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TRANSPORT AND MAGNETIC PROPERTIES OF Gd$_{3-x}$D$_x$S$_4$

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Résumé.- Nous avons mesuré les propriétés magnétiques et de transport pour Gd$_{3-x}$D$_x$S$_4$. Nous interprétons les propriétés, magnétiques par un modèle d’échange homogène d-f avec une petite énergie de Fermi, que nous avons précédemment traité. Les propriétés de transport ne peuvent pas être expliquées par un modèle normal de polaron magnétique indépendamment lié. La localisation magnétique Wigner est proposée.

Abstract.— Magnetic and Transport Properties of Gd$_{3-x}$D$_x$S$_4$ were measured. Magnetic properties are understood by a homogeneous d-f exchange model with a small Fermi energy treated before by us. Transport properties are not understood by a usual separated bound magnetic polaron model. Magnetic Wigner localization is proposed.

Gd$_{3-x}$D$_x$S$_4$ is an interesting material in the sense that the vacancies in Gd sites enter, randomly even at T=0 in the concentration range 0<x<1/3 and form amorphous like magnetic semiconductor Gd$_2$S$_3$ for x=1/3. Detailed investigation was done by IBM group for four single crystals and they interpreted their data by the bound magnetic polaron model /1/. There exist, however, a lot of ambiguity in their model. We prepared some single crystals of Gd$_{3-x}$D$_x$S$_4$ near x=1/3. One of samples is similar to No. 4 of IBM sample. So, we extended measurement to lower temperature and higher magnetic field to get more informations and to establish the model. A new model is proposed.

1. Magnetic Properties.— From the inverse susceptibility, the effective moment is estimated to be $\mu_{eff} = 7.9\mu_B$ and the paramagnetic Curie temperature $\Theta = 20K$. The number of carrier and the mobility are estimated to be $n_c = 2.77\times10^{-20}/cm$ or $0.015/Gd$ and $\mu = 2.67cm^2/Vs$. from the Hall effect at room temperature. These values are similar to those of No. 4 in IBM samples. Optical reflectivity at room temperature is fitted well by the usual Drude form giving the following values. Optical dielectric constant $\varepsilon = 6.9$, effective mass ratio $m^*/m = 0.53$ and the relaxation time $\tau = 8.2 \times 10^{-16}s$. From the Arrott plot, the Curie temperature is estimated to be $T_c = 2K$. It is difficult to estimate the spontaneous moment accurately due to non linear effect which exists even at high temperature as shown in Fig. 1, but it seems to increase gradually even at $T_c$, saturates gradually with a knick at 4.2K below which it is constant, $2.1\mu_B$. 30% of the real saturation moment $7\mu_B$.

In the pure Gd$_2$S$_3$, $\Theta$ is estimated to be about -10K /1/ and some kind of, including glass like, antiferromagnetic order is expected to occur at lower temperature, even if it is not clear. To check this point, the effective exchange molecular field $J_{eff}(0) <s>$ is evaluated as functions of $<s>$ and the applied field $H_{ext}$.

Fig. 1 Magnetization vs external field for low and high temperature.

As seen in figure 2a as an example, $J_{eff}(0)$ equals $J(0)$ determined from $\Theta$ for small $<s>$, but deviates rapidly with increasing $<s>$, and finally becomes negative. The results are summarized in figure 2b for various external fields. Sharp drops in $J_{eff}(0)$ seen in the curves for $H_{ext} = 40$ and 64kOe, both of
which correspond to \( T=4K \) and approaches \( J_0(0)=-1K \) which is expected in the pure \( \text{Gd}_2\text{S}_3 \), are tentatively assigned as the Néel temperature at which the antiferromagnetic component appears. When \( (J_{\text{eff}}(0)-J_0(0))<s> \) is plotted as a function of \(<s>\), it increases initially with \(<s>\) but becomes nearly constant, \( ZK \), for \(<s>>1\). This suggests strongly that \( 2(J_{\text{eff}}(0)-J_0(0))<s> = J_{\text{df eff}}(0) <s> \), the exchange field due to the 5d electron, and the conduction electron Pauli spin \(<s>\) saturates at \(<s>=1\). This is actually reasonable because then, assuming that \( x \) part of \( n_0 \) are polarized, we have \( xJ_{\text{df eff}}(0) = 270K=0.023 \text{ eV} \), which is reasonable considering the facts that at the bottom of the band the share of the atomic 5d character is not large /2/ and that the self-energy term should be subtracted to get \( J_{\text{df eff}}(0) \) from \( J_{\text{df eff}}(0)/3/ \). The energy difference for the \( \frac{1}{2} \) spin conduction electron at \(<s>=1\), \( 2J_{\text{df eff}}=0.0477^{-1}\text{eV} \), is also reasonable compared with the Fermi energy, with is estimated from the effective mass mentioned above and the three valleys structure common in the 5d band /2/ to be 0.075eV. The homogeneous model seems to be applicable well and the magnetic properties are explained by that model as we treated before /4/. \( x \) seems to be 0.5<\( x <1 \) increasing gradually with \(<s>\).

This is also consistent with the magnetization below 4K as shown in figure 1.

2. Transport Properties.— Electrical resistivity measurements have been carried out down to 1.37K and up to 85 kOe of applied field on two samples cut from the same batch as shown in figure 3.
The results are interpreted as follows. As seen in Ce$_2$S$_3$/5/, the materials should be metallic without the d-f exchange interaction. The localized state is formed by the magnetic polaron effect which seems to be formed below 50K causing the activation energy 110K in the resistivity. Below $T_c$, the binding energy of the magnetic polaron decreases rapidly causing the activation energy smaller. For 9<T<15, the resistivity decreases with decreasing T because of rapid increasing of <$s$>. Below 4.2K, <$s$> is constant and thus the activation energy becomes constant again but very small, 7K. For further decreasing of T, the hopping conduction region with a lower activation energy appears as seen in figure 3a below 1.7K for the cases of H=10 and 15 kOe. With increasing field, <$s$> increases causing decreasing activation energy and the localized magnetic polaron seems to be unstable for <$s$> > 2, or H>30 kOe in the present case, causing the metallic conduction. It is important to note that in three IBM samples in which $n_c$ varies factor three, the metal insulator transition seems to occur at a common values <$s$> $\sim$ 2 /1/. This is hard to understand on the spacially well separated small magnetic polaron model proposed by IBM group. It is also inconsistent with the observed small activation energies and the magnetic properties mentioned before. Alternatively, we propose magnetic Wigner localization.

Wigner crystal is formed by electrons near the Fermi energy when the again in the correlation energy overcome the loss in the Kinetic energy /6/. This occurs, however, only in a narrow band such as $4f$ bands. /6/ In the present case, the d-f exchange interaction encourages the localized and the Wigner crystal is stabilized, which is formed with a small number of conduction electrons and thus each localized state extends over a large space overlapping many electrons with lower energies. The ward, magnetic polaron, used before thus should be replaced by the present magnetic Wigner localized state. All unusual properties reported before are explained on this model. Detail will be given in a separate paper.

References


/2/ Kasuya, T., in this conference.


