$Ba_3ZnRu_2O_9$および関連物質が示す特異で多彩な磁気状態

Various novel magnetic states of $Ba_3ZnRu_2O_9$ and related compounds

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（概要）

We have investigated a novel type of quantum spin liquid in $Ba_3ZnRu_2O_9$, which has a hexagonal lattice of Ru^{5+} dimers. We have studied the magnetic behavior of the Nb-doped system $Ba_3Zn(Ru_{1-x}Nb_x)_2O_9$, where the Nb^{5+} $(4d)^0$ is non-magnetic ion and disturbs the formation of the Ru^{5+} dimer. By the Nb-doping, a ferromagnetic transition appears at around 100 K for $Ba_3Zn(Ru_{1-x}Nb_x)_2O_9$ $(0.08 < x)$. In order to study the magnetism of quantum spin liquid and/or ferromagnetic states at low temperature, we performed neutron powder diffraction of three samples $Ba_3Zn(Ru_{1-x}Nb_x)_2O_9$ (x = 0.25, 0.40, and 0.50). The three magnetic Bragg peaks with tiny diffraction intensity for x=0.25. We have found that these magnetic Bragg peaks vanish at $x_c = 0.27$. We have also studied the Ca-doped system $Ba_3Zn_{1-y}Ca_yRu_2O_9$, where paired spins in the Ru_2O_9 dimer form a non-magnetic spin singlet state with spin gap for $Ba_3CaRu_2O_9$ (y=1). we have also performed neutron powder diffraction of two samples $Ba_3Zn_{1-y}Ca_yRu_2O_9$ (y = 0.25 and 0.50). From the results of neutron diffraction measurements, we discuss a novel scenario and various magnetic states in $Ba_3ZnRu_2O_9$ and related compounds.

キーワード：スピン液体、Ru ダイマー、特異な磁気状態、長距離磁気秩序

（1 行あける）

１．目的

We have investigated a novel type of quantum spin liquid in $Ba_3ZnRu_2O_9$, which has a hexagonal lattice of Ru^{5+} dimers. In the temperature (T) dependence of the magnetic susceptibility (χ) of $Ba_3ZnRu_2O_9$, no trace of the Curie tail or glassy behavior has been detected down to 50 mK.[1,2)] We studied the magnetic behavior of the Nb-doped system, $Ba_3Zn(Ru_{1-x}Nb_x)_2O_9$, where Nb^{5+} $(4d)^0$ is a non-magnetic ion that disturbs the formation of the Ru^{5+} dimer. The $\chi - T$ curves of the Nb-doped system also show no trace of the Curie tail at low temperatures, indicating that the local Ru^{5+} spin induced by Nb-doping does not act like a free spin. The spin liquid state of $Ba_3ZnRu_2O_9$ has been found to be robust by impurity doping. By the Nb-doping, a ferromagnetic transition appears at around 100 K for $Ba_3Zn(Ru_{1-x}Nb_x)_2O_9$ $(0.08 < x)$. To study magnetic dynamics at low temperatures, we have attempted to perform neutron powder diffraction of three samples $Ba_3Zn(Ru_{1-x}Nb_x)_2O_9$ (x = 0.25, 0.40, and 0.50) down to 10 K. In contrast, for $Ba_3CaRu_2O_9$, paired spins in the Ru_2O_9 dimer form a spin singlet, resulting in the non-magnetic ground state with a spin gap (SG). It is interesting in the magnetic behavior for mixing compounds $Ba_3Zn_{1-y}Ca_yRu_2O_9$. We have also attempted to perform neutron powder diffraction of two samples $Ba_3Zn_{1-y}Ca_yRu_2O_9$ (y = 0.25 and 0.50).

２．方法

We measured the powder neutron diffraction profiles of three samples $Ba_3Zn(Ru_{1-x}Nb_x)_2O_9$ (x = 0.25, 0.40, and 0.50) and $Ba_3Zn_{1-y}Ca_yRu_2O_9$ (y = 0.25 and 0.50) using HRPD in JRR-3. Measurement temperatures are at lowest temperature (< 10 K) and 130K.

３．結果及び考察

Figure 1 shows neutron diffraction profiles of $Ba_3Zn(Ru_{1-x}Nb_x)_2O_9$ with x=0.25 taken at 4 K and 130K. Black line of Fig. 1 indicates the intensity difference between the data at 4 K and at 130K. From the previous experiments, we observed the magnetic Bragg reflections 113, 202, and 204 for the non-doped sample $Ba_3Zn(Ru_{1-x}Nb_x)_2O_9$ with x=0. In this time, we can see the tiny intensities of the magnetic reflections 113, 202, and 204, indicating that the magnetic ground state of the spin system for x=0.25 is magnetic long-range ordered state. We do not observe the magnetic Bragg peaks for x=0.40 and 0.50. Because we can detect three magnetic Bragg peaks only, it is not easy to determine the magnetic structure of this system. We are analyzing the magnetic structure comparing that of the similar antiferromagnetic materials $Ba_3MRu_2O_9$ (M=Co, Ni, and Cu). Figure 2 shows the Nb concentration x-dependence of the magnetic Bragg intensities for $Ba_3Zn(Ru_{1-x}Nb_x)_2O_9$, indicating the intensities monotonically decrease with increasing y. From fitting, the phase boundary of long-range ordered state is evaluated at x_c=0.27.

Figure 3 shows neutron diffraction profiles of $Ba_3Zn_{1-y}Ca_yRu_2O_9$ with y=0.25 taken at 4 K and 130K. Brown line of Fig. 3 indicates the intensity difference between the data at 4 K and at 130K. We can see the tiny intensities of the magnetic reflections 113, 202, and 204, indicating that the magnetic ground state of the spin system for y=0.25 is magnetic long-range ordered state. More experimentation is needed to understand the Ca-doping effect for $Ba_3Zn_{1-y}Ca_yRu_2O_9$.

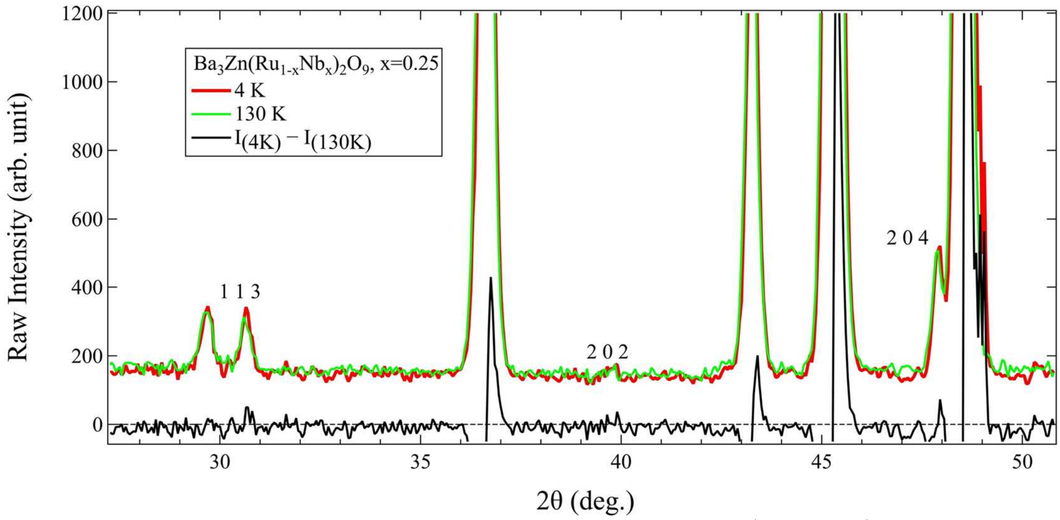

Fig. 1: Profiles of the neutron diffraction of $Ba_3Zn(Ru_{1-x}Nb_x)_2O_9$ with x=0.25 taken at 4 K and 130K. Black line indicates the intensity difference between the data at 4 K and at 130K.

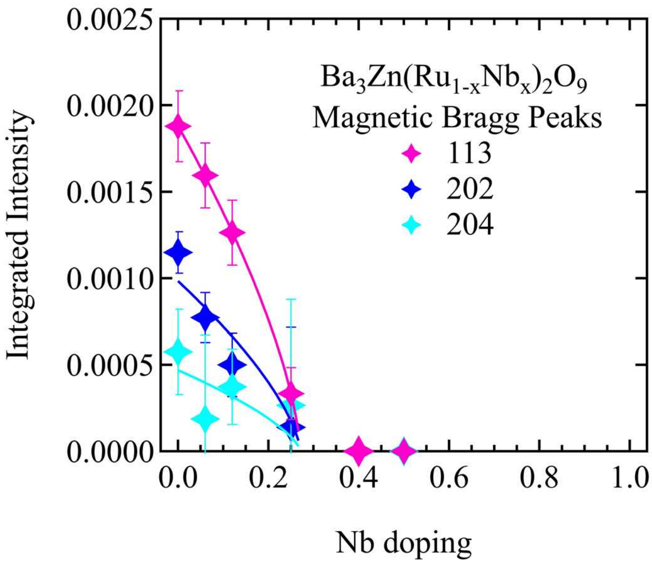

Fig. 2: The Nb concentration x-dependence of the magnetic Bragg intensities for $Ba_3Zn(Ru_{1-x}Nb_x)_2O_9$.

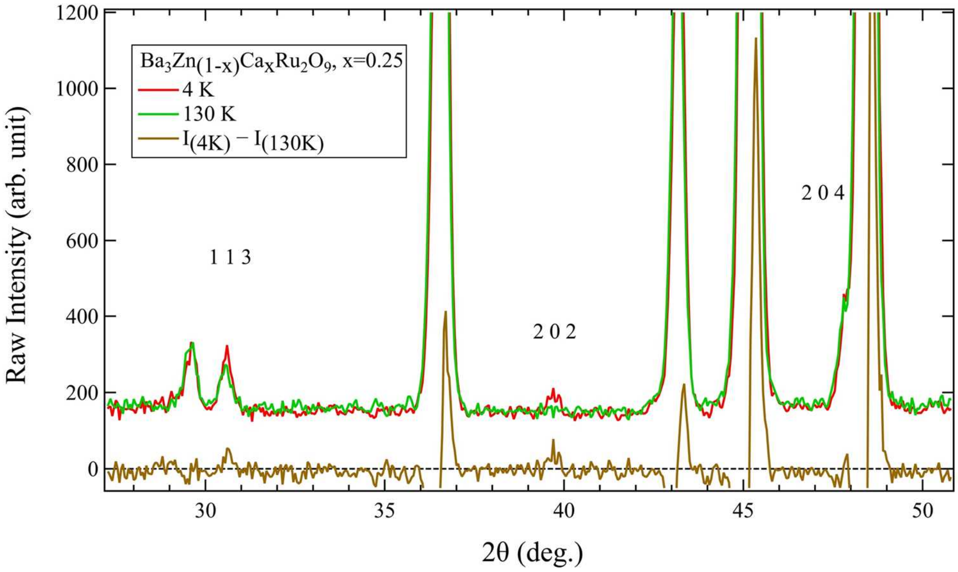

Fig. 3: Profiles of the neutron diffraction of $Ba_3Zn_{1-y}Ca_yRu_2O_9$ with y = 0.25 taken at 4 K and 130K. Brown line indicates the intensity difference between the data at 4 K and at 130K.

４．引用(参照)文献等

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