Magnetic Excitation of BaCoS₂ near the Mott Transitions Induced by Pressure and Carrier-Number Control

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Neutron inelastic measurements have been carried out to study the dynamical magnetic properties of BaCoS₂ near the Mott transitions induced by two different means, by the application of pressure and by carrier-number control. The low energy excitation spectra have been found to be significantly different between thus induced metallic phases with similar Néel temperatures and similar antiferromagnetic staggered moments at low temperatures.

[BaCoS₂, Mott transition, neutron inelastic scattering, magnetic fluctuation]

1. Introduction

BaCo_{1-x}Ni_xS₂ has a layered structure which consists of an alternating stacks of rock-salt type BaS layers and Co_{1-x}Ni_xS layers, and is expected to have two-dimensional (2D) electronic nature [1]. BaCoS₂ is in the Mott-insulating state and antiferromagnetic with the Néel temperature T_N 305 K. The spin structure was determined by neutron diffraction studies, where Co²⁺ ions were found to have the spin $s=3/2$ [2]. The electronic configuration is probably $(yz \text{ or } zx)^1 (3z^2-r^2)^1 (x^2-y^2)^1$ [2,3]. By the Ni²⁺ (d⁸)-substitution for Co²⁺ (d⁷), electrons are introduced into the Co_{1-x}Ni_xS layers and the system becomes paramagnetic and metallic in the whole temperature (T) region when x exceeds 0.22. In the x -region around 0.22, Hall coefficients R_H , thermoelectric powers S and magnetic susceptibility χ have been found to exhibit anomalous T -dependence similar to those of high- T_c oxides [4]. In high- T_c oxides, the anomalous behavior of various physical properties is related to the strong low energy magnetic excitations surviving even in the hole-doped metallic phase near the Mott transition. Therefore, it is important for the understanding of the physical properties of the present system to study its low energy magnetic excitations near the Mott transition.

For this system, it has been revealed that the Mott transition can also be induced by external pressure $p \sim 1.5$ GPa [5,6]. Then, an interesting question arises how the electronic characteristics are different between the metallic phases in the vicinity of the Mott transitions induced by the pressure and by the carrier-number control. Neutron inelastic scattering measurements have been carried out near the Mott transitions to study dynamical magnetic properties of BaCoS₂ [7] under pressure and BaCo_{0.82}Ni_{0.18}S₂ at ambient pressure [3], where their magnetic characteristics have been found to be significantly different between these metallic phases [7].

2. Experimental

Single crystals of BaCoS₂ and BaCo_{0.82}Ni_{0.18}S₂ used in the present experiments were grown as described elsewhere [8]. BaCo_{0.82}Ni_{0.18}S₂ undergoes the antiferromagnetic transition at T_N

~ 180 K and exhibits metallic (non-metallic) T -dependence of the resistivity above (below) T_N . Neutron measurements were carried out by using the triple axis spectrometers, ISSP-PONTA and HQTAS installed at JRR-3M of JAERI in Tokai. The (002) reflection of pyrolytic graphite (PG) were used for both monochromator and analyzer. In the inelastic measurements, typical horizontal collimation were 40'-40'-80'-80'. The crystals were oriented with the [110] axis vertical, where $Q=(h,h,l)$ points in the reciprocal space can be reached. In high pressure measurements, a clamping-type cell was used [9]. The pressure was measured by monitoring the lattice constant of a NaCl crystal. Pressure values shown in the present paper are the ones determined at room temperature. Judging from the p -dependence of T_N , the present estimation of p is slightly larger

Fig. 1. Magnetic scattering profiles of BaCoS₂ observed at various temperatures and at various energies.

Fig. 2. Temperature dependence of the square of inverse correlation length κ_{RPA}^2 determined by using the RPA form.

than that of ref. [10]. This difference is probably caused by the fact in these experiments different kind of pressure media were used and therefore the T -dependences of p were different. Further details of the present experiments are described in refs. [3] and [6].

3. Results and Discussion

Figure 1 shows the magnetic scattering intensities of BaCoS_2 measured at ambient pressure along (h, h, l) at various temperatures above T_N and at various transfer energies E . A peak structure has been observed around $h=1/2$ even above T_N . The peak intensity has only weak l -dependence, which mainly originates from that of the magnetic form factor. These results show that the present system has the 2D magnetic nature. Similar 2D-like magnetic excitations have also been observed for $\text{BaCo}_{0.82}\text{Ni}_{0.18}\text{S}_2$. These data have been analyzed above T_N by applying the formalism of the random phase approximation (RPA), to see to what extent the theory can describe the observed characteristics of the dynamical magnetic behavior of this strongly correlated electrons [3]. The RPA form used here to describe the spectral function $\chi''(q, \omega)$ is

$$\chi''(q, \omega) = \frac{\alpha_0 \omega \Gamma_0 \bar{\chi}_q}{\Gamma_0 (\kappa_{\text{RPA}}^2 + q^2)^2 \xi_0^4 + \omega^2}, \quad (1)$$

where Γ_0 is a characteristic energy of spin fluctuations, κ_{RPA} is an inverse correlation length, $\bar{\chi}_q$ is the magnetic susceptibility of noninteracting spin system at q measured from Q_{AF} , the magnetic Bragg point below T_N , ξ_0 is the length scale ($\sim$ lattice constant), α_0 is a scale factor. First, the T -dependent parameters Γ_0 and α_0 were determined by using the observed ω -dependences of the q -integrated intensity χ' and the profile width at a fixed T . Then, κ_{RPA}^2 was

calculated at each T by using eq. (1) with these parameters. As shown in Fig. 2, where κ_{RPA}^2 is plotted against T , RPA presents a qualitative explanation of the data. However, κ_{RPA}^2 does not approach zero as $T \rightarrow T_N$. In contrast to the case of $\text{BaCo}_{0.82}\text{Ni}_{0.18}\text{S}_2$, κ_{RPA}^2 of BaCoS_2 deduced by RPA approaches zero when $T \rightarrow T_N$ [7]. Then, the nonvanishing behavior of κ_{RPA}^2 as $T \rightarrow T_N$ observed for $\text{BaCo}_{0.82}\text{Ni}_{0.18}\text{S}_2$ may be caused by the randomness effects introduced by the Ni-substitution for Co, although it is tempting to attribute it to an effect of the strong electron correlation which cannot correctly be described within the RPA treatment.

Figure 3 shows the T -dependence of magnetic Bragg intensities of BaCoS_2 taken at $(1/2, 1/2, -2)$ for various p -values. In the figure, the data for $\text{BaCo}_{0.82}\text{Ni}_{0.18}\text{S}_2$ at ambient pressure are also shown, where anomalous decrease of the intensity at ~ 70 K with decreasing T is possible spin-structure change caused by the sulfur deficiency. Figure 4 shows the magnetic scattering profiles of BaCoS_2 taken at $E=3$ meV for various p -values. The data under the pressure $0 \leq p \leq 1.4$ GPa were taken at room temperature ($\sim T_N$). The data at 1.5 GPa were taken at 225 K ($\sim T_N$). The similar data of $\text{BaCo}_{0.82}\text{Ni}_{0.18}\text{S}_2$ taken at ambient pressure and at $T \sim T_N$ are also shown. In the Figure, the relative intensities can be compared, because they are scaled by the integrated intensities of the nuclear Bragg reflections. The

Fig. 3. Temperature dependence of magnetic Bragg intensities of BaCoS_2 measured at various pressure values, and that of $\text{BaCo}_{0.82}\text{Ni}_{0.18}\text{S}_2$ at ambient pressure. The antiferromagnetic moments per $(\text{Co}_{1-x}\text{Ni}_x)$ atom at low temperatures are also shown.

Fig.4. Magnetic scattering intensities of BaCoS_2 measured at $E=3$ meV for various p -values, and that of $\text{BaCo}_{0.82}\text{Ni}_{0.18}\text{S}_2$ at ambient pressure. The zero point of each vertical axis is shown by the horizontal line with the corresponding pressure value.

Fig.5 Magnetic form factors $f(Q)$ deduced from the integrated intensities of the magnetic Bragg reflections taken for BaCoS_2 and $\text{BaCo}_{0.82}\text{Ni}_{0.18}\text{S}_2$ at various pressures. Closed and open symbols correspond to the data at $Q=(1/2, 1/2, l)$ and $(3/2, 3/2, l)$, respectively.

magnetic excitation spectra of BaCoS_2 at 3 meV changes drastically with pressure, while T_N and ordered moments do not exhibit such a drastic reduction. For $\text{BaCo}_{0.82}\text{Ni}_{0.18}\text{S}_2$ the magnetic excitation spectra can clearly be observed at $E=3$ meV, which has clear contrast to the almost complete disappearance of the scattering peak in BaCoS_2 under the pressure of ~ 1.5 GPa (Note that their T_N values and the antiferromagnetic moments at low temperatures are rather similar as shown in Fig. 3.)

Figure 5 shows the magnetic form factors deduced for BaCoS_2 at various p -values and for $\text{BaCo}_{0.82}\text{Ni}_{0.18}\text{S}_2$ at ambient pressure. With increasing p , a significant change of the form factors of BaCoS_2 at $(3/2, 3/2, l)$ can be seen at around $p \sim 1.5$ GPa. The results indicate that magnetic electrons begin to spread within the CoS planes at the corresponding pressure. They cannot be explained by the redistribution of electrons among three 3d-orbitals and suggest an increasing tendency of the itinerant nature of the electrons within the CoS planes with increasing pressure.

These results imply that the metal-insulator transition under pressure is induced by the increase in the intra plane transfer energy t , or by the weakening of the correlation effect. With increasing pressure, the kinetic energy of the electrons increases and as a result of this change, the magnetic excitation spectra become significantly broader in the energy space than those in the metallic phase derived by carrier-number control. The "localized" nature of the electron spin is not, on the other hand, easily weakened by the carrier-number change through the Mott insulator $\rightarrow$ metal phase boundary. The present finding may provide useful information for the understanding the electronic nature of high- T_c oxides.

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