Zero-Magnetization Ferromagnet Proven by Helicity-Switching Compton Scattering

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A single crystal of gadolinium-doped $SmAl_2$ has zero magnetization in the midst of the ordered temperature region, despite the probable ferromagnetic spin ordering. The asymmetry in Compton-scattering intensity when switching between right- and left-handed polarization of incident 150-keV synchrotron radiation provides decisive proof that ferromagnetic order is really there, and that spin and orbital magnetic contributions cancel. The experiments also show that the spin direction at this zero-magnetization state is rather stable against the external magnetic field and, nevertheless, reversible by a preceding control of temperature and an external field.

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The word "ferromagnet" has been used for material having a bulk magnetization in general, for the historical reason that the oldest ones comprise ferrous ions. Nowadays, magnetic materials have been classified, depending on the interionic spin arrangement, as ferromagnetic, antiferromagnetic, ferrimagnetic, and so on, where "ferro" means the simple parallel coupling between the spins. Since the spin of an electron is accompanied by a magnetic moment, the ferromagnet thus defined usually implies a macroscopic magnetization too, and hence not inconsistent with the former usage. However, a ferromagnet whose magnetization arises mainly from one of the rare-earth ions, Sm^{3+}, can exhibit a ferromagnetic spin order with no net magnetization at a specific temperature T_{comp} in theory, due to the exact compensation between the partial magnetizations originating from electron's spin and its orbital motion [1,2]. This unique property is thought to be one of the ideal characters for the devices processing the spin of a charged particle, because, despite the uniform spin polarization, it does not generate a stray magnetic field which might perturb the motion of a charged particle or its spin state through the Lorentz force, the magnetic force, the Larmor precession, and so on. The present kind of material may, then, find application in the fields, where the quantum-mechanical exchange interaction rather than the classical electromagnetic one plays an essential role, such as spin electronics manipulating not only the electric current but also the spin polarization. From such a point of view, as a matter of practical importance, the controllability of T_{comp} has been demonstrated by the magnetic measurements for the polycrystalline samples of ferromagnetic pseudobinary compounds $(Sm_{1-x}R_x)Al_2$, where R is rare earth [3].

The peculiar magnetism of ferromagnetic Sm compounds has been interpreted so far in terms of the different temperature dependences of the spin and orbital polarizations almost canceling each other, and, as far as the resultant macroscopic magnetization is mainly concerned, no conflict between the experiment and the theory has been encountered [1–4]. It is, however, also natural that some people should have suspicions about the real existence of such a ferromagnetic spin ordering just at the zero-magnetization state, due to lack of the direct observation. Considering, in addition, that the potential technological applications as mentioned above are essentially based on this point, the establishment of firm evidence is certainly desirable. But how can we prove it? Probably, the most hopeful way is to utilize the interaction between the electron's spin and the helicity of light. In this sort of interaction, the spin or the magnetization reversal in magnetic materials and a change between the right-circular and left-circular polarization of the probe light often result in an equivalent effect. Therefore, we can check and see the existence of the electron spin polarization in the target by switching the helicity of light while keeping the direction of electron's spin intact. Note that the spin direction in a material having no spontaneous magnetization could not be flipped as we want by an external magnetic field. The relevant photon-electron interactions include the Kerr effect, the magnetic circular dichroism, the spin-resolved photoemission, and so forth, but the most immediate and suitable means for the present purpose is thought to be the so-called magnetic Compton scattering, where the observed effect is directly related to the wave function of the polarized electrons and proportional to the spin part of the magnetization in disregard to the orbital one [5]. In this Letter, we report that such a magnetic Compton-scattering experiment with the state-of-the-art synchrotron-radiation technology has admirably revealed the spin ordering in question that had been conjectured.

In the present experiment, a single-crystal sample of $(Sm_{0.976}Gd_{0.024})Al_2$, whose ferromagnetic nature seems unquestionable considering the properties of the relevant system family [3,6,7], was used with the intention of excluding the ambiguity of the polycrystalline effect. The

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sample was grown in a sealed tungsten crucible by the Bridgeman method. Figure 1 shows the results of the magnetic measurements, which were made using a SQUID magnetometer. The compensation temperature T_{comp} is observed to be 67.5 K, and the magnetic order-disorder transition appears to occur at $T_{\mathrm{c}} \sim 125$ K. As shown in Figs. 1(b) and 1(c), the field strength required to reverse the magnetization becomes larger as the temperature approaches T_{comp}. This is considered to be a natural consequence of the magnetic inertness around T_{comp}, due to the marginal compensation between the spin and orbital magnetizations, and suggests the stability of the spin direction against the external magnetic field in that temperature range, if the ordering persists. As will be briefly discussed in the last part of this report, such an irresponsiveness of the spin direction might be useful for a spin-distinction function in the future application. The temperature dependence denoted by crosses in Fig. 1(a) is the one measured under *no* magnetic field throughout with rising temperature, after the sample was once cooled down to 2 K in a field of 5 T. The smooth crossing over a zero line at T_{comp} implies that the information about the spin direction should not be lost when the system goes through the zero-magnetization region. This result can be regarded as a collateral evidence for the persistence of the spin ordering at T_{comp}.

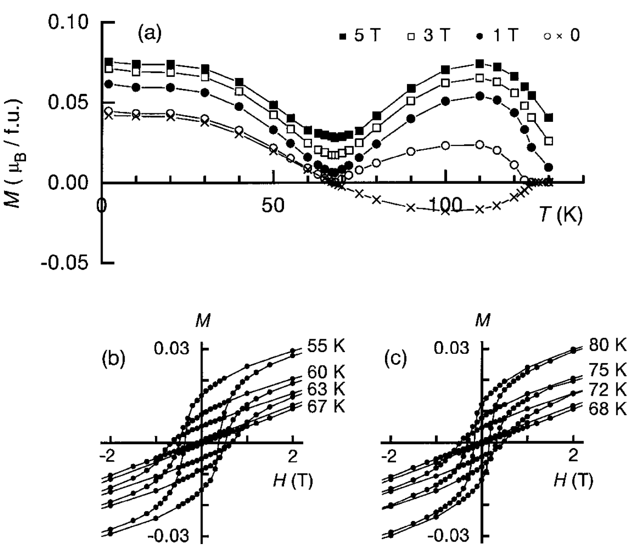

FIG. 1. Magnetic properties of a single crystal of $(Sm_{0.976}Gd_{0.024})Al_2$. The magnetization readings (M; f.u., formula unit) are ones along the easy axis of $\langle 111 \rangle$ when a magnetic field is applied in that direction. The upper figure (a) shows the magnetization versus temperature. Data points denoted by solid squares, open squares, solid circles, and open circles are taken from the first quarter of the magnetization versus applied field hysteresis loop at each temperature measured with the field changing in a sequence of $+5\ \mathrm{T} \rightarrow 0 \rightarrow -5\ \mathrm{T} \rightarrow 0 \rightarrow +5\ \mathrm{T}$. Crosses denote the temperature dependence of the remanent magnetization measured under a zero field throughout (see the text). The lower figures (b) and (c) show the hysteresis curves against the applied field in the vicinity of T_{comp}.

The "magnetic Compton" means a fraction of the relativistic effect in the Compton-scattering process, and a sufficient flux of circularly polarized high-energy x rays is needed for the measurement. In the present case, moreover, the degree of circular polarization has to be switchable as described before. These requirements have been basically met with the optical system of the synchrotron-radiation beam line NE1 of PF-AR (Photon Factory–Advanced Ring). The combination of the 6.5 GeV storage ring and the light-source device, called the elliptical multipole wiggler, can generate intense high-energy x rays of right or left elliptical polarization [8], and the brand-new doubly bent monochromator with the annealed Si(400) crystal can reflect and focus an adequate flux of a monochromated beam of up to 160 keV onto a sample [9]. Since the magnetic Compton experiments to date have been usually made with a short-period alternation of the magnetic field direction instead of the helicity of light for some practical reasons, we have examined, as a first step, the feasibility of the helicity-switching experiment with the 150-keV x rays. And then, after a couple of tests and technical solutions, we have eventually established the method with the helicity switching at long intervals of a few hours [10].

Figure 2 shows the magnetic effects in Compton profiles measured in both ways of the field and helicity switchings, each of which is obtained as the difference between the profiles for two directions of the magnetic field or the x-ray helicity. The results for Fe with the characteristic dip in the low-momentum region [5] imply the correct data taking for both methods. The obtained asymmetric ratios $A \equiv (I_{\mathrm{e}} - I_{\mathrm{o}})/(I_{\mathrm{e}} + I_{\mathrm{o}})$ were both about $+0.8\%$, where I_{e} (I_{o}) means the Compton-scattering intensity, normalized by the elastic one, under the even (odd) relation between the applied field direction and the x-ray helicity. Setting a single-crystal target of $(Sm_{0.976}Gd_{0.024})Al_2$ (ca. $\phi 10$ mm $\times$ 8 mm) such that the direction of easy magnetization, $\langle 111 \rangle$, should be parallel to the applied field and also coincide with the x-ray scattering vector, we confirmed again that the present helicity-switching method should give the same result as the conventional field-switching one at 8 K [Fig. 2(b)], where the sample has a finite magnetization. Note that the polarity of the magnetic effect has changed from the case of Fe, giving an agreement with the interpretation that, for $(Sm_{0.976}Gd_{0.024})Al_2$, the orbital magnetization exceeds the spin one and the latter orients opposite to the direction of the resultant magnetization or the applied field well below T_{comp}. The shape of the profile could be, roughly speaking, interpreted as the summation of the broad component of the $4f$ electrons and the conduction-electron contribution in low-momentum region. Slight difference from the Hartree-Fock profile shown in the tail part might be ascribed to the anisotropic effect.

Figure 3 shows two magnetic Compton profiles taken at T_{comp} with the helicity switching; one is the spectrum

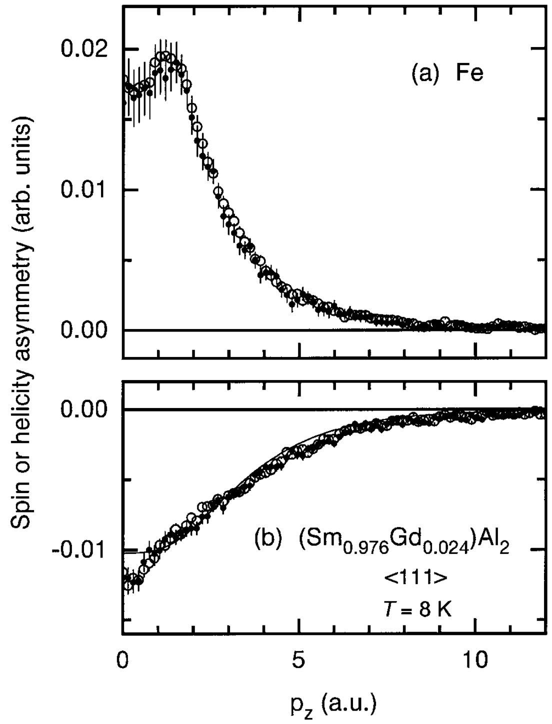

FIG. 2. Magnetic Compton profiles taken by the field- and helicity-switching methods, which are denoted by dots and open circles, respectively. The field-switching profiles were measured by alternating magnetic fields of ± 2 T every 30 sec while keeping the x-ray helicity unchanged. The helicity-switching ones were measured under a constant field of $+2$ T with reversing the x-ray helicity at intervals of 2 h (on the occasion of refilling the ring current). The values of the asymmetric ratio A of the field- and helicity-switching data are $+0.78\%$ and $+0.82\%$ for Fe and-0.59% and -0.58% for $(Sm_{0.976}Gd_{0.024})Al_2$, respectively. Scattering angle is 160°. Error bars show the statistical accuracies. A solid line shown in (b) is the relativistic Hartree-Fock profile for the $4f$ electrons of the free Sm atom [11] arbitrarily scaled for comparison, convoluted with the experimental resolution of 0.7 a.u.

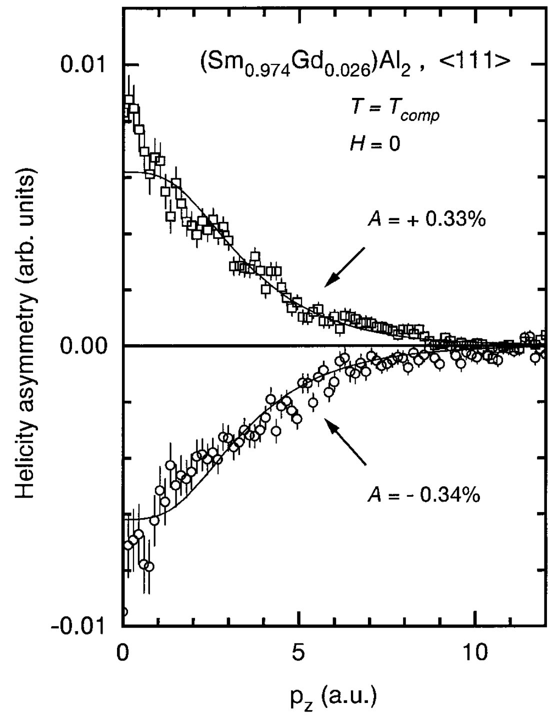

FIG. 3. Magnetic Compton profiles taken at T_{comp} by the helicity-switching method with no applied magnetic field. Two profiles of opposite polarities are different in the temperature and magnetic field process prior to the measurements (see the text). Solid curves are the relativistic Hartree-Fock result for the free Sm atom [11].

measured after the magnetic field is once applied parallel to the x-ray scattering vector at a temperature *below* T_{comp} and then the temperature approaches T_{comp} from the *low* temperature side as the field is diminished to 0 (circles), and the other is the spectrum measured after the field is applied parallel to the x-ray scattering vector again at a temperature *above* T_{comp} and the temperature is similarly set for T_{comp} from the *high* temperature side (squares). The obtained magnetic effects clearly show that the spin moments retain their ferromagnetic ordering just at the zero-magnetization state, at least during the several-hour measurements. Besides, the two spectra of opposite polarities imply that the spin direction at T_{comp} can be reversed by the preceding control of temperature and an external magnetic field, making use of the property that the spin magnetization contributes positively and negatively to the total one at above and below T_{comp}, respectively.

This asymmetry does not seem, furthermore, affected very much by the magnetic field, as long as the temperature is kept T_{comp}; $A = -0.33\%$ for $+0.5$ T and $A = -0.28\%$ for -0.5 T, whereas $A = -0.34\%$ for a zero field. This result is consistent with the magnetization versus field behavior shown in Fig. 1. The magnetic hardness or inertness of the spin direction at T_{comp} in addition to the persistence of the ordering reminds us of the potential application of this material in spin technology again. In order to detect the spin-dependent effect properly, in many cases, we have to extract the difference between the phenomena with two possible spin states in either probe or object substance, as has been done in the present Compton-scattering experiments. This difference technique is actually useful, for instance, to distinguish the spin image from the topographical one in the so-called spin-polarized scanning tunneling microscopy. With a probe tip made of the present kind of ferromagnet, it might be rather easy to reverse only the sample spins by an external field, while the probe spins remain unchanged, and to get the contrast ascribed purely to the surface spin structure. The ferromagnetic spin order at T_{comp} could, thus, materialize a unique spin-sensitive, nonmagnetic device, and also the microfabrication of this material together with a soft-magnetic one or a superconducting one may offer a novel function. Now that the existence of a zero-magnetization ferromagnet has been established, it may be the time when an actual realization of such a spin device or a comparison of its performance with that of the existing one should become a subject of not only the scientists but also the industrial researchers being interested in these topics.

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[10] At the outset, we have confirmed that undesirable changes in the beam properties, such as the skew of the polarization principal axis, should not be accompanied with the reversal in polarity of the light-source device. To properly obtain the magnetic Compton signal with helicity switching at long intervals, furthermore, it is crucial to reduce the count-rate dependence of signal-peak shape or energy resolution in electronics as much as possible. In the case of our measurement system using the common spectroscopy amplifier (the shaping time = 1 μs) and 200-MHz analog-to-digital converter with pile-up rejecter, the counting rate had to be adjusted about 8000 cps (counts per second) or less. Higher count rate resulted in the failure to compensate the nonmagnetic parts of spectra and/or the base lines in the subtraction profile between the two measured with the long-interval switching. Note that, as 10 sets of detector elements and the respective independent signal-processing channels were used in the present measurements, the total count rate should amount to about 10 times, i.e., $\sim 8 \times 10^4$ cps.
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