Determination of the antiferromagnetic exchange constant between nearest-neighbor Gd^{3+} ions in $Pb_{0.95}Gd_{0.05}Te$

A. Bruno, J. P. Lascaray, and M. Averous
Groupe d'Etudes des Semiconducteurs, Université des Sciences et Techniques du Languedoc, Place E. Bataillon, 34060 Montpellier Cédex, France

J. M. Broto and J. C. Ousset
Laboratoire de Physique des Solides, Institut National des Sciences Appliquées, Avenue Rangueil, 31077 Toulouse Cédex, France

J. F. Dumas
Physicochimie des Matériaux Solides Inorganiques, Université des Sciences et Techniques du Languedoc, Place E. Bataillon, 34060 Montpellier Cédex, France

(Received 22 July 1986; revised manuscript received 29 September 1986)

For what is thought to be the first time high-field magnetization measurements are performed on a semimagnetic semiconductor with rare-earth-like magnetic ions (Gd). Saturation is reached at a field of 12.5 T. The experimental results are well fitted by the cluster model. The value of J_{NN} is determined: $J_{NN} \simeq -1.2$ K.

INTRODUCTION

In this paper we present what are probably the first measurements of the magnetization of a semimagnetic semiconductor in which the magnetic ion is a rare-earth metal (Gd) in high magnetic field. The exchange constant between nearest-neighbor magnetic ions (J_{NN}) is determined for $Pb_{1-x}Gd_xTe$. A very interesting feature of this compound is the low value of J_{NN}, which enables us to observe the total saturation of the magnetic clusters.

Recent measurements of the magnetic field dependence of the magnetization in high magnetic field ($B > 15$ T) were performed for wide-gap semiconductors.[1-5] The magnetization curve shows a steplike character. The data have been analyzed using a generalized cluster model,[1-6] in which the magnetization is calculated as a sum of the magnetizations of small clusters. The role of the magnetic pairs was found to be important. Measurements of magnetization versus magnetic field were also performed for small-gap semiconductors, but the magnetic field did not exceed 15 T.[7,8] The most used magnetic ion was manganese. The phenomenological function

$$\langle S_2 \rangle = S_0 \beta_{5/2}[\tfrac{5}{2} g\mu_B B/K_0(T+T_0)]$$

did not describe accurately the experimental results.

In this paper, we study the magnetization of $Pb_{0.95}Gd_{0.05}Te$ in magnetic fields up to 32 T, at a temperature $T = 1.8$ K. The results are analyzed in terms of the cluster model to determine the nearest-neighbor exchange integral J_{NN}.

RESULTS AND DISCUSSION

The sample of $Pb_{1-x}Gd_xTe$ used in this work was polycrystalline with a Gd concentration of about 5%.

The magnetization was measured in pulsed magnetic fields of up to 32 T, using two concentric opposing pick-up coils. Two measurements were made: the first with the sample outside the coils and the second with the sample inside the coils. The magnetization is taken as the difference between the two measurements. The experimental setup was not calibrated, and the absolute value of the magnetization was not determined. Data were taken during the increase of the magnetic field. The rise time (34 ms) is large enough to assume a steady state during the measurement. Moreover, we have verified that there is not a significant difference between magnetization curves obtained in the increasing and decreasing fields.

Figures 1 and 2 show the magnetization versus magnetic field. Since the magnetization remained constant in the range 18–32 T, only the data obtained up to 22 T are represented for clarity. The crosses represent the experimental data and the full lines are the theoretical magnetizations obtained from the cluster model. In this model the total magnetization is calculated as the sum of two terms M_s and M_p: M_s is the contribution of isolated Gd^{3+} ions and M_p is the pair contribution to the magnetization. For $x < 0.05$ the contribution of clusters of more than two ions could be neglected. M_s can be written as

$$M_s = M_{st}\beta_{7/2}[7\mu_B B/k_B(T+T_0)] \ ,$$

where M_{st} is the saturation of the total magnetization except pairs, and $\beta_{7/2}$ is the Brillouin function for $S_0 = \frac{7}{2}$ in the case of Gd^{3+}. T_0 is a fitting parameter used for the first time by Gaj, Planel, and Fishman.[9] It represents the phenomenological antiferromagnetic interaction of the relatively isolated Gd^{3+} with distant neighbors. The pair contribution M_p can be obtained taking the pair Hamiltonian in the form

$$H_p = -2J_{NN}S_1S_2 + g\mu_B S_p B \ ,$$

where $S_p = S_1 + S_2$, $S_1 = S_2 = \frac{7}{2}$ are the spins of the two members of the pair, and $g = 2$ was previously determined by ESR measurements.[10] The energy levels of the pair for

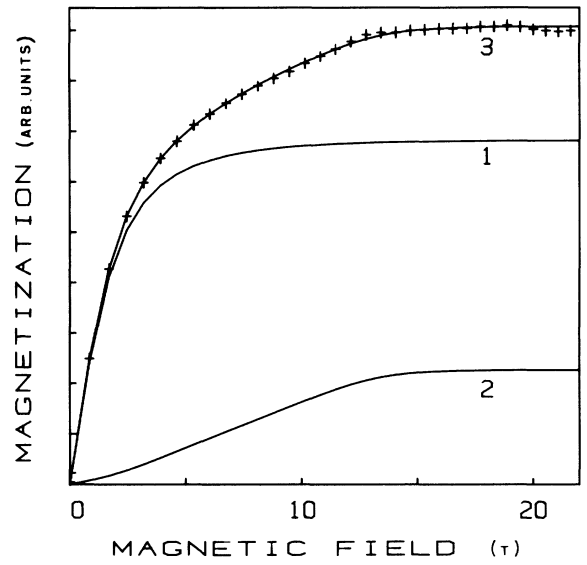

FIG. 1. Magnetization of $Pb_{0.95}Gd_{0.05}Te$ as a function of magnetic field at 1.8 K. Crosses show the data points; solid curve 3 shows the best fit to the data; and curves 1 and 2 represent, respectively, the calculated contribution of singles (Brillouin law) and pairs.

this Hamiltonian are

$$E_{pi} = -J_{NN}[S_{pi}(S_{pi}+1) - (\tfrac{63}{2})] + g\mu_B m_{pi} B \ ,$$

where $S_{pi} = 0,1,\ldots,7$ and $m_{pi} = S_{pi}, S_{pi}-1,\ldots,-S_{pi}$.

The total magnetization of the pairs can be expressed as the thermodynamical average of the magnetization of each level:

$$M_p = -(M_{pt}/2S_0)\frac{\sum m_{pi}\exp(-E_{pi}/kT)}{\sum\exp(-E_{pi}/kT)} \ ,$$

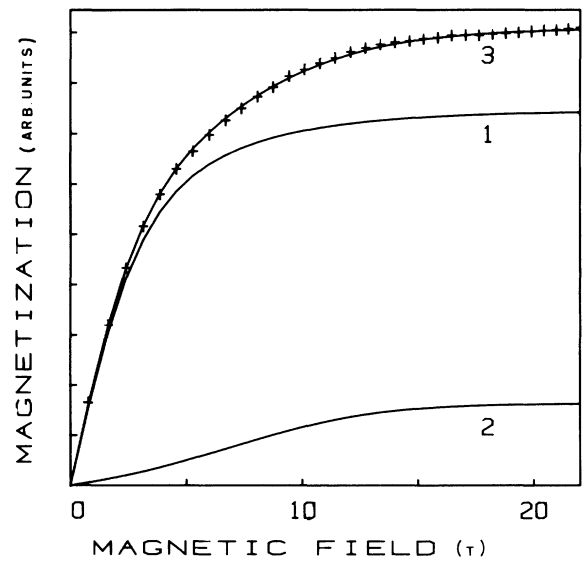

FIG. 2. Same as Fig. 1 at 4.2 K.

TABLE I. Values of the best-fit parameters.

T (K)	T_0 (K)	J_{NN} (K)	M_{st}/M_t
1.8	2.7	−1.2	0.75
4.2	3.2	−1.2	0.82

where the summation is taken over all the states of the pair and M_{pt} is the saturation value of the pair magnetization:

$$M_{pt} = M_t - M_{st} = g\mu_B S_0 x P_2 \ ,$$

where M_t is the total saturation magnetization, x is the Gd concentration, and P_2 denotes the probability of finding one ion in a pair.

In this paper, we have neglected the interaction between the pair and more distant neighbors. Nevertheless, as can be seen, the experimental data are in good agreement with the calculated solid curve. The best fit parameters values are listed in Table I.

It has been found that (i) J_{NN} is constant for the two temperatures. This value is small, $J_{NN} = -1.2$ K. The J_{NN} value deduced by the assumption that the linear part of the magnetization is due to the pair contributions is in quite good agreement with the value obtained by the susceptibility measurement using a Curie-Weiss law.[11] (ii) T_0 is roughly T independent between 1.8 and 4.2 K. (iii) The ratio M_{st}/M_t increases with temperature. As a matter of fact, when T increases, the contribution of the pairs represented by J_{NN} becomes negligible and M_{st} tends to M_s (isolated Gd ions).

The first part of the curve, Fig. 1, up to 2 T, follows a Brillouin law corresponding mainly to the isolated ions. The linear part $2 < B < 12.5$ T is attributed to the pair contribution. As a matter of fact, when $T \ll 2\,J_{NN}$, the pair magnetization could be described by a sum of Boltzmann functions.[1-5] When $T \simeq 2|J_{NN}|$, the contribution of each level could not be separated, and one observes a linear increase versus B, with a slope $(P_2/14|J_{NN}|)(g\mu_B/k)$ as in our case. Above 12.5 T the magnetization reaches the saturation value. The same behavior is observed at 4.2 K (Fig. 2). The pair contribution is less marked because the evolution of the Brillouin law when T increases.

CONCLUSION

High-field magnetic measurements have been presented on a new semimagnetic semiconductor $Pb_{1-x}Gd_xTe$. Due to the specific character of gadolinium, the saturated magnetization was reached at about 12.5 T. The experimental data are well fitted by the cluster model. The model yields a value of $J_{NN} = -1.2$ K. This small value of the exchange interaction is comparable to the value previously determined for $Pb_{1-x}Mn_xTe$ by Anderson and Gorska.[7]

ACKNOWLEDGMENTS

The Groupe d'Etudes des Semiconducteurs, Laboratoire de Physique des Solides and Physicochimie des Matériaux Solides Inorganiques are associated with the Centre National de la Recherche Scientifique.

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