Temperature-dependent magnetic Compton scattering study of spin moments in $Ce(Fe_{0.94}Ru_{0.06})_2$

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We report on the study of spin moments in $Ce(Fe_{0.94}Ru_{0.06})_2$ using magnetic Compton scattering. The measurements on polycrystalline sample were carried out at SPring-8, Japan with 175 keV elliptically polarized synchrotron radiation at 70, 90, 120, 150, and 190 K in 2 T field. The temperature variation of the magnetic effect exhibits clearly the double magnetic transition, i.e., from antiferromagnetic to ferromagnetic and ferromagnetic to paramagnetic transition in agreement with the resistivity and magnetization studies on this material. A comparison of temperature-dependent spin moments in the present sample with $Ce(Fe_{0.96}Ru_{0.04})_2$ shows interesting features of spin momentum density.

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The magnetic properties of intermetallic compounds, in particular RT_m where R stands for rare earths and T_m for transition metals, are peculiar and among these, Ce shows a characteristically different behavior because of strong hybridization between the valence like $4f$ electrons and conduction electrons.[1] Consequently, $CeFe_2$ and its pseudobinaries have been attracting attention continuously almost for the last three decades.[2–18] This interest has been renewed by reports on the fragile nature of ferromagnetism at low temperature and the observation that the magnetic behavior of the $Ce(Fe_{1-x}M_x)_2$ compounds (where M is usually a $3d$,$4d$, or $5d$ transition metal dopant) is strongly dependent on the nature and quantity of the impurity added. A couple of years ago, we had undertaken a magnetic Compton study on $Ce(Fe_{0.96}Ru_{0.04})_2$ in which the characteristic double magnetic transition was clearly observed.[19] It was also found that the spin moments in the ferromagnetic (FM) region showed a variation very similar to that seen in magnetization measurements. In an exhaustive study on Ru doped $CeFe_2$,[11] it has been observed through resistivity, susceptibility, and magnetization measurements that an increase in Ru concentration from 4% to 8% reduced the ferromagnetism. Also, the two phase transition temperatures changed ($T_N \sim 70$ K and $T_C \sim 180$ K) and the region of the FM phase shrank. It was, therefore, thought of interest to attempt a magnetic Compton study of $Ce(Fe_{0.94}Ru_{0.06})_2$ compound to examine whether these fine features, namely, decline in ferromagnetism and shrinkage of FM region, showed up in the spin momentum density, and to analyze this in terms of the individual site moments.

The measurements on the polycrystalline $Ce(Fe_{0.94}Ru_{0.06})_2$ sample (3 mm diameter $\times$ 5 mm length) were carried out in the reflection geometry at the high energy inelastic scattering beamline BL08W at SPring-8, Japan. Since the details of experimental setup are already published,[20] only salient features of the experiment are given here. During the measurements, the peak brightness of the elliptical multipole wiggler was 1.378×10^{17} phs^{-1} mrad^{-2} mm^{-2} per 0.1% bandwidth. The white beam was monochromatized by a bent Si (620) asymmetric Johann-type monochromator and focused to a 1.1 mm high and 1.6 mm wide spot at the sample position. The asymmetry angle for (620) reflection was 1° and the beam energy width $\Delta E/E$ was calculated to be 1.1×10^{-3}. The energy of incident radiation was selected as 175 keV with elliptical polarization ($P_c \sim 0.5$) so as to obtain enough intensity and a reasonable magnetic effect.

The radiation scattered at 178° was energy analyzed by a ten-element (each 100 mm$^2 \times 15$ mm) Ge solid-state detector (SSD). Each element had its own power supply, amplifier, and counting chain, which were adjusted to ensure identical energy calibrations. The momentum resolution was 0.50 a.u. The digitized data were collected by multichannel analyzer (Labo Co., Japan) and the data accumulated in the analyzer were transferred to a computer using the MS-Windows-based "MagCompton" program. The sample magnetizing field of 2.0 T was aligned almost along the x-ray scattering vector. The magnetic field in the sample was reversed in the sequence of (+−−+−++−), where (+) and (−) represent the relative directions of the magnetic field [(+) being parallel to the scattering vector and (−) vice versa]. A switching time of

6 s and dwell time of 60 s were used to ensure good signal averaging. The temperature of the sample was varied from 70 to 190 K with a two-stage Gifford-McMahon-type refrigerator operated with helium gas circulation, which was attached to a superconducting magnet and through a cold finger. The usual corrections for absorption, magnetic cross section, and fluctuation in beam intensity as determined by ionization chamber were applied to the raw data. Since the analyzed Compton line lies in a relatively narrow energy range where the efficiency of the Ge SSD is close to 100%, no energy-dependent detector efficiency correction was deemed necessary. Subtraction of two normalized data sets (I^+, I^-), consisting of both charge and magnetic components, yielded the magnetic Compton profile because the charge profile cancels out. The corrected data were then transformed from energy to momentum scale in atomic units. Since the spin-dependent multiple scattering is always very small $(\sim 1\%)$,[21] no correction for multiple scattering was applied. The magnetic Compton profiles were folded at $p_z=0$ to improve the statistical precision of the data. Finally, the magnetic Compton profiles were normalized to deduce spin moments per formula unit as per the procedure of Ref. 19. The magnetization measurements were made using SQUID magnetometer (Quantum design MPMS5) at 2 T field.

Figure 1(a) shows the temperature-dependent total spin moment determined from the present magnetic Compton experiment and the magnetization measurements under zero field cooling (ZFC) in an applied field of 2 T for $Ce(Fe_{0.94}Ru_{0.06})_2$. Also shown here are the results on $Ce(Fe_{0.96}Ru_{0.04})_2$ reproduced from our earlier work.[19] In addition to the earlier data, a new graph for the 4% compound under ZFC has been added to examine the origin of magnetism in the low-temperature phase. The closeness between the two curves for 6% compound again points to very small orbital magnetism in these compounds in the FM state. The present data show clearly the onset of ferromagnetism from paramagnetic phase around 190 K and then the transition from ferromagnetism to antiferromagnetism around 70 K. It may be mentioned that the magnetization data published for the 4% Ru-doped sample in Ref. 19 was, in fact, obtained in the field-cooled (FC) mode in which case, as mentioned by Sokhey *et al.*,[10] the sample (4% Ru doped) does not reach the low-temperature equilibrium antiferromagnetic (AFM) state, at least down to 5 K. The FC data therefore do not represent the actual AFM state and the difference between magnetic Compton scattering and the earlier bulk magnetization data lies in the mode of magnetization measurement. In view of the ZFC results shown here, it can be concluded that there is little orbital contribution in the low temperature range (<50 K) in both 6% and 4% Ru doped $CeFe_2$. In the case of doped $CeFe_2$, the transition temperature (T_N) from FM to AFM state has a strong dependence on the applied magnetic field. The transition temperature changed from 90 K to about 70 K as the field was increased from 0.03 T to 2 T as observed by other workers also.[11]

Magnetization and susceptibility data indicate a progressive decrease in T_C and enhancement of T_N as doping is increased.[11,22] The change may be rapid or gradual depending on the nature of dopant. Figure 1(a) shows that as compared to 4% Ru doped $CeFe_2$,[19] at 2 T applied field, the antiferromagnetic transition temperature T_N is around 70 K in place of 50 K and T_C is 190 K. This suggests that for 6% Ru doped $CeFe_2$ the ferromagnetism exists between 70 K and 190 K, whereas in the case of 4% doping, it was observed to be between 50 K and 200 K.[19] Thus, the FM region has shrunk in agreement with the observation of Roy and Coles[11] as mentioned earlier.

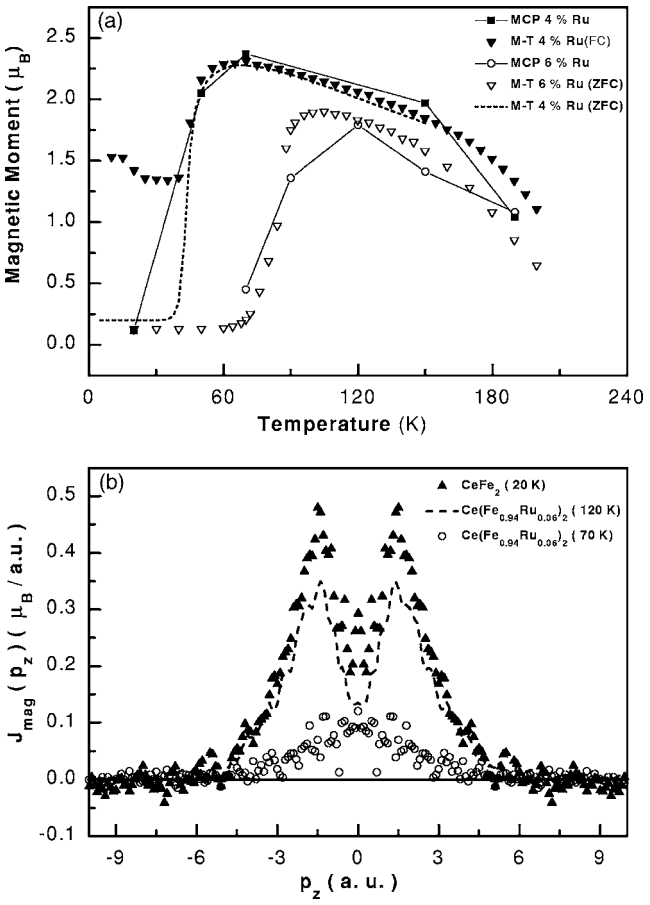

FIG. 1. (a) Temperature variation of magnetic moment for $Ce(Fe_{0.94}Ru_{0.06})_2$ measured by SQUID magnetometer (M-T) in ZFC mode and magnetic Compton scattering (MCP); see text for details. (b) Magnetic Compton profiles of $Ce(Fe_{0.94}Ru_{0.06})_2$ at 70 K and 120 K under 2 T applied field and $CeFe_2$ at 20 K.

The area of magnetic Compton profile $\int_{-\infty}^{\infty} J_{mag}(p_z)dp_z$ provides number of magnetic electrons or spin moment (μ_s). Figure 1(b) shows magnetic Compton profiles of $Ce(Fe_{0.94}Ru_{0.06})_2$ at 70 K and 120 K along with base compound at 20 K. It is obvious that the doped $CeFe_2$ approaches the AFM phase at 70 K but at 120 K it exhibits clearly the FM behavior. So it can be concluded that there is a drastic loss of magnetic effects at 70 K in the 6% Ru doped $CeFe_2$, but, as the temperature is increased, $Ce(Fe_{0.94}Ru_{0.06})_2$ regains its ferromagnetism. Also, it should be noted that the maximum value of the total spin moment in the FM region in this compound is smaller than that for the 4% Ru doped compound in agreement with the results of Roy and Coles[11] obtained by other techniques as discussed earlier.

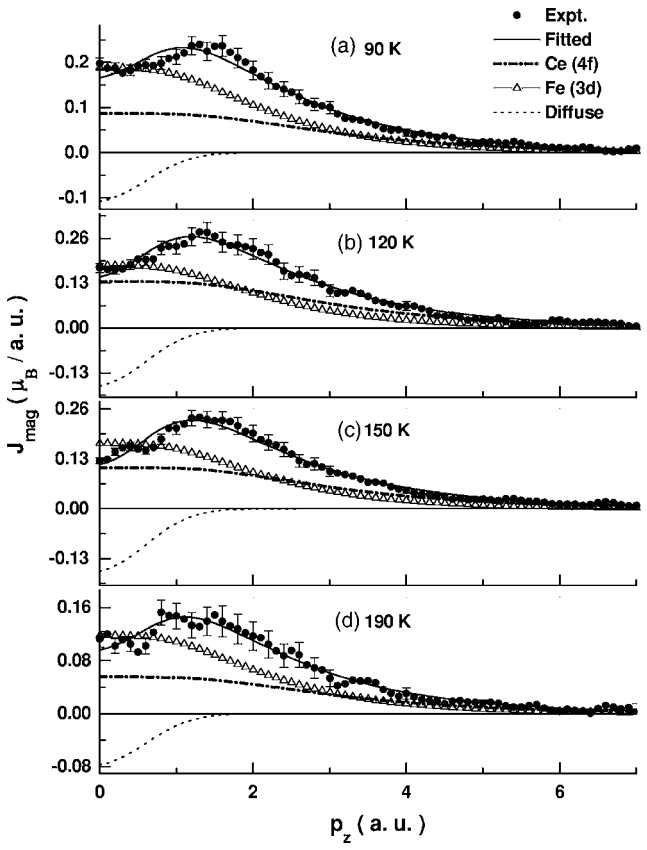

FIG. 2. Decomposition of the magnetic Compton profiles of $Ce(Fe_{0.94}Ru_{0.06})_2$ into the Fe ($3d$), Ce ($4f$), and diffuse (Fe $3p,4s$; Ce $5d$ and Ru $4d$) spin moments at given temperatures. All theoretical profiles are convoluted with a Gaussian of FWHM=0.5 a.u. to mimic the experimental resolution.

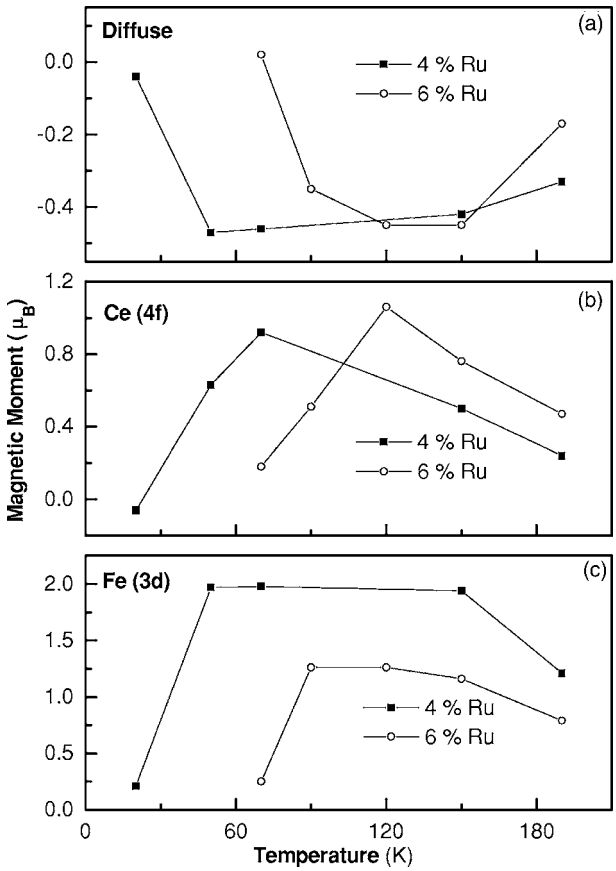

FIG. 3. (a) The temperature variation of individual spin moments in 4% and 6% Ru doped $CeFe_2$ for (a) diffuse electrons, (b) Ce ($4f$) electrons, and (c) Fe ($3d$) electrons.

As is known, the magnetic Compton scattering is sensitive to the spin moments only and this arises from superposition of the contribution from different electrons. As was done in our earlier work (Ref. 19), individual site magnetic moments were obtained in terms of the contributions from different sites by splitting the magnetic Compton profile into its individual components and their temperature variation can be determined. Luckily, in a material such as $CeFe_2$, the momentum distributions of Fe ($3d$) and Ce ($4f$) electrons and diffuse (Fe $3p,4s$; Ce $5d$; and Ru $4d$) electrons are characteristically different. For the diffuse electrons, the Compton profile can be taken as parabolic given by $J(p_z)=ap_z^2+b$ for $p_z<p_F$ and 0 for $p_z \geqslant p_F$ where constants a and b are the functions of a number of free electrons and Fermi momentum p_F. For other electrons, namely Fe ($3d$) and Ce ($4f$), in the absence of band-structure-based profiles, the free atom profiles are a reasonable approximation for the low resolution experiment such as the present one. Our experience in the analysis of magnetic Compton profiles guides us that in such measurements the central dip in the magnetic Compton profile can be fairly well approximated by an inverted parabola having Gaussian full width at half maximum (FWHM) of 1.3 a.u. which corresponds to the FWHM of a free electron Compton profile of the specimen. All these profiles have been convoluted by a FWHM of 0.5 a.u. to incorporate the effect of instrumental smearing.

Figures 2(a)–2(d) show the magnetic Compton profiles for different temperatures, 90, 120, 150, and 190 K, respectively, split into individual components. Due to poor statistics and the loss of magnetic effects, the magnetic Compton profile at 70 K is not shown here.

In Figs. 3(a)–3(c), the temperature-dependent spin moments at each site for the 6% Ru doped sample are presented. Also shown here are the corresponding values for the 4% doped compound taken from Ref. 19. From these figures it can be seen that the diffuse contribution is almost the same in the two compounds with the difference that the value of $-0.45\ \mu_B$ was constant for the temperature range 50–150 K for the 4% compound but for the present sample it is constant over a smaller range of 120–150 K. It suggests possibly some correlation effect between the diffused electrons and Fe/Ce spin moments. The maximum value of Ce ($4f$) moments is almost the same in the FM state for both the compounds but with a shift in temperature. Interestingly, though the spin moment on the Fe ($3d$) site almost remains constant in the FM phase in both the cases, its value is different in the two cases, being smaller in the 6% doped compound. The range of constancy in the spin moments is also smaller in the present case. This is the cause for reduced spin moment.

Besides the Fe site, the largest spin moment in the 6% Ru doped sample at 120 K is due to the Ce ($4f$) electrons, which plays a major role. Coming back to Fig. 3(a)–3(c), in the

temperature range 90–190 K, the spin moment is least at 190 K because of the decrease in Fe (3*d*) and Ce (4*f*) moments. At 70 K, our qualitative data show that the magnetic moment at the Fe (3*d*) and Ce (4*f*) sites drops drastically; the diffuse contribution is also negligible but still positive. This accounts for the loss of magnetic effect at this temperature. It suggests the existence of AFM transition. As regards the sign of Ce and Fe moments, Figs. 3(b) and 3(c) suggest that for both 4% and 6% doping it is parallel, contrary to that for pure $CeFe_2$.[2,5,19] This may possibly be because of Ru doping. It was similar to our earlier studies on pure and 4% Ru doped $CeFe_2$.[19] It requires more measurements on 1%–3% doped $CeFe_2$ compounds.

The temperature-dependent magnetic Compton profiles of $Ce(Fe_{0.94}Ru_{0.06})_2$ have been presented. The variation of magnetic moment with the temperature depicts metamagnetic transition in this sample also. Going beyond the verification of the induced magnetic moments with temperature in the Ru doped $CeFe_2$, we have determined the magnetic moments at different sites of the sample. Band structure calculations, magnetic circular dichroism studies, and more magnetic Compton scattering measurements on 1%–3% Ru doped samples are required for examining the role of dopants in this system.

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