Magnetic Property and Hall Resistivity of Rare Earth Intermetallic Compounds HoAg_<i-x> In_<x>

Magnetic Property and Hall Resistivity of Rare Earth Intermetallic Compounds $HoAg_{1-x}$ In_x

Katsuma YAGASAKI*

Synopsis

Measurements of the magnetization and the Hall resistivity have been made on the polycrystals of $HoAg_{1-x}$ In_x compounds. The compound is antiferromagnetic for $x=0\sim0.2$ and ferromagnetic for $x=0.3\sim0.5$. The Néel and the Curie temperatures take maximum around $x=0.1$ and 0.4, respectively. The ordinary Hall coefficient is positive for all the compounds. It takes minimums at $x\fallingdotseq0.1$ and 0.4 where the magnetic transition temperature takes maximums. The spontaneous Hall coefficient R_s is negative for $x=0$, 0.1, and 0.5 and positive for $x=0.2\sim0.4$. It takes a maximum at $x\fallingdotseq0.3$. The behavior of R_s is not the same manner as the three compound systems of RAg_{1-x} In_x (R=Gd, Tb, and Dy), and cannot be explained by the localized spin model only.

§1 Introduction

Unpaired electrons in 4f shell are responsible for the magnetic moment of rare earth ions. The 4f electrons are closely bound inside the outer closed shells of 5s and 5p, so that the direct overlap between 4f electrons of neighoring ions is negligible, but they are coupled together quite strongly through the conduction electrons.

The pseudobinary compounds of RAg_{1-x} In_x with CsCl type structure have been investigated in order to obtain information on the relation between the magnetic property and the concentration of valence electrons, where the number of conduction electrons per rare earth ion is believed to be controlled by varing indium concentration x.

The author [1),2)] has crried out the measurements of the magnetization, electrical resistivity, Hall resistivity, and magnetoresistance on the heavy rare earth systems of RAg_{1-x} In_x (R=Gd, Tb, and Dy). It was found that the conduction bands of these compounds are, like YZn compound, composed of the majority d-hole band and the minor s-electron band, the carrier concentrations of which are about one per unit cell decreasing with increasing x and about 10^{-4} for d-hole and s-electron bands, respectively. The magnetic properties of these compounds can be explained on the basis of RKKY theory by using the band structure obtained by the author. The transport

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*Dept. of Physics, Colledge of Science, Univ. of the Ryukyus.

properties are also considerably revealed by these measurements, but insufficiently understood yet. The more numbers of studies on the similar compounds are needed.

In this work, the magnetic property and the Hall effect on $HoAg_{1-x}$ In_x compounds have been measured.

§2 Experimental Procedure

The specimens were prepared by a plasma-jet melting of appropriate mixture of terbium, silver and indium metals in argon atmosphere. The purities of the elements were 99.9% for holmium and 99.999% for silver and indium. After annealing at 700℃ for one day in a high vacuum, all the specimens were examined by X-ray diffraction to have a sigle phase of the CsCl type of crystal structure.

The specimens were shaped into spheres of about 2 mm in diameter for measurements of the magnetization and the susceptibility, and narrow thin plates of 0.3mm ×3mm×15mm for the Hall effect. The thickness of the specimens for the Hall effect was measured to an accuracy of 1/1000 mm by using a dial comparator.

The magnetization and the magnetic susceptibility were measured in applied fields

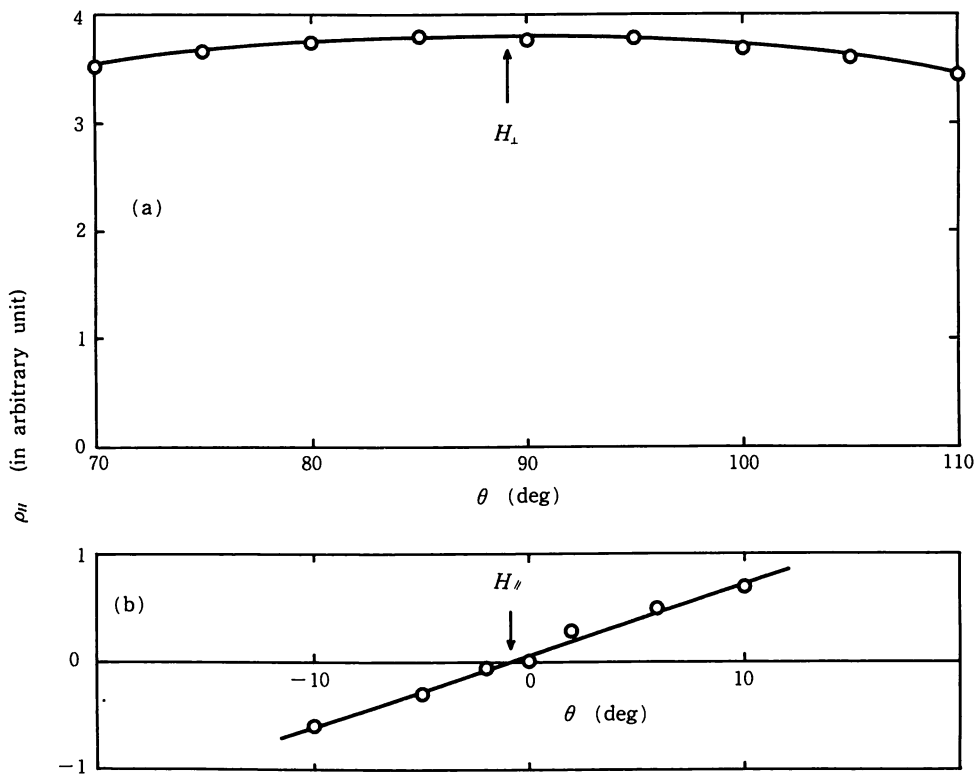

Fig. 1 A plot of the Hall resistivity, ρ_H, against the field direction, θ, for HoAg at 77 k.

up to 15.2 kOe by means of a Faraday type magnetometer in a temperature range from 4.2 K to room temperature. The Hall resistivity was measured by a standard two-probe technique in the paramagnetic temperature from 77 K to room temperature in applied fields up to 12 kOe.

In measurements of the Hall effect, it is most important to set the surface of the specimen to be exactly perpendicular to the applied field. Fig. 1(a) shows the Hall resistivity measured in a field of 12 kOe at 77 K for HoAg when the direction of the field is varied around the roughly adjusted position. At the position where the Hall potential takes a maximum, the field is exactly perpendicular to the surface of the specimen. As the curve of the maximum, however, is very broad, the maximum point is hard to determine very sharply. Fig. 1(b) shows ρ_H measured around the position turned by 90 degrees from the maximum position, where the field is almost parallel to the surface of the specimen. As the perpendicular component of the field becomes smaller, the measurements become more difficult, nevertheless, with accurate measurements, the point of $\rho_H=0$ can be determined very sharply as shown in Fig. 1(b). In this experiment the point of $\rho_H=0$ was determined in the first place, and the field was turned by 90 degrees.

§3 Results and Discussion

The lattice parameters obtained at room temperature are plotted against x in Fig. 2. The lattice parameter increases almost linearly with increasing x.

The magnetization curves measured at 4.2 K are shown for all the compounds in Fig. 3. The curve for $x=0\sim0.2$ is a straight line passing through the origin and it

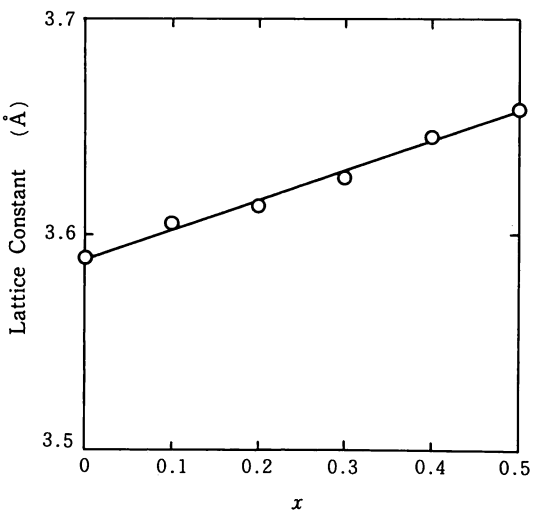

Fig. 2 The concentration dependence of the lattice parameter for $HoAg_{1-x}$ In_x compounds.

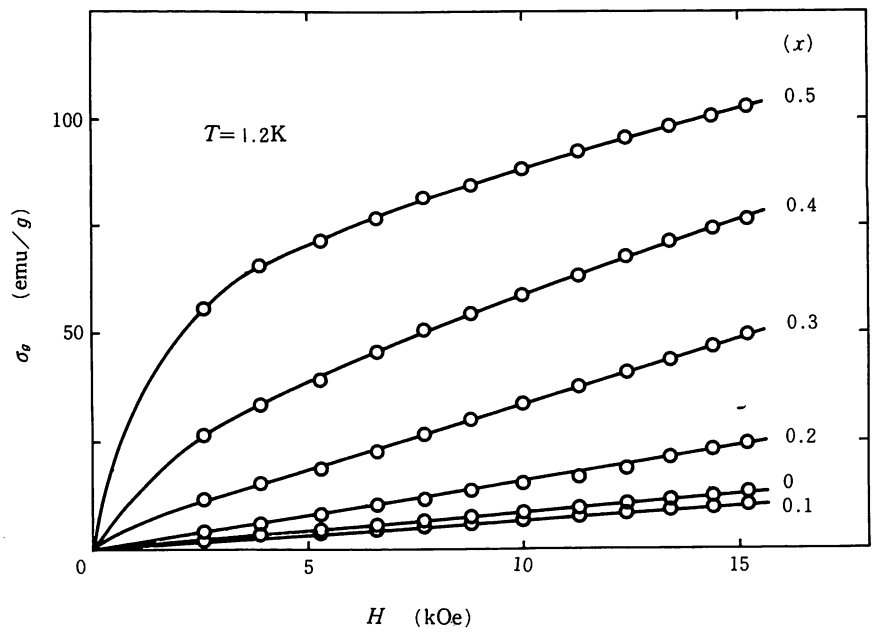

Fig. 3 The magnetization curves measured at 4.2 K for all the compounds.

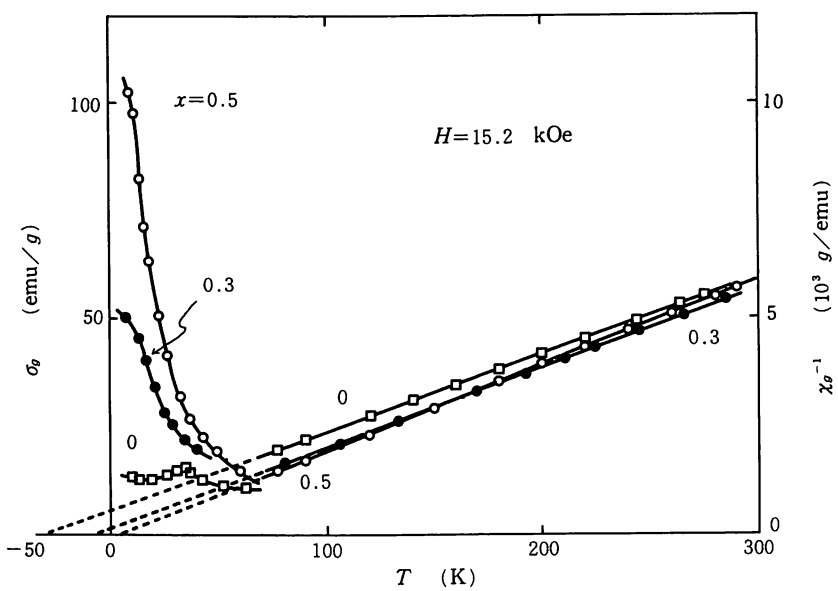

Fig. 4 The temperature dependences of the magnetization, σ_g, and the reciprocal susceptibility, χ_g^{-1}.

shows that the magnetic ordering at low temperature is antiferromagnetic. On the other hand the compound for $x=0.3\sim0.5$ seems to have the spontaneous magnetization. The magnitude of magnetization of $2\sim5$ μ_B in a field of 15.2 kOe, which increases with increasing x, however, is very small compared with the saturation value of $10\mu_B$. The temperature dependence of the magnetization per gram, σ_g, and the inverce susceptibility, χ_g^{-1} measured in 15.2 kOe are shown in Fig. 4 for $x=0$, 0.3, and 0.5, as an example of the measurements. The curve of σ_g-T for $x=0\sim0.2$ has a peak, and T_N was determined as the temperature of the peak. For $x=0.3\sim0.5$, σ_g decreases with increasing temperature and T_C was determined from the Arrot's plot of the magnetization at each temperature. In the paramagnetic temperature, χ_g follows well the Curie Weiss law. The effective numbers of Bohr magnetons obtained from $\chi_g^{-1}-T$ curve are 11.0, 10.9, 10.8, 10.7, 10.7 and 10.5 for $x=0\sim0.5$, respectively. These values are a little larger than the theoretically calculated value assuming the tripositive free ion of Ho, and they show a tendency to decrease with increasing x. The magnetic phase diagram thus obtained is shown in Fig. 5. The compound is antiferromagnetic for $x=0\sim0.2$ and

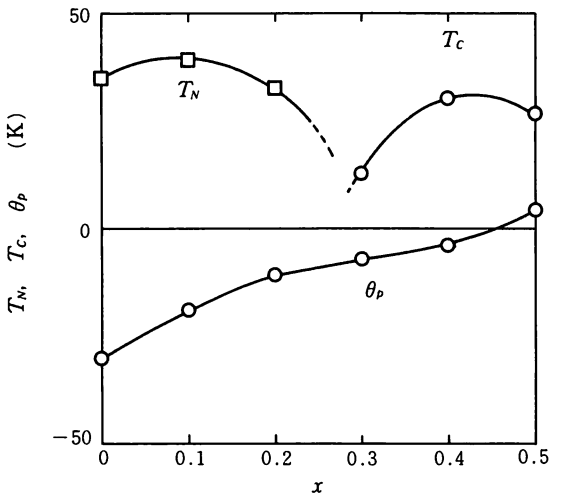

Fig. 5 The magnetic phase diagram for $HoAg_{1-x}$ In_x compounds.

ferromagnetic for $x=0.3\sim0.5$, and T_N and T_C take a maximum around $x=0.1$ and 0.4, respectively. The paramagnetic Curie temperature, θ_p, increases with increasing x and changes the sign from negative to positive between $x=0.4$ and 0.5. With regard to the other compound systems, $GdAg_{1-x}$ In_x[3] is antiferromagnetic for $x=0\sim0.2$, and ferromagnetic for $x=0.2\sim0.5$, $TbAg_{1-x}$ In_x[1] is antiferromagnetic for all the compounds, and the ordering structure is $(\pi\pi0)$ for $x=0\sim0.3$ and (00π) for $x=0.3\sim0.5$, and $DyAg_{1-x}In_x$[4] is, like Tb system, antiferromagnetic for all the compounds. It should be noted that the ferromagnetic appeares again in Ho system. With all these systems, θ_p is negative for $x=0$ and increases with incresing x, and the concentration at which θ_p

changes its sign from negative to positive shifts to larger x with increasing 4f electrons from Gd to Ho.

The Hall resistivity, ρ_H, is expressed by the empirical formula;

$$\rho_H = R_0 H + R_1 M,$$
$$R_1 = (4\pi - N) R_0 + R_s, \tag{1}$$

where R_0, R_1, and R_s are the ordinary, the extraordinary, and the spontaneous Hall coefficients, respectively, and N is the demagnetizing factor, which is nearly equal to 4π in the specimen used by this experiment. In the paramagnetic region,

$$M = \chi H = \frac{C}{T - \theta_p} H, \tag{2}$$

then we obtain

$$\rho_H = R_0 H + R_s C (T - \theta_p)^{-1} H, \tag{3}$$

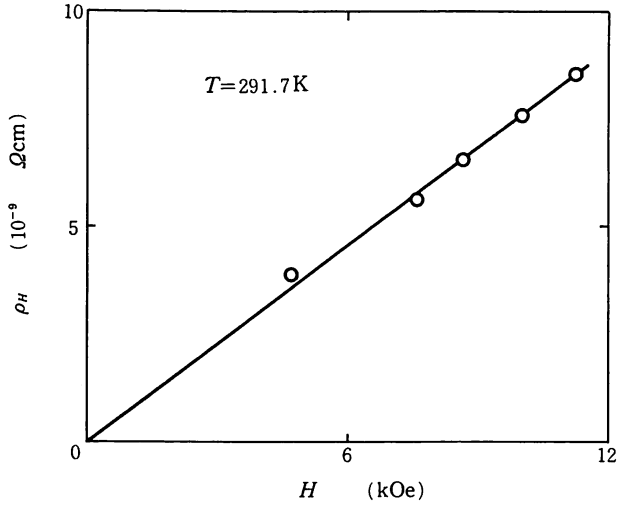

Fig. 6 A plot of ρ_H against H for HoAg.

where C is Curie constant. In Fig. 6, ρ_H is shown as a function of applied field, as an example, for HoAg. The experimental points lie on a straight line so well that eq. (3) is supported. Since ρ_H/H is determined uniquely at all temperatures, we obtain following equation;

$$\rho_H/H = R_0 + R_s C (T - \theta_p)^{-1}. \tag{4}$$

In Fig. 7, ρ_H/H is plotted against $(T-\theta_p)^{-1}$ for all compounds. According to eq. (4), the value of R_0 can be obtained as the intersection of the extrapolation of the line and the ordinate, and R_s as the slope of the line. Fig. 8 shows x dependence of R_0 and R_s thus determined. As same as Gd~Dy systems[2] R_0 is positive for all

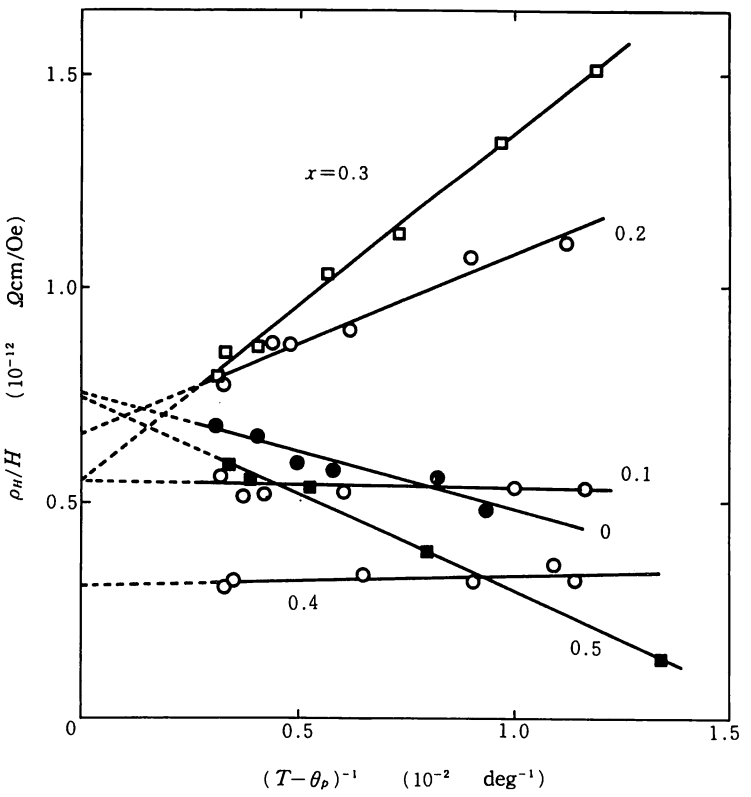

Fig. 7 A plot of ρ_H/H against $(T-\theta_p)^{-1}$.

compounds and it shows that the hole carriers are responsible mainly for electrical conduction in these compounds. R_0 takes two minimums around x=0.1 and 0.4, where T_N and T_C take maximums, respectively. It shows that the electronic state is closely related to the magnetic interaction. R_s is negative for x=0, 0.1, and 0.5 and positive for x=0.2~0.4. It takes a maximum at $x \fallingdotseq 0.3$. With regard to $HoAg_{1-x}$ In_x compounds, x dependence of R_s is quite different from other three compound systems of Gd~Dy in which R_s is negative for $x \geqq 0.1$ and the magnitude increases abruptly with increasing x.

Concerning the anomalous Hall effect on ferrites in which carrier concentration depends on temperature, experimental[5] and theoretical[6] results indicate that R_s is proportional to R_0 in all temperatures. The author showed that R_s normalised by R_0, R_s/R_0, is proportional to 4.0-th power of the total disorder resistivity, $\rho_d(\infty)=\rho_m(\infty)+\rho_v+\rho_{im}$, for Gd~Dy systems. However, in this Ho system, R_s/R_0 depends almost on $\rho_d(\infty)$ in the same manner as the other three compound systems for $x \leqq 0.3$ but does not for $x \geqq 0.4$. This implies that not only the magnetic scattring, $\rho_m(\infty)$, but also the electrostatic potential and impurity scatterings, ρ_v and ρ_{im}, are too responsible for the spontaneaus Hall coefficient in a complicated manner.

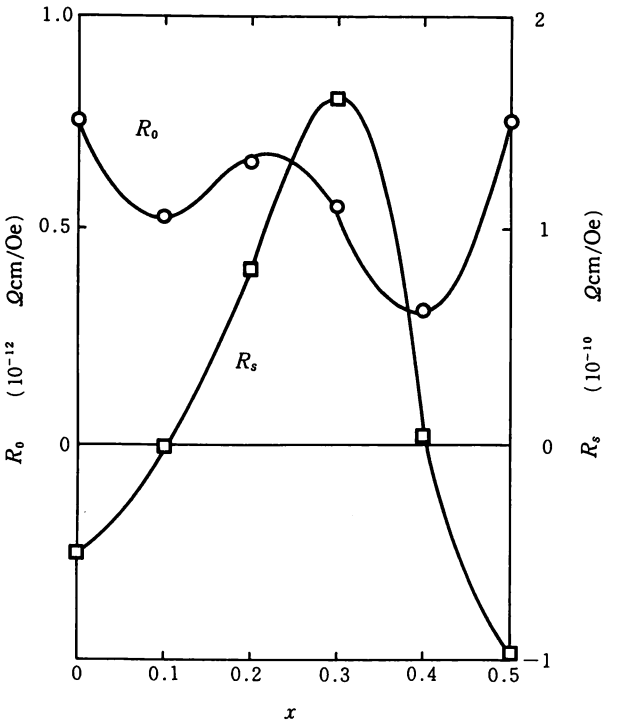

Fig. 8 R_0 and R_s plotted against x.

On the basis of the localized spin model, Irkhin and Abelskii[7] have shown that the spontaneous Hall coefficient R_s is expressed as follows;

$$R_s \propto \frac{\lambda_{eff}}{E_F M(0)} \rho_m(\infty), \tag{5}$$

where λ_{eff} is the effective L–S coupling constant and $M(0)$ is the magnetic moment at $T=0$ K. With regard to RAg, as the electrostatic potential disorder resistivity, ρ_v, is zero and the impurity scattering, ρ_{im}, is small, the total disorder resistivity is mainly composed of the spin disorder resistivity, $\rho_m(\infty)$. So that R_s/R_0 for RAg is expected to follow eq. (5). Fig. 9 shows R_s/R_0 for RAg plotted against $\rho_m(\infty)/E_F M(0)$ where the Fermi energy E_F is assumed to be constant, for $M(0)$ the gJ value of tripositive free R ion is used and for $\rho_m(\infty)$ the value obtained by the author is used. The experimental points is almost linear though dispersion is somewhat large. It is emphasised that the line do not pass through the orign but through a point of negative value of $R_s/R_0 = -85$ on the ordinate. This behavior indicates that R_s may be composed of a term proportional to $\rho_m(\infty)$ and a term of negative constant. The origin or mechanism of this large negative constant term is not explained by the existing theory. As for the details, we shall wait to see what the future holds. To get more detailed information, measurements on the similar compound systems of light rare earth are in progress.

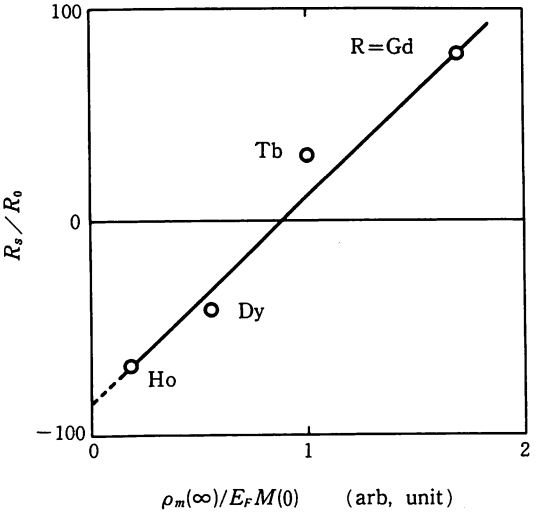

Fig. 9 R_s/R_0 plotted agginst $\rho_m(\infty)/E_F M(0)$ for RAg (R=Gd, Tb, Dy, and Ho)

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