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^{121,123}Sb-NQR Investigation for Possible Ferromagnetic Ordering in Alkaline-Earth Filled-Skutterudite: $SrFe_4Sb_{12}$

M. Matsumura[1], H. Tou[2], E. Matsuoka[2] , T. Takabatake[2] and M. Sera[2]

[1]Department of Material Science, Faculty of Science, Kochi University, Kochi, 780-8520
[2]Department of Quantum Matter, ADSM, Hiroshima University, Higashi-Hiroshima, 739-8530

^{121,123}Sb-NQR spectrum and the nuclear spin-lattice relaxation time T_1 have been measured in the range of temperature from 5 K to 300 K in the Alkaline-Earth Filled-Skutterudite, $SrFe_4Sb_{12}$, to investigate microscopically a possible ferromagnetic order recently reported in the macroscopic measurements. The Curie-Weiss (CW) law of the magnetic susceptibility at $T > 150$ K with the effective moment μ_{eff} $\sim$2.87 μ_B/f.u. and the positive Weiss temperature θ $\sim$50 K and, moreover, the small magnetic hysteresis and the small magnetization of 0.4 μ_B/f.u. at 5 T at $T \ll \theta$ suggest an itinerant weak ferromagnetic order occurring in this compound.

The narrow three NQR lines for ^{123}Sb (nuclear spin I=7/2) and the two NQR lines for ^{121}Sb (I=5/2) were observed in a whole range of temperature, indicating only one Sb site existing in the crystal at least in a view of NQR. The quadrupole frequency ν_Q and the asymmetry parameter η are successfully estimated from the ratio of the respective highest NQR lines, $^{121}\nu_{\rm NQR}/^{123}\nu_{\rm NQR}$. The value of ν_Q decreases monotonically with increasing temperature and η is 0.39 being independent of temperature, which implies no changes of the crystal symmetry in the temperature range of measurements. In the sense of the static NQR spectrum, a clear hyperfine splitting was not observed at $T \ll \theta$ except for the additional increase of the linewidth of about 5 KHz below 60 K being close to θ in the FFT of echo signal. This suggests two cases; too small coupling between ferromagnetic moments and Sb nucleus to make a clear splitting, or an existence of substantial part of the specimen showing no ferromagnetic order.

The nuclear relaxation rate $1/T_1T$, which is confirmed to be dominated by the magnetic fluctuations, shows a broad peak at around 60 K. Above about 100 K, $1/T_1T$ is found to follow a function, $1/T_1T(\mathrm{secK})^{-1}$=0.66+49.87/($T$-36.77), where the first and the second terms are interpreted as a relaxation due to the orbital current of 5p electrons at Sb site ($(1/T_1T)_{orb}$), and as a relaxation due to spin fluctuations of conduction electrons ($(1/T_1T)_{spin}$), respectively. The following CW law of $(1/T_1T)_{spin}$ is consistent with the T-dependence of the static susceptibility, implying a dominant ferromagnetic spin fluctuations in this compound. Below 100 K, $(1/T_1T)_{spin}$ deviates downward from CW law as like the deviation of χ. Since the relation $(1/T_1T)_{spin} \propto \chi$ is hold close to the peak temperature 60 K, the suppression of $1/T_1T$ is ascribed to that of χ. The origin of the suppression is not clear at present.

The rapid decrease of $1/T_1T$ below 60 K is usually ascribed to a development of the ferromagnetic order, however, it may be too early to judge an existence of a homogeneous ferromagnetic order because of the lack of the hyperfine splitting in the NQR spectrum. There exists another possibility for the decrease of $1/T_1T$ if we assume an inhomogeneous ferromagnet; a small part with ferromagnetic order and a large part still in a exchange enhanced paramagnetic state with a broad peak of χ around 60 K. The broad peak of χ would bring about the decrease of $1/T_1T$ if the relation $(1/T_1T)_{spin} \propto \chi$ would hold below 60 K.

The nuclear relaxation rate following CW law with $\theta > 0$ not following Korringa relation suggests that ferromagnetic spin fluctuations is enhanced in this compound. The long range ferromagnetic order is still under discussion at present because of the lack of a clear hyperfine splitting in NQR spectrum.