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Magnetism of the antiferromagnetic spin-3/2 dimer compound $CrVMoO_7$ having an antiferromagnetically ordered state[†]

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In a magnetically ordered state, two types of magnetic excitations exist: gapless transverse-mode (Nambu-Goldstone mode) and gapped longitudinal-mode (amplitude Higgs mode) excitations. The former is well known as spin wave excitation. The longitudinal-mode (L-mode) was observed by inelastic neutron scattering experiments in the pressure-induced magnetically ordered state of $TlCuCl_3$, which is a three-dimensional (3D) interacting antiferromagnetic (AF) spin-1/2 dimer compound.[1–3] While the L-mode has weak intensity and spontaneously decays into a pair of transverse-modes, it is well defined in the ordered state in the vicinity of the quantum critical point for 3D systems.[4]

As for low-dimensional systems, it is difficult to observe the L-mode in longitudinal susceptibility by inelastic neutron scattering, since the longitudinal susceptibility exhibits an infrared singularity that can obscure the amplitude peak at a finite energy. In terms of scalar susceptibility, however, the L-mode can be well defined both in 2D and 3D systems. In magnetic systems, the L-mode was actually observed by Raman scattering experiments (which can measure the scalar susceptibility) in the pressure-induced ordered state of $KCuCl_3$ and in the magnetic-field-induced ordered state of $TlCuCl_3$.[5–7]

According to the results of theoretical investigations on interacting AF spin-cluster compounds, the L-mode excitations can be observed in the antiferromagnetically ordered state that appears on cooling under atmospheric pressure and zero magnetic field.[8] A shrinkage of the ordered magnetic moments by quantum fluctuations leads to a large intensity of the L-mode excitations. If the ground state of the isolated spin cluster is a spin-singlet state, the shrinkage of ordered moments can be expected in an ordered state that is generated by introducing intercluster interactions. We expect that an interacting AF spin-3/2 dimer model can be applied to the compound $CrVMoO_7$, judging from its crystal structure (Fig. 1).[9,10]

We studied the magnetic properties of $CrVMoO_7$ in powder form, using magnetization, specific heat, electron spin resonance, neutron diffraction, and inelastic neutron scattering measurements. An antiferromagnetically ordered state appeared below $T_N = 26.5 \pm 0.8$ K. The magnetic susceptibility at high temperatures was close to that calculated for the isolated AF spin-3/2 dimer with an intradimer interaction of $J = 25 \pm 1$ K and $g = 1.92 \pm 0.02$. We were able to explain the magnetization curves on the basis of the interacting AF spin-3/2 dimer model with an effective interdimer interaction of $J_{\rm eff} = 8.8 \pm 1$ K. The magnitude of the ordered moment was $0.73(2)\mu_B$, which is much smaller than the classical value of $\sim 3\mu_B$. Using inelastic neutron scattering measurements, the magnetic excitations were observed, and the results were qualitatively explained on the basis of the interacting AF spin-3/2 dimer model.

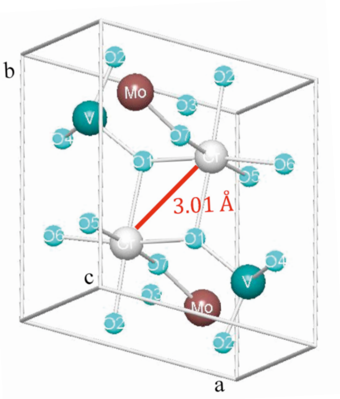

Fig. 1. The unit cell of $CrVMoO_7$. An AF spin-3/2 dimer is formed by two neighboring Cr^{3+} ions with a distance of 3.01 Å.

In conclusion, $CrVMoO_7$ is a rare spin-dimer compound that shows an antiferromagnetically ordered state under atmospheric pressure and zero magnetic field. Though we could not confirm the L-mode in this study, the magnetic excitations of L-mode would be observable in *single crystalline* $CrVMoO_7$.

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