Strong molecular field effect from Gd on 3d-electronic states of Cu in concentrated Gd-Cu non-crystalline alloys

K Yano¹, J Fukuoka², S Yamada³, H Sakurai³, S Okada¹, H Adachi⁴, H Kawata⁴, E Kita⁵ and I Nakai²

¹College of Science and Technology, Nihon University, Funabashi 274-8501, Japan
²Graduate school of Engineering, Tottori University, Tottori 680-8552, Japan
³Department of Production Science and Technology, Gunma University, Gunma 376-8515, Japan
⁴KEK, Photon Factory, Ibaraki 305-0801, Japan
⁵Institute of Applied Physics, University of Tsukuba, Ibaraki 305-8573, Japan

E-mail; kyano@phys.ge.cst.nihon-u.ac.jp

Abstract. The molecular field effect from Gd on the 3d electronic states of Cu was studied from both a macro-scopic magnetization measurement and a micro-scopic way of magnetic Compton profile MCP in the rich content of Gd where the molecular field from Gd was increased as high as possible. From magnetization measurement, the magnetic moment of Gd was estimated to be about 7.0 μ_B in Gd₆₇Cu₃₃ and Gd₇₀Cu₃₀ alloys, respectively. The Curie temperature Tc was found to increase gradually from 140 K for Gd₆₀Cu₄₀ to 150 K for Gd₇₀Cu₃₀. The MCP measurement revealed that some kinds of spin–polarization of 3d electrons in Cu were not detected effectively. That is, the 3d electronic states of Cu were stable and not affected effectively even in the stronger molecular field applied.

1. Introduction

Magnetism in Rare Earth (RE)-transition metal (TM) compounds and alloys has been investigated intensively from fundamental and applied sides [1, 2]. In RE-TM systems, the Ni is well known to lose its magnetic moment at RENi₂ and in more contents of RE. This phenomenon is explained by a charge transfer model that the outer shell electrons of RE transfer and occupy the 3d-electronic states (band) of Ni (TM). That is, the 3d band of Ni is completely occupied at ReNi₂ compound [3, 4]. However very recently, the Ni was found to retain and does not lose its magnetic moment at GdNi₂ and even in GdNi compound [5, 6, 7].

The RE-Cu system, especially Gd-Cu system where the magnetic structure is the simplest, has been one of the most fascinating compounds and alloys [8, 9, 10]. Since the Cu is naturally expected to be non-magnetic, the information about magnetic natures such as exchange interaction energy J_{Gd-Gd} of the RE in RE-TM system is expected to be separated from those of TM and to be obtained clearly without the interferences of TM. However there was an obstacle that the magnetization does not saturate in Gd-Cu system, especially in Cu-rich concentration regions. On the other hand, in Gd-rich region (Gd=50 and 60 at%), amorphous Gd-Cu (a-Gd-Cu) alloys were investigated aiming at deriving the magnetic properties such as J_{Gd-Gd} and so on employing magnetization measurements and the magnetic Compton profile method MCP [10]. In the study, the statistical accuracy in MCP
measurement was not sufficient and the analytical result was no so clear. In addition, the idea of the molecular field effect from Gd on the 3d electronic states was not included [10].

In this study, the effect of strong molecular field upon the electronic states of 3d of Cu was investigated in detail from macro- and micro-scopic points of view. For this aim, the samples of Gd-rich contents for amorphous Gd$_x$Cu$_{1-x}$ (X=60, 67 and 70) were selected.

2. Experimental procedure

Samples of a-Gd$_x$Cu$_{1-x}$ (X=60, 67 and 70) alloys were prepared in the form of ribbons with melt-spinning method of single-roller system. The atmosphere of the chamber was introduced with pure Ar gas. The rotation seed of Cu roll was varied from 3,000 to 4,000 (rpm) depending on the content of Gd.

Temperature dependence of magnetization $M(T)$ was measured with vibrating sample magnetometer (VSM) under a magnetic field up to 11 kOe in a temperature between 4.2 K and 250 K. Saturation magnetization $M_s$ was determined by extrapolating the inverse magnetic field ($1/H$) to zero. Magnetic moment per Gd was derived from $M_s$ under the assumption that Cu is non-magnetic. Curie temperature was determined by Arrott-plot.

Furthermore, magnetic Compton scattering profile (MCP) experiments were carried out at AR-NE1 beam line. Circular-polarized X-rays from an elliptical multi-pole wiggler were monochromatized and focused by a single channel, cut bent Si crystal. The energy of the incident X-rays used was 135 keV. Temperature of the sample was kept at 10 K and 110 K employing a closed-type refrigerator in the magnetic field of 1 Tesla.

3. Results and Discussion

The magnetization as a function temperature was measured for a-Gd$_{67}$Cu$_{33}$ between 4.2 K and 250 K in the magnetic fields of 10,000, 8,000, 6,000, 4,000, 2,000, 1,000 and 50 Oe, respectively. The result is shown in figure 1 (a) and the magnetization process $M-H$ for the same sample at 4.2 K is shown in (b). From figure 1 (a), the magnetic structure is considered to be a simple ferromagnetism. The Curie temperature $T_c$ is found to be about 150 K from low-field (50 Oe) magnetization and this result coincided to that determined by the Arrott-plot. The $T_c$ increases gradually with the increase of Gd concentrations from 100 K (Gd$_{50}$Cu$_{50}$), 140 K (Gd$_{60}$Cu$_{40}$) to 150 K (Gd$_{67}$Cu$_{33}$). Figure 1 (b) shows that this sample is magnetically soft with little anisotropy, however this sample shows a little high field-susceptibility. Therefore the saturation magnetization $M_s$ was determined by the $1/H$-plot and the

Figure 1. Magnetization as function of temperature for a-Gd$_{67}$Cu$_{33}$ alloy (a) and the magnetization process at 4.2 K (b). The applied magnetic fields in (a) were 10,000, 8,000, 6,000, 4,000, 2,000, 1,000 and 50 Oe, respectively.
magnetic moment per Gd atom is derived to be about $7 \mu_B$ under the assumption that the Cu is non-magnetic.

The magnetic Compton profile MCP at 10 K in a field of 1 Tesla is shown in figure 2. The open circles are measured MCP data and the solid and one-dotted lines are the calculated results, respectively, employing the Hartree-Fock calculation for the 4f-electrons of Gd [11]. The best fitting result is found to be between the solid line and the one-dotted line. In RE-TM system, the 3d-electrons of TM and the 4f-electrons of RE play a dominant role in the magnetism and the nearly-free electrons such as 4s, 5d and 6s electrons contribute to exchange-interactions such as RKKY interaction and spin-polarization which attributes to a magnetic moment [12]. From the figure 2, the measured MCP can be fitted well by the calculated MCP for 4f-electrons of Gd in the region of $P_z > 2$ (a.u.). Taking into account that the MCP for 3d-electrons differs clearly from that for 4f-electrons in the region of $P_z > 2$ a.u. [7, 11, 12], it is found that the 3d-electrons in Cu does not contribute to the MCP and they are magnetically inactive in essence. Furthermore, the MCP contributed from s, p-like electrons is well known to be dominant in $P_z < 2$ a.u. [12] and after all, it can be concluded that the measured MCP is composed of the 4f-component of Gd and the s, p-like electrons component of the constituents (=Gd+Cu). Employing the evaluation method that the magnetic moment is proportional to the area of MCP [12], we can estimate the value of the magnetic moment of 4f-component and s, p-like component, respectively. Under the assumption that the magnetic moment of 4f is $6.8 \mu_B$ [10], the value of s, p-like electrons is derived to 0.22-0.45 $\mu_B$ and total. Accordingly, the total magnetic moment from MCP measurement becomes 7.02-7.25 $\mu_B$ and a little larger than that of macroscopic measurement. The value estimated from MCP is nearly the same that obtained for a-Gd60Cu40 and a-Gd70Cu30 alloys. The discrepancy between the microscopic MCP and the macroscopic VSM measurement can be attributed to the scatter of the concentrations of sample.

Figure 2. The magnetic Compton profile MCP for aGd$_{67}$Cu$_{33}$ alloy at 10 K in magnetic field of 1 Tesla. The open circles are the measured MCP and the solid and one-dotted lines are the calculated ones for Gd-4f electrons.

Figure 3. The magnetic Compton profile MCP for aGd$_{67}$Cu$_{33}$ alloy at 110 K in magnetic field of 1 Tesla. The open circles are the measured MCP and the solid line is the calculated one for Gd-4f electrons.
The measurement of MCP at 110 K in the magnetic field of 1 Tesla was also carried out and is shown in figure 3. The MCP at 110 K resembles to that at 10 K and no temperature dependence of MCP is observed.

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