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Evidence for positive polarity of the spin moment in hcp Sm determined from a magnetic Compton-scattering experiment

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The magnetic moment of Sm^{3+} in hcp Sm was studied with a magnetic Compton-scattering experiment with the aid of a pulse magnet. We have successfully obtained direct evidence for a positive contribution of the spin moment to the total moment, opposite to the case for a free ion. The enhancement of the spin moment by incorporation of the conduction electron polarization is observed. [S0163-1829(97)52134-5]

It is well known that the magnetic moment of a rare-earth ion originates mainly from the localized $4f$ electrons and that its magnitude is not sensitive to the environment of the ion. However, this is not true for Sm^{3+}, where the narrow energy intervals among the consecutive J multiplets and the small value of the $4f$ moment resulting from almost cancellation between the spin part and the orbital one make a drastic change of the $4f$ moment in magnitude and character possible. Such a modification could be caused in solids by the exchange interaction and the crystal fields, both of which induce a relatively strong admixture of the excited J levels into the ground one through the off-J matrix elements of the Hamiltonian. In metallic cases, furthermore, the conduction electron spin polarization, whose magnitude could be same order as that of the $4f$ moment in the case of Sm^{3+}, would have a considerable influence on the magnetic properties. These effects have been studied so far by several scientists, but in interpreting the experimental results it is difficult to take account of all these effects simultaneously and to make it clear how each effect works.

We found before that hcp Sm, which is a metastable phase prepared by quenching from the melt using a single roller apparatus, is a rather unusual ferromagnet with an ordering temperature T_c of 160 K, a quite small magnetization of the order of $0.1\mu_B$, the abnormal thermomagnetic curve having a broad maximum, and so on.[1] Recently, we have analyzed the temperature dependence of the total moment based on a model with a trivalent ion in a metallic matrix considering the lowest three multiplets, the spin-orbit interaction, the crystal fields, the exchange interaction, and the conduction electron spin polarization, and it has been concluded that the effect of the conduction electron polarization is crucial to the peculiar magnetic behaviors of this metal.[2] One of the interesting results of this analysis is that in hcp Sm the spin part of the magnetic moment, which is enhanced by the conduction electron spin polarization, exceeds the orbital part of it. In this case, the spin moment is expected to be parallel to the total moment and the orbital one to be antiparallel, namely the situation is opposite to the case for the free Sm^{3+} ion. To verify this interpretation from a microscopic point of view, we have then made a magnetic Compton-scattering (MCS) experiment.

The magnetic Compton profile (MCP) $J_{\mathrm{mag}}(p_z)$, where p_z represents the z component of the electron momentum and z means the direction of the scattering vector of x rays, is the spin-dependent part of the so-called Compton profile. Using the incident x rays with a component of a circular polarization, this is experimentally obtained from the difference between the energy spectrum of Compton scattered x rays from the sample magnetized along the z axis and that from the sample magnetized in the opposite direction along the same axis. This technique is quite powerful for examining the complicated origin of the Sm^{3+} moment in metals due to the striking features of MCP (Ref. 3) described below. First, only the spin part of the magnetic moment is reflected in an MCP. This feature enables us to determine whether the spin moment is parallel or opposed to the total moment, i.e., the sign of an MCP can be an index for the polarity of the spin moment compared to that of the resultant magnetization.[4] Second, since the MCP gives the momentum distribution of spin polarized electrons, the contribution of the conduction electrons is clearly distinguishable from that of the $4f$ electrons; the former are distributed narrowly in the low momentum region, whereas the latter have a broad Compton profile, rather flat in the low momentum region. Up until now, microscopic information on the Sm^{3+} moment in several metallic substances has been obtained through the neutron form factor[5] with great efforts to solve the problem of the high thermal-neutron capture cross section of natural Sm, but it is to be noted that in the above sense MCS is suitable to the aim of this study and that in the case of a polycrystalline sample, such as hcp Sm, the available method is nothing but MCS.

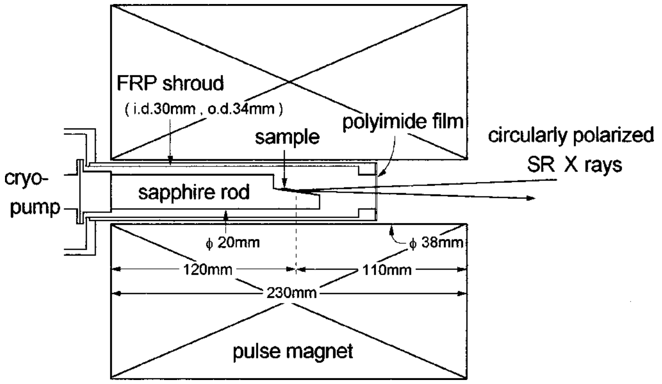

FIG. 1. Schematic arrangement around the sample. The dimension of the target, made of an array of cut ribbon samples, is about 3 cm in length, 8 mm in width, and 0.14 mm in thickness. The x rays are impinged with an angle of about 6° from the surface of the target.

The experiment was performed using circularly polarized synchrotron radiation (SR) x rays emitted from the elliptical multipole wiggler[6] installed at the 6.5-GeV TRISTAN accumulation ring of the National Laboratory for High Energy Physics. The incident x rays with an energy of 69.5 keV were focused on the sample by a doubly bent Si(111) monochromator[7] and the energy spectra of the scattered x rays were measured using a multisegmented Ge solid-state detector.[8,9] The scattering angle was about 160°. The data were collected in the sequence of $ABBAABBA\ldots$, where $A(B)$ means the case that the magnetization is (anti)parallel to the scattering vector and each of them took 20 or 30 s. To overcome the magnetic hardness of the sample, pulsed magnetic fields of 10 T were used[10] to reverse the magnetization of the sample set on the inclined plane of the tip of a sapphire holder (Fig. 1). In addition, the measurement was done at 150 K, which is 10 K below the ordering temperature. Note that the sample holder must be made of a single crystal of an insulator which has a high thermal conductivity, because a yield of eddy current due to a pulsed magnetic field of 10 T for a few milliseconds might cause a movement or a heat-up of the holder. For the present setup, consequently, just 2 s are enough for the interval time between A and B. The spin dependence of the scattering intensity was obtained as the difference between the data of A and those of B, which were normalized before subtracting one from the other by the intensity of the elastic scattering or that of the Sm K emissions.

Figure 2 shows the obtained MCP. The total accumulation time is about 18 h for each direction of the magnetization, the evaluated momentum resolution is 0.80 atomic units (a.u.), and the statistical accuracy is indicated by the error bars. The solid and open circles mean the cases, respectively, where the elastic peak and the fluorescence peaks are used in the data normalization. The nearly same profiles in both cases ensure that the profile intrinsic for the sample was successfully detected.

The positive values of the obtained MCP indicate the positive contribution of the spin moment to the total moment. We have also confirmed this result by comparing with the MCP of a $SmCo_5$ standard sample, also observed on the positive side, arising mainly from the spin moment of the Co ion positively contributing to the resultant magnetization. The polarity of the spin moment in hcp Sm elucidated by the present measurement is unusual for the light rare earths, but consistent with our interpretation of the magnetic data.[2]

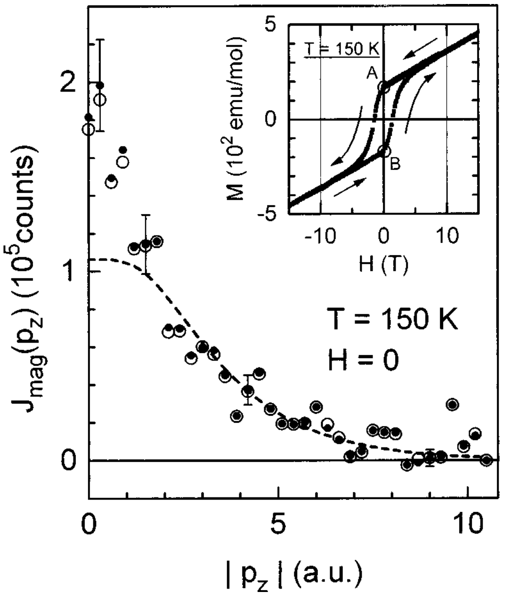

FIG. 2. The spin-dependent Compton profile of hcp Sm. The solid and open circles mean the cases, respectively, where the elastic peak and the fluorescence peaks are used in the data normalization. The broken line is the relativistic Hartree-Fock Compton profile of atomic $4f$ electrons (Ref. 11). The inset shows the field dependence of the magnetization of the sample prepared the same way, which was measured by a vibrating sample magnetometer with a water-cooled electromagnet at the High Field Laboratory for Superconducting Materials, Tohoku University. The hysteresis shows that a magnetic field of 10 T is enough to reverse the magnetization at 150 K. The points A and B indicate the measuring positions of the Compton profiles.

To separate the conduction electron contribution in the low-momentum region, we have fitted the experimental data in the region of $|p_z|>3$ a.u. with the relativistic Hartree-Fock Compton profile[11] for the $4f$ electrons (a broken line in Fig. 2) and attributed the residual part to the conduction electron contribution. As is shown in Fig. 2, a good fit was obtained in the region of $|p_z|>2$ a.u., similar to the MCP of the Gd metal.[9] Based on the fact that the area of MCP is proportional to the magnitude of the spin moment, the ratio of the magnitude of the conduction electron polarization to that of the $4f$ spin moment is estimated to be $17\pm3\%$, also giving a good agreement with that from the magnetic analyses of 15–16%.[2,12]

In conclusion, we have applied MCS to the microscopic study of the Sm^{3+} moment. Observing the spin part in isolation from the total moment, it was verified that in hcp Sm the spin moment is enlarged by the appreciable incorporation of the conduction electron spin polarization and contributes positively to the total moment, contrary to the case of the free Sm^{3+} ion. Furthermore we have successfully demonstrated the availability of a pulse magnet in the MCS experiment in the low-temperature region by the present experiment. This means that many ferro- or ferrimagnets with large coercivities at low temperatures, such as rare-earth intermetallics, have now become targets of the MCS study.

For the ferromagnetic intermetallics of Sm^{3+} with nonmagnetic ions in general, the cancellation between the magnetizations arising from the spin and the orbital part of the $4f$ moment of each Sm^{3+} ion, which have different temperature dependences,[2] results in the peculiar magnetism. In addition, the conduction electron polarization can affect the resultant magnetization significantly. Measurements of the $4f$ spin, orbital, and the diffuse moment separately utilizing SR x rays are quite helpful to the understanding of the magnetic properties of these compounds and will be interesting topics in near future. The MCS experiment will be undoubtedly one of the powerful methods.

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[4] Since an MCP is the difference between the normal Compton profiles in two cases, it takes positive or negative values naturally, which we call here ''the sign.'' Needless to say, since the absolute sign of an MCP depends on the definition, i.e., which profile is subtracted from the other one, the signs of MCP's to be compared with each other have to be determined in a same fashion.

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[11] F. Biggs, L. B. Mendelsohn, and J. B. Mann, At. Data Nucl. Data Tables **16**, 201 (1975). The profiles tabulated therein are calculated for atoms, where the electronic configuration of Sm is $(4f)^6$ rather than $(4f)^5$ for the tripositve ion. However, this is probably not a serious problem since the difference between the $4f$ profile calculated for the Sm atom and that for Pm, which has five electrons in the $4f$ orbital and the nuclear charges smaller by 1, is very small.

[12] In fact, the value of this ratio depends on the fitting momentum range, e.g., 23 ± 3 % for $|p_z|>2.5$ and 26 ± 3 % for $|p_z|>2$. The cause of this ambiguity is insufficient statistical accuracy due to the fact that the present measurement had to be done just below the ordering temperature, where the thermal average of the spin moment effective to the MCP is small. Considering these values, the estimated ratio seems somewhat higher than that obtained from the magnetic analyses in Ref. 2.