# Successive Transitions in Spin-dimer Compound $Cs_3V_2Cl_9$

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$A_3M_2X_9$ (A = Cs, Rb : M = transition metal elements: X = Cl, Br) compounds with trigonal space group $P6_3/mmc$ are composed of isolated di-nuclear complexes $[M_2X_9]^{3-}$ and their magnetic properties are explained within an isolated or weakly coupled spin dimer model. Because these spin dimers are arranged in a triangular form, the spin frustration effect is expected to appear when magnetic phase transition occurs via finite interdimer interactions. $Cs_3V_2Cl_9$, one member of the $A_3M_2X_9$ family with magnetic ion $V^{3+}$ ($S = 1$), was previously studied via magnetic susceptibility and inelastic neutron scattering measurement using powder sample[1)] and no magnetic ordering was observed above 1.5 K. We recently synthesized a single crystal of $Cs_3V_2Cl_9$ and measured susceptibility $\chi(T)$ and specific heat, and we found successive phase transition at $T_N \approx 4$ K and $T_n \approx 15$ K.[2)]

These successive phase transitions are not explained within the framework of the isolated dimer model, and they show the presence of non-negligible interdimer interaction. $\chi(T)$ shows no anomaly at $T_n$ although $T_N$ is accompanied by the cusp-like anomaly of $\chi(T)$. The lower temperature transition is suggested to be an antiferromagnetic transition although the spin structure is not clear. The higher temperature transition is not a mere crystal structure transition because the transition temperature depends on the applied magnetic field.

We measured the $\mu$SR of $Cs_3V_2Cl_9$ to clarify the nature of the successive phase transitions. Figure 1 shows the zero-field muon spin relaxation (ZF-$\mu$SR) spectra measured down to 1.6 K. At relatively high temperatures, the spectra follow Gaussian curves. As the temperature decreases, the spectra changes from Gaussian to an exponential curve following $a_s \exp(-\lambda_s t) + a_f \exp(-\lambda_f t)$, where $\lambda_s$ and $\lambda_f$ denote slow and fast relaxation rates, and $a_s$ and $a_f$ represent amplitudes of the asymmetry of slow and fast components, respectively. Solid lines in Fig. 1 are fitted results. Figure 2 shows the temperature dependence of $\lambda_s$. A distinct peak is observed at around $T_N$, which confirms that a magnetic long range order occurs at this temperature. No anomaly of lambda is observed at $T_n$, which indicates that an internal field does not appear. The phase transition at $T_n$ is not accompanied by the internal field, whereas the value of $T_n$ depends on the applied magnetic field. One candidate for the transition at $T_n$ is a spin nematic order wherein quadrupole moments, not magnetic moments, play the role of an order parameter. Because the nematic state does not break the time-reversal symmetry, usual magnetic probes including muon do not detect this transition.[3)] Several theoretical studies indicate the occurrence of the nematic order in an $S = 1$ triangular lattice antiferromagnet or spin dimer magnets. The findings obtained by this $\mu$SR experiments make it more likely that $T_n$ is the nematic transition.

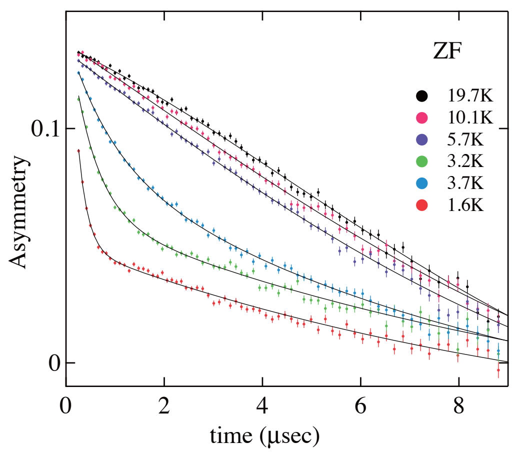


Fig. 1. Temperature dependence of the ZF-$\mu$SR spectra of $Cs_3V_2Cl_9$.

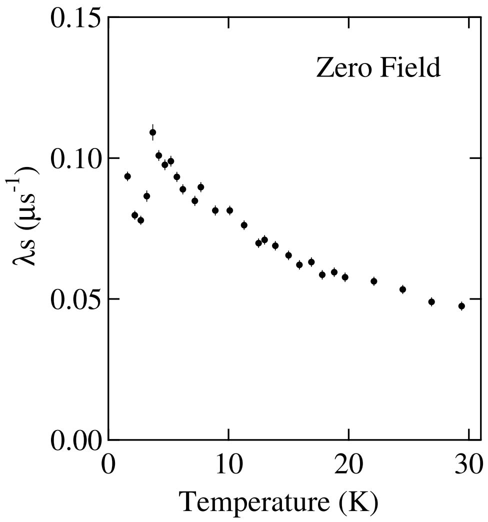


Fig. 2. Temperature dependence of the relaxation rate $\lambda_s$.

References
1) B. Leuenberger *et al.*, Inorg. Chem. **25**, 2930 (1986).
2) H. Kikuchi, T. Tanaka, Y. Fujii, A. Matsuo, K. Kindo, Abstract of the 19 International Conference on Magnetism. July 8–13, 2012 Busan, Korea, p. 167.
3) A. Smerald *et al.*, Phys. Rev. B **88**, 184430 (2013).

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