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Spin form factor study of $GdAl_2$

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Recently, we have developed the x-ray diffraction method to obtain the atomic form factors of the electron spin polarization in ferromagnetic crystals[1-3]. Since given by this method is information about the electron spin polarization rather than the magnetization, the obtained spin form factors for the rare earths or the actinides having large orbital magnetic moments should be very different in shape from the neutron magnetic form factors. This point will be advantageous when the experiments are combined with the theoretical electronic structure calculations, where the convincing prescription for the treatment of the orbital magnetic moment has not been established yet.

The cubic Laves-phase compounds $R\mathrm{Al}_2$ (R = rare earth) are ferromagnetic for most of rare earths and suitable for the present spin form factor study. We have applied this method to $GdAl_2$. This study has a twofold meaning. First, this was done as part of our systematic study on the spin density distribution around the rare-earth ions. From this point of view, the gadolinium ion is a key one, because the spin polarization of the localized electrons is expected to be almost isotropic and the experimental spin form factors can be compared with $\langle j_0 \rangle$, the numerical expectation value of the 0th-order spherical Bessel function with respect to the theoretical *4f* radial wave function. Second, according to the previous research reports, the magnetic properties of $GdAl_2$ are not necessarily as understandable as expected, and we thought that the findings from our spin form factor study could contribute to a better understanding of this material.

The polarized neutron study of $GdAl_2$ has been done by Abell *et al.* in the early 1980's and the Gd magnetic form factors at 4.2K have been reported[4]. The polarized neutron sees the orbital magnetic moment together, if any, and detects the magnetic moment perpendicular to the scattering plane. In our experiment, on the other hand, the probe photon sees the spin polarization parallel to the scattering vector. The magnetic parts of the atomic form factors determined from these two experiments will be identical when they arise solely from the spin polarization and the spin density distribution does not depend on the direction of the polarization.

The experiments were done at the station 3C3 with a bending-magnet light source. The single-crystal samples were prepared by the Bridgeman method at the Institute for Solid State Physics, the University of Tokyo, and, prior to the measurements, the magnetic properties were checked by a SQUID magnetometer at the Cryogenic Center, the University of Tokyo. We measured three series of reflections of (h00), (hh0), and (hhh) at several temperatures. The details of the experiments are the same as described in Ref. 3.

The spin form factors in medium through high $\sin\theta/\lambda$ region were obtained and found to change in roughly accordance with $\langle j_0 \rangle$ for the free Gd^{3+} ion calculated with the Hartree-Fock method or the density-functional-theory one for each series of reflections. The values of the form factors multiplied by the spin magnetic moment were, however, lower in comparison with those estimated from the magnitude of the magnetization at the same temperature. Similar phenomenon has been also reported in the neutron case[4], but the reduction rate seems larger in our x-ray case. Moreover, we found that the thermal variation for the (hhh) series exhibits a broad maximum at around 50 through 100K (Fig. 1), while the magnetization varies monotonically in the ferromagnetically ordered region ($T \leq T_C$~170K). Such a phenomenon was not observed for the (h00) and (hh0) series.

The behavior described above might be related to the incomprehensible results found in some previous studies of this compound.

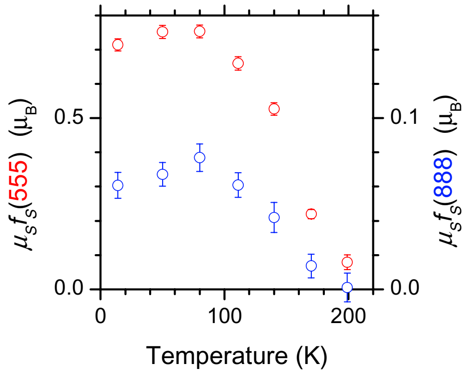

Fig. 1 The temperature dependence of the values of the spin form factors, f_S, multiplied by the spin magnetic moment, μ_S, for the (555) and (888) reflections of $GdAl_2$, which are marked with red and blue circles, respectively.

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