$Ba_3Zn_{1-x}Ca_xRu_2O_9$ の特異な磁気状態

Novel magnetic states of $Ba_3Zn_{1-x}Ca_xRu_2O_9$

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（概要）

We study on a 6H hexagonal ruthenates $Ba_3ZnRu_2O_9$ and related compounds, where $Ba_3ZnRu_2O_9$ is a candidate quantum spin liquid (QSL) system. Recently, we have found tiny magnetic Bragg intensity of $Ba_3ZnRu_2O_9$ at 10 K, indicating that the novel ground state has magnetic long-range ordered component with tiny-ordered moment. On the other hand, $Ba_3CaRu_2O_9$ has non-magnetic ground state, where paired spins in the Ru_2O_9 dimer form a spin singlet state with a spin gap (SG). By Ca-doping of $Ba_3ZnRu_2O_9$, we can control the magnetic ground state from novel QSL state to SG one. In order to clarify the Ca-doping effect to magnetic state, we carried out neutron powder diffraction of two samples $Ba_3Zn_{1-x}Ca_xRu_2O_9$ (x = 0.25 and 0.50) using HRPD at JRR-3. For x=0.25, the three magnetic Bragg peaks with tiny diffraction intensity, which is less than that of x=0. Moreover, no magnetic Bragg peak was observed for x=0.50. In order to clarify the x-dependence of magnetic Bragg peak and magnetic long-range ordering, we have carried out neutron powder diffraction of two samples $Ba_3Zn_{1-x}Ca_xRu_2O_9$ (x = 0.15 and 0.35) using HRPD at JRR-3. From the results, we found that the magnetic Bragg peaks vanish at $x_c \sim 0.35$.

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)] Recently, we have found tiny magnetic Bragg intensity of $Ba_3ZnRu_2O_9$ at 10 K, indicating that the novel ground state has magnetic long-range ordered component with tiny-ordered moment. 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).[3,4)] It is interesting in the magnetic behavior for mixing compounds $Ba_3Zn_{1-x}Ca_xRu_2O_9$. We carried out the neutron powder diffraction of two samples $Ba_3Zn_{1-x}Ca_xRu_2O_9$ (x = 0.25 and 0.50). In order to study the magnetic ground states of the Ru^{5+}-spin (S=3/2) of the Ca-doping systems $Ba_3Zn_{1-x}Ca_xRu_2O_9$, we planned to additional neutron powder diffraction experiments using HRPD of two samples $Ba_3Zn_{1-x}Ca_xRu_2O_9$ (x = 0.15 and 0.35).

２．方法

We measured the powder neutron diffraction profiles of two samples $Ba_3Zn_{1-x}Ca_xRu_2O_9$ (x = 0.15, and 0.35) 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 $Ba_3Zn_{1-x}Ca_xRu_2O_9$ with x = 0.15 taken at 10 K and 130K. Brown line of Fig. 1 indicates the intensity difference between the data at T = 10 K and at 130K. From the previous experiments, we observed three magnetic Bragg reflections 113, 202, and 204 for the non-doped sample $Ba_3Zn_{1-x}Ca_xRu_2O_9$ with x = 0. For x = 0.15, these magnetic Bragg intensities become less than that of x = 0. Figure 2 shows neutron diffraction profiles of $Ba_3Zn_{1-x}Ca_xRu_2O_9$ with x=0.35 taken at 10 K and 130K. We have observed no magnetic Bragg peak for x = 0.35. Figure 3 shows the Ca concentration x-dependence of the magnetic Bragg intensities for $Ba_3Zn_{1-x}Ca_xRu_2O_9$, indicating the intensities monotonically decrease with increasing x. Ca substitution for Zn does not change the fundamental magnetic structure, it just reduces the ordered moment of the

Ru^{5+}-spins. From fitting, the phase boundary of long-range ordered state is evaluated at $x_c = 0.35$. In the near phase boundary at $x_c = 0.35$, spin system is considered to be quantum spin liquid state.

In this time, we can see the tiny intensities of the magnetic reflections 113, 202, and 204. Since there are only three peaks, it is not possible to determine the magnetic structure without making any assumptions. Then, by referring to the magnetic structures of similar materials $Ba_3MRu_2O_9$ (M=Co, Ni, Cu, *etc*.), magnetic structure of $Ba_3ZnRu_2O_9$ has been found to have similar periodicity to that of $Ba_3NiRu_2O_9$[5]. The detailed magnetic structure of $Ba_3ZnRu_2O_9$ is currently under analysis.

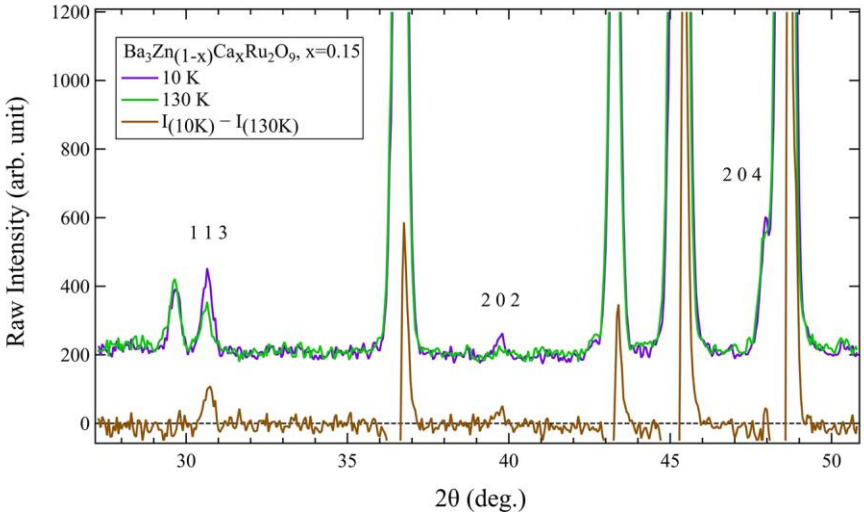

Fig. 1: Profiles of the neutron diffraction of $Ba_3Zn_{1-x}Ca_xRu_2O_9$ with x = 0.15 taken at T = 10 K and 130K. Brown line indicates the intensity difference between the data at 10K and at 130K.

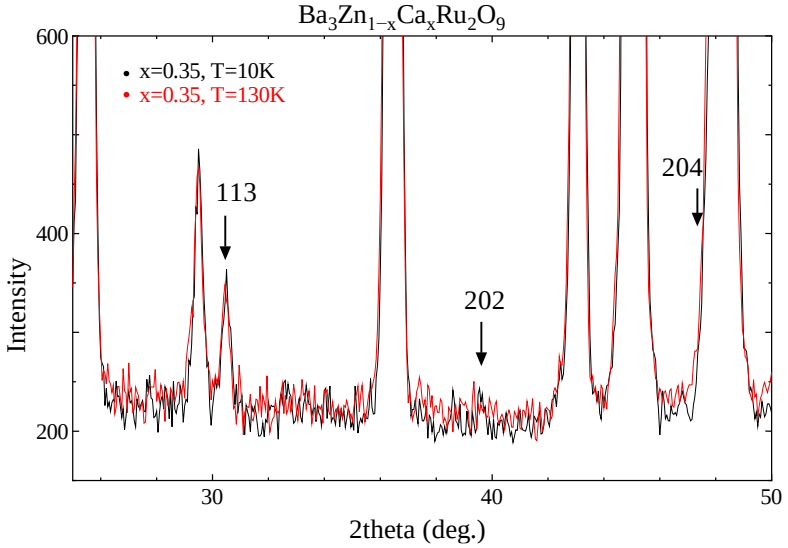

Fig. 2: Profiles of the neutron diffraction of $Ba_3Zn_{1-x}Ca_xRu_2O_9$ with x = 0.35 taken at 10 K and 130K.

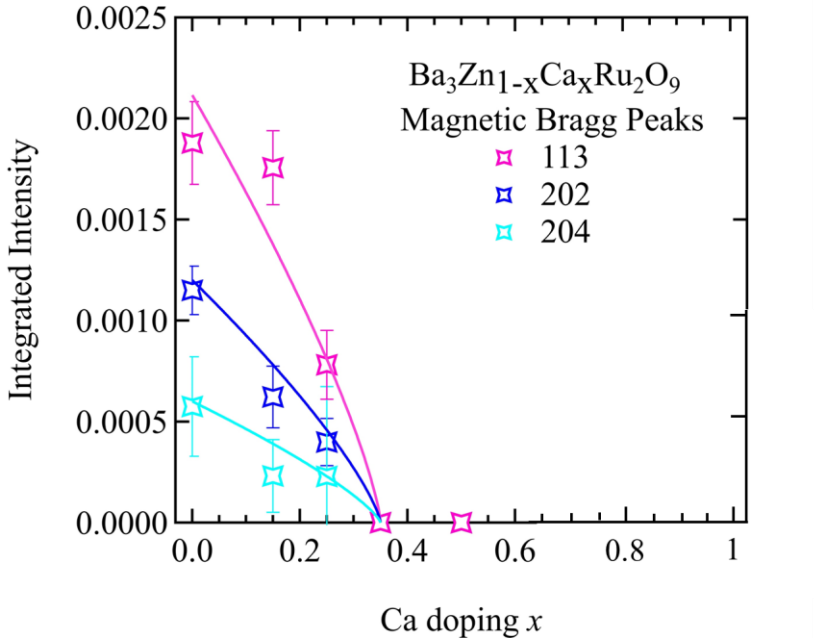

Fig. 3: The Ca concentration *x* - dependence of the magnetic Bragg intensities for $Ba_3Zn_{1-x}Ca_xRu_2O_9$.

４．引用(参照)文献等

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