Modulated Magnetic Structure in the Rare-earth Clathrate $Eu_8Ga_{16}Ge_{30}$

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In the clathrate compound $Eu_8Ga_{16}Ge_{30}$, the guest magnetic ions of Eu^{2+} within the tetrakaidecahedral cages are rattling among off-center positions even within the ferromagnetically ordered state below T_C=36 K.[**?**, **?**] The resistivity and low-field magnetization for y=0 show broad humps at 24 K and 20 K, respectively.[**?**] These anomalies rapidly disappear upon substituting Si for Ge in $Eu_8Ga_{16}Ge_{30}$, implying the existence of multiple ferromagnetic structures only in the pure ternary compound. Furthermore, the Curie temperature T_C decreases from 36.2 K for $Eu_8Ga_{16}Ge_{30}$ to 32.6 K for $Eu_8Ga_{16}Ge_{22.3}Si_{7.3}$, and in conjunction with this decrease, the jump of the specific heat at T_C is doubled. These observations suggest that a delicate modulated magnetic structure in pure $Eu_8Ga_{16}Ge_{30}$ is unstable against Si substitution and changes to a more robust and uniform ferromagnetic state with Si substitution.

In the present work, we performed neutron diffraction measurements on a single-crystalline sample $Eu_8Ga_{16}Ge_{30}$ prepared with ^{153}Eu isotope for avoiding strong absorption of neutrons. Figure **??** shows the temperature dependence of the integrated intensity (left-hand scale) and the peak width (right-hand scale) of the 022 peak. Above T_C, the 022 peak was observed because of the nuclear contribution. Below T_C, the intensity start increasing, and the peak width increases at around T_C and slowly decreases. These are attributed to development of ordered magnetic moments below T_C, being consistent with the results of the magnetization measurements. At around $T^*\sim$20 K, the intensity shows hump, and the peak width reaches a minimum value. Further cooling below T^*, the intensity steeply increases, and the peak width first increases, and then decreases. These complex behaviors at around T^* can not be explained by a simple ferromangetic structure. We now analyses the $\boldsymbol{q}$-dependence of the intensity of the magnetic contribution in order to confirm magnetic structures at the intermediate range of $T^*<T<T_C$ and at the ground state of $T<T^*$.

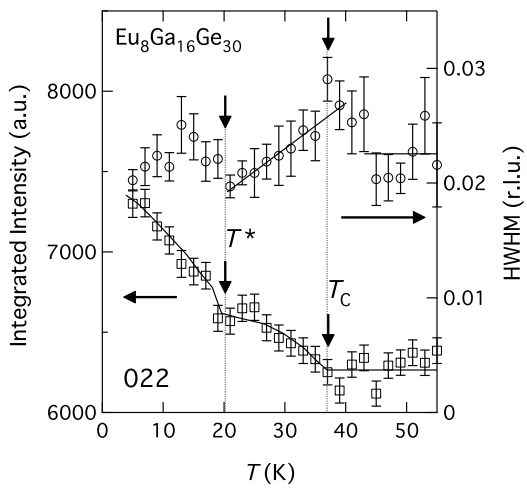

Fig. 1. Temperature dependence of the integrated intensity and the peak width of the peak at $\boldsymbol{Q}$=(022).