ゼロ磁化強磁性体（Sm, Gd）Al_2の円偏光スイッチングによる $Sm\ M_{4,5}$ 内殻吸収磁気円二色性

Sm $M_{4,5}$ core level XMCD studied by circularity switching method for zero-magnetization ferromagnet (Sm, Gd)Al_2

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(Sm,Gd)Al_2 is expected to be a ferromagnetic material with zero net magnetization because of the cancellation between spin and orbital magnetic moments at a certain compensation temperature. Our previous X-ray magnetic circular dichroism, XMCD, studies (proposal No. 2002A0493 and 2002B0681) confirmed the ferromagnetic order for both spin and orbital magnetic moments at the compensation temperature. To study the mechanism of the cancellation, the temperature dependence of spin and orbital magnetic moments needs to be studied. The experimental error of the previous experiments is not small enough to get reliable result. In this work, we did the experiment by switching the circularity of photons in every data point to avoid the experimental error related with the long time period. The experimental results with very small error clearly show that the magnitude of the ratio of orbital to spin magnetic moment of Sm 4f electrons decreases with the decrease of temperature.

Introduction

Samarium atoms, with the outermost electron configuration of $4f^6 6s^2$, usually contribute three electrons to the conduction band when combined into solids and the atoms become Sm^{3+} with the $4f^5$ configuration. The ground electronic state of Sm^{3+} can be, in principle, described by Hund's rules labeled as $^6H_{5/2}$. Then we can know that the size of the spin magnetic moments M_S is almost same as that of the orbital one M_L. Since the 4f electron number is less-than-half-filled, the total angular momentum has J=|L-S | , that M_S and M_L tend to be aligned antiparallel due to the spin-orbit interaction and only a small net magnetic moment of $(5/7)^{1/2}\mu_B$ is left. The contribution of M_S and M_L to the net magnetic moment, namely their projections on the total angular momentum J, can be evaluated to be $(25/35^{1/2})\mu_B$ and $(30/35^{1/2})\mu_B$ respectively, that the contributions from M_L is larger and M_L (M_S) is parallel (antiparallel) to the net magnetization.

Because of the small net magnetization left in Sm^{3+}, the contributions from magnetic moments of conduction electrons and doped ions to the magnetism of samarium compounds becomes important [1-3] and the tuning of the magnetic properties was demonstrated by Adachi and Ino that the magnetization became zero at a certain compensation temperature T_{comp}, when some of the samarium atoms in $SmAl_2$ were substituted with the gadolinium ones. The reason is that Gd^{3+} has only spin magnetic moment and can cancel out the surplus orbital magnetic moment in Sm^{3+}. They suggest that $(Sm,Gd)Al_2$ is a ferromagnet (with long-range order for both spin and orbital magnetic moments) without net magnetization (spin and orbital magnetic moments cancel each other out due to their antiparallel coupling) at T_{comp}. [4] This kind of material has a broad prospect for the applications, for example, spin-polarized STM and spintronics devices, which utilize the quantum-mechanical interactions related with the electron spins where the long range macroscopic magnetic interaction is harmful to the function of spin-based effects. In spin-polarized STM measurements, the magnetic structure of the sample surfaces can be destroyed by the stray magnetic field from the magnetized probe tips and the zero-magnetization tips, which can still supply spin polarized electrons, are highly desired. Adachi et al. studied $(Sm,Gd)Al_2$ by Magnetic Compton Scattering (MCS) and their results confirmed the existence of ferromagnetic ordering for the spin component of the total magnetic moment. [5] To confirm the ferromagnetic property completely and to study the mechanism of this peculiar magnetic character, the individual behavior of the spin or orbital magnetic moment for each electronic state should be studied quantitatively. XMCD, x-ray magnetic circular dichroism, is most suitable for this task because we can evaluate spin and orbital magnetic moments separately for a certain electronic state (for example, 4f or itinerant 5d states) in an element selective (Sm or Gd) manner through the tuning of the photon energy. In this work, the Sm $M_{4,5}$ absorption edges are used which gives us the information about Sm 4f electrons.

Experimental

The measurements were carried out at the beam line BL25SU of SPring-8 synchrotron radiation facility. The sample studied here is $Sm_{0.982}Gd_{0.018}Al_2$ polycrystal, same as that used in reference 4 and its T_{comp} is about 81K. Even for non-ferromagnetic materials, small XMCD can still be observed under an external magnetic field. To eliminate this possibility, the sample was magnetized in advance at a certain temperature (defined as T_m hereafter) and the external magnetic field was removed before the measurements. In previous experiments (proposal No. 2002A0493 and 2002B0681), two successive scans were carried out, that after the first scan, the circular polarization of the incident photons was reversed and the XMCD was obtained as the difference of absorptions in these two scans. For the estimation of experimental error, every measurement was done for two times.

Results and Discussion

From our previous experimental results, the following results were obtained for Sm 4f electrons. The ferromagnetic order for both M_S and M_L at T_{comp} was confirmed. M_S and M_L were found always to align antiparallel with each other as understood from the Hund's third rule. When $T_m < T_{comp}$ ($T_m > T_{comp}$), M_S is negative (positive), that is, antiparallel (parallel) to the net magnetization. This agrees with the results of the MCS study [5], showing that the magnitude of the total spin magnetic moment ($M_{ST}=M_S+m_S$) is smaller (larger) than the total orbital one (M_{LT}=M_L+m_L) at the temperature lower (higher) than T_{comp}, where m_S and m_L are the contributions from electrons other than Sm 4f ones. From our knowledge, the main part of m_S comes from Gd 4f and Sm 5d electrons and the m_L is negligibly small. Then we can infer that the magnitude of ratio M_{LT}/M_{ST} increases with the decrease of temperature and equals to one at T_{comp}. The size of M_L was found always larger than that of M_S, which agrees with the estimation made before. When temperature decreases, the size of both M_S and M_L were found increase. The amplitude of M_L/M_S was found almost unchanged or rather decreases as shown in

Fig. 1 by star marks. We can see the experimental error is too large compared with the change of temperature dependence of the ratio, so higher experimental accuracy is needed. In this work, measurements were done by switching the circularity of photons in every photon energy to avoid the experimental error related with the long time period between the two scans in previous measurements and the result is shown in

Fig. 1 by open circle marks. We can see clearly that the magnitude of ratio M_L/M_S decreases with the decrease of temperature. This shows that the increase of the amplitude of M_{LT}/M_{ST} is not due to the Sm 4f electrons. The magnetic moments of Sm 5d and Gd 4f electrons may play important roles in the subtle adjustment of magnetic moments to the zero magnetization. From the above results, the whole story of the magnetism of (Sm,Gd)Al_2 can be expressed as follows.

When the temperature is below the Curie temperature, both M_S and M_L have long-range ferromagnetic ordering, that the spin (orbital) magnetic moment of Sm 4f electron aligns with each other and the spin ones are antiparallel with the orbital ones. When temperature T > T_{comp}, as shown in , the size of the total orbital magnetic moment M_{LT}≈M_L is smaller than that of the spin one M_{ST}=M_S +m_S. When the temperature decreases, for Sm 4f electrons, both the magnitudes of M_L and M_S increase and the increasing rate of M_L is slower than that of M_S, but the change of M_{LT} is faster than that of M_{ST}, which is due to the temperature dependence of magnetic moments of Sm 5d electrons or Gd ions. At T_{comp}, the amplitude of M_{LT} becomes the same as that of M_{ST}, and results in zero net magnetization as observed.

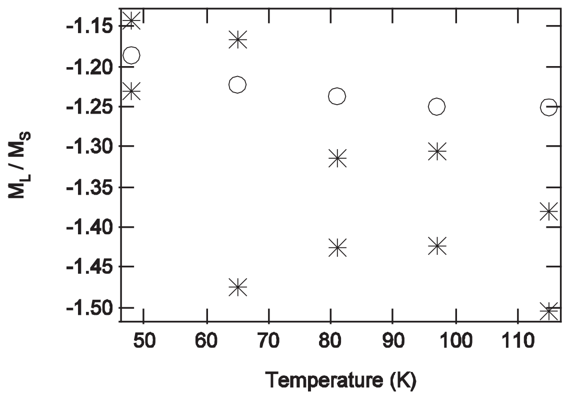

Fig. 1 The temperature dependence of ratio M_L/M_S of Sm 4f electrons in $Sm_{0.982}Gd_{0.018}Al_2$ obtained from our XMCD results. The star marks are the results got by previous experiments and the open marks are the result of this work by circularity switching method. In every temperature, two measurements were taken to estimate the experimental error. For circularity switch method, the experimental error is so small that no difference between the two measurements can be found.

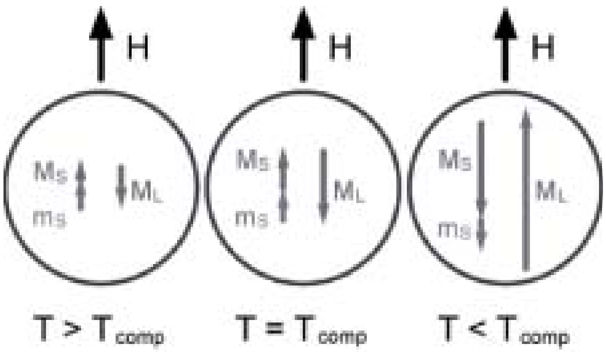

Fig. 2 The temperature dependence of the spin and orbital magnetic moments is schematically described for Sm 4f (ML and MS) as well as other (mS) electronic states (see text).

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