 K by the lattice constant of NaCl encapsuled with sample. The sample was mounted with the [110] and [001] axes parallel to the scattering plane. We used hexagonal unit cell to describe the reciprocal lattice points. In this notation (1/2 1/2 0) and (0 0 1.7) corresponds to the antiferromagnetic wave vector for the AFI and AFM phase, respectively.

§3. Results

Figure 1 (a) shows the pressure dependence of the (1/2 1/2 0) magnetic Bragg peak from AFI phase at T=1.5 K. A resolution-limited (1/2 1/2 0) peak was observed below 1.3 GPa, while no trace of the magnetic peak was observed for $p \geq 2.4$ GPa. It means that the AFI phase was completely suppressed above 2.4 GPa. Figure 1 (b) shows the (00l) profile around the (0 0 1.7), where a strong incommensurate SDW peak is observed in $V_2O_{3.04}$. In V_2O_3, no SDW peak was observed down to 1.5 K from ambient pressure up to 3.2 GPa. From the statistical error of the present experiment, the upper

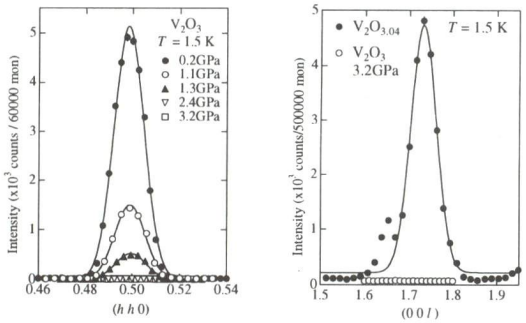

Fig. 1. (a) The pressure dependence of the (1/2 1/2 0) AFI peak in V_2O_3 observed at T= 1.5 K. (b) The (10l) scan in $V_2O_{3.04}$ and the one of metallic phase in V_2O_3 at 3.2 GPa.

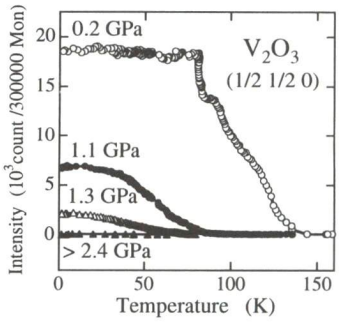

Fig. 2. The temperature dependence of the (1/2 1/2 0) AFI peak intensity for various pressures.

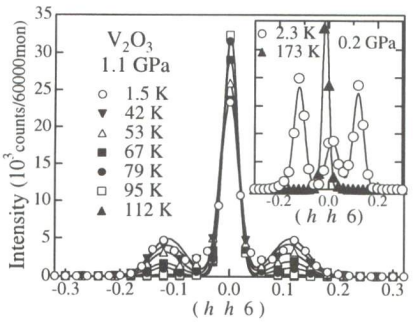

Fig. 3. The temperature dependence of the (006) transverse scan for p = 1.1 GPa. The inset of Fig. 3 shows the transverse profile of the (006) nuclear peak observed in the AFI phase at 2.2 K and PM phase at 173 K.

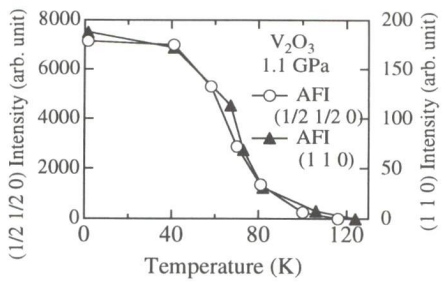

Fig. 4. The temperature dependence of the intensities of (1/2 1/2 0) magnetic and (110) nuclear peaks from AFI phase at 1.1 GPa.

limit of the moment size is estimated to be 2x10^{-4} μ_B from the structure factor calculation, assuming the same magnetic structure as in the SDW state in $V_2O_{3.04}$. Furthermore we have found no magnetic peak in (h h l), (0 0 l), (h h 1), and (h h 3) scans. Therefore we conclude that there is no magnetic ordering in the metallic state of V_2O_3 stabilized by the pressure.

Figure 2 is the temperature dependence of the (1/2 1/2 0) peak intensity for AFI phase as a function of pressure. For p = 2.4 GPa no intensity was observed. At lower pressures, the (1/2 1/2 0) intensity increases gradually and saturates at low temperatures. The pressure dependence of the saturated intensity could be understood either the coexistence of the AFI with the PM phase or the change in magnetic moment.

Figure 3 is the temperature dependence of the (006) transverse scan for p = 1.1 GPa. The AFI peak was observed below 100 K and the intensity increases with decreasing temperature. Surprisingly, the (006) peak from the PM phase was also observed down to 1.5 K. The inset of Fig. 3 shows the results for p = 0.2 GPa, which showed MI transition in whole sample volume. A sharp (006) peak in the PM phase splits into three reflections associated with the MI transition. The results for p = 1.1 GPa indicate that the PM phase coexists with the AFI phase at this pressure. From these profiles we found that the MI transition is of first order at high pressures where the PM phase does not completely disappear at low temperatures. Figure 4 shows the temperature dependence of the intensities of (1/2 1/2 0) magnetic and (110) nuclear peaks from the AFI phase at 1.1 GPa. As is shown in Fig. 4, the magnetic intensity shows almost the same temperature dependence as the nuclear peak. This result indicates that the temperature dependence of the magnetic peak intensity can be explained in terms of the volume change in the AFI phase without significant change in the magnetic moment. This negligibly weak temperature dependence is reasonable because the exchange interaction in AFI phase is very large. This result indicates that the MI transition maintains strong first order at even near critical pressure, where only small amount of PM phase is transformed into the AFM phase.

Figure 5 shows the constant-Q profile at (1 0 0.3) for the metallic V_2O_3 at 4.2 K with pressure of 2.5 GPa. This wave number corresponds to the magnetic Bragg point in $V_{2-y}O_3$. At this point a strong magnetic response has been reported in $V_{2-y}O_3$ as well as the PM phase in V_2O_3 above 200 K at ambient pressure. We have confirmed that no trace of the AFI and SDW phase was involved in our sample under pressure by the elastic experiments. Although the signal is weak and scattered due to the absorption and background from the pressure cell, we observed an inelastic peak around 4 meV. The dotted line in Fig. 5 is the background measured at (1 0 1.2). The inset of Fig. 5 is the (1 0 l) scan with fixed energy transfer at 2 meV and 4.8 meV, respectively. We found that the inelastic response exhibits a broad maximum around l = 0. It is particularly interesting to compare these data to the one of the non-stoichiometric sample. Figure 6 shows the constant-Q profile at the magnetic Bragg point (1 0 2.3) for $V_2O_{3.04}$ at 1.5 K in the ambient pressure. A broad quasi-elastic like response was observed as reported before.[10,11] In the constant-E scan along the (1 0 l) direction a broad maximum was observed around l = 0. This peak is due to the superposition of the magnetic response at (1 0 ±0.3) which

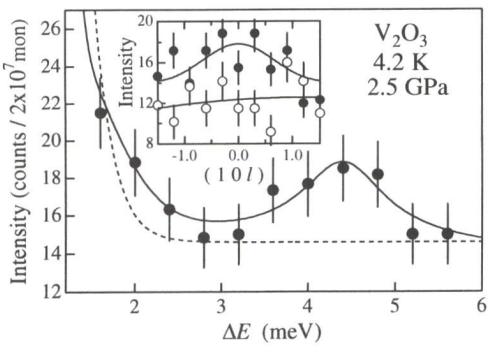

Fig. 5. The constant-Q profile at (1 0 0.3) for the metallic phase of V_2O_3 at 4.2 K stabilized with pressure of 2.5 GPa. The dotted line is the background. The inset shows the constant energy scan along the (10l) with ΔE at 2 meV (open circles) and 4.8 meV (close circles), respectively. The lines are guide to the eyes.

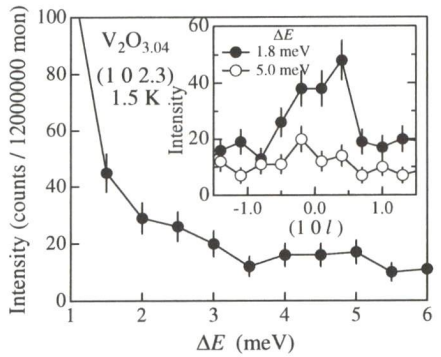

Fig. 6. The constant-Q profile at the magnetic Bragg point (1 0 2.3) for $V_2O_{3.04}$ at 1.5 K in the ambient pressure. The inset is the constant-E scan along the (10l) direction with ΔE at 1.8 meV (close circles) and 5 meV (open circles), respectively. The lines are guide to the eyes.

come from the twin structure of our sample. By comparing these data, we noticed a clear suppression of the low energy excitation in V_2O_3. It reminds us the spin gap behavior in the strongly correlated systems like high-T_c compounds.

§4. Discussions

From our systematic neutron elastic scattering experiments under high pressures, no magnetic ordering has been observed in the metallic phase of stoichiometric V_2O_3 stabilized by hydrostatic pressure. The absence of the magnetic ordering has been confirmed even in the metallic phase coexisting with AFI phase down to 1.5 K, which is much lower than T_N ($\sim$ 10 K) in non-stoichiometric sample. The metallic phase in coexisting state can be considered to be at the boundary of the MI transition. Therefore we conclude that in stoichiometric V_2O_3 there is no itinerant weak antiferromagnetic state at the boundary of the MI transition. This result is different from the non-stoichiometric case. The SDW state appears in $V_{2-y}O_3$ even for a very dilute defect concentration with $y = 0.012$, but not pure system.[11] Therefore the SDW state is peculiar to the charge doping introduced by defects. It implies that the appearance of the SDW state is not inevitable near the MI transition in V_2O_3. Furthermore, our result gives rise to an interesting question, if the SDW state is a homogeneous and intrinsic state or not. Anyhow it is clear that the coexistence of magnetic guest site and host site should be taken more serious to consider the magnetic structure and excitation in non-stoichiometric system.

Very recently Takigawa *et al.* reported a steep decrease of Knight shift and the relaxation $1/T_1$ in V_2O_3 under pressure.[8] They pointed out a striking similarity to the spin gap behavior observed in normal state of lightly doped cuprate superconductors. In addition, they concluded that nuclear relaxation at high temperatures is mostly due to orbital contribution. Our neutron scattering study indicates that the peak of relaxation rate is at least partly associated with spin contribution. These results would be indicative of the fact that the opening gap in spin channel in the metallic state near MI transition is an inevitable phenomenon in strongly correlated electron system. For this purpose further high-pressure inelastic scattering experiments are necessary to reveal the spin-gap nature in V_2O_3.

§5. Conclusions

High pressure neutron scattering experiments have been carried out in order to study the magnetic ordering and excitation in V_2O_3 up to 3.2 GPa. No itinerant weak antiferromagnetism has been observed in the metallic phase down to 1.5 K. We found a remarkable suppression of the low energy spin excitation in paramagnetic metallic phase stabilized by pressure at low temperatures. This result is consistent with a recent high pressure NMR study which reported a clear suppress of the spin relaxation and Knight shift.

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