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LETTER TO THE EDITOR

An anomalous magnetic phase transition at 10 K in Nd_7Rh_3

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Abstract
The compound Nd_7Rh_3, crystallizing in a Th_7Fe_3-type hexagonal structure, has been shown recently by us to exhibit a signature of a 'magnetic phase-coexistence phenomenon' below 10 K after field cycling. This is uncharacteristic of stoichiometric intermetallic compounds, and bears a relevance to the trends in the field of 'electronic phase-separation'. In order to characterize this compound further, we have carried out dc magnetic susceptibility (χ), electrical resistivity, magnetoresistance and heat-capacity measurements as a function temperature (T = 1.8–300 K). The results reveal that this compound exhibits another unusual finding at the 10 K transition in the sense that the plot of $\chi(T)$ shows a sharp increase in the 'field-cooled' cycle, whereas the 'zero-field-cooled' curve shows a downturn below the transition. In addition, the sign of magnetoresistance is negative and the magnitude is large over a wide temperature range in the vicinity of the magnetic ordering temperature, with a sharp variation at 10 K. The results indicate that the transition below 10 K is first-order in its character.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Recently, we have reported [1] isothermal magnetization (M) and electrical resistivity (ρ) behaviour as a function of externally applied magnetic field (H) for the binary intermetallic compound Nd_7Rh_3 [2], crystallizing in a Th_7Fe_3-type hexagonal structure [3]. This compound exhibits two magnetic phase transitions, one at ($T1=$) 32 K and the other at about ($T2=$) 10 K. On the basis of isothermal magnetization behaviour, we have proposed [1] that both the transitions are essentially antiferromagnetic (AF) in character. The most intriguing property of this compound is that there is a magnetic-field-induced first-order antiferromagnetic-to-ferromagnetic transition around 10 kOe appearing below $T2$ and that there is a co-existence of antiferromagnetic and 'super-cooled' ferromagnetic (F) phases after the magnetic field is reduced zero, but without spin-glass anomalies. The phase co-existence phenomenon for a stoichiometric intermetallic compound, mimicking the behaviour of manganites, is unusual

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and bears significant relevance to the topic of 'electronic phase-separation' [4, 5] as discussed in our earlier paper [1]. We also found evidence for the 'memory' of high-field conductivity at low temperatures after the magnetic field is reduced to zero, which is taken as evidence for percolative electrical conduction through less resistive ferromagnetic clusters dispersed in more resistive antiferromagnetic clusters. In this letter we present the results of measurements of dc magnetic susceptibility (χ) and ρ as a function of temperature (T) in the presence of various external fields, as well as heat capacity (C), to bring out further interesting aspects of this compound.

2. Experimental details

The polycrystalline sample employed in the present investigation is the same as that in our previous studies [1]. Temperature dependent (1.8–300 K) dc magnetization measurements in the presence of several fixed magnetic fields (100 Oe, 500 Oe, 1 kOe and 5 kOe) were performed employing a commercial (Quantum Design) superconducting quantum interference device (SQUID) as well as a vibrating sample magnetometer (VSM) (Oxford Instruments). The $\rho(T, H)$ behaviour (1.8–300 K) was obtained with the help of a Physical Property Measurements System (PPMS) (Quantum Design). The same commercial set-up was used to measure heat capacity by a relaxation method; in addition, the $C(T)$ behaviour was obtained by an adiabatic heat-pulse method employing a home-made set-up.

3. Results and discussion

3.1. Magnetic susceptibility

The results of dc χ measurements in the presence of various dc magnetic fields are shown in figure 1. ZFC curves represent 'zero-field-cooled' data obtained while warming from 1.8 K in the presence of a desired field after cooling the specimen to 1.8 K in zero field, while field-cooled-warming (FCW) curves were obtained after cooling the sample in the presence of a field. The data were collected during cooling as well for a field of 100 Oe. One of the insets shows the plot of inverse χ obtained in a field of 5 kOe and this plot is linear above 40 K, typical of paramagnets; the value of the effective moment is nearly the same ($\sim$3.72 μ_β/Nd ion) as that of free Nd^{3+} ion and the sign of the paramagnetic Curie temperature (23 K) is positive, indicating the existence of ferromagnetic correlations. There is a distinct peak in $\chi(T)$ in the vicinity of 30 K, indicative of the onset of long-range magnetic order. It should be noted that the peak appears at 32 K for $H = 100$ Oe, whereas for $H = 5$ kOe the peak appears at a slightly lower temperature (30 K). This implies a marginal suppression of the magnetic transition temperature by the application of magnetic field, thereby establishing that the magnetic ordering near 32 K is of an antiferromagnetic type, despite a positive value of the paramagnetic Curie temperature. ZFC–FCW curves do not show any bifurcation near 32 K and hence spin-glass freezing needs to be ruled out. At lower temperatures, we see an additional broad feature around 20 K for $H = 100$ Oe, which is gradually broadened as the magnetic field is increased. There is a distinct bifurcation of ZFC–FCW curves around 20 K. It appears that, in addition to the 10 K-magnetic transition as reported in the literature [2], there could be another magnetic transition around 20 K. The 20 K-feature was not reported in the previous literature [2]. It may be recalled that there are three distinct crystallographically equivalent sites for Nd [3] and it is not clear whether the three different types of Nd ions order magnetically at different temperatures or whether there is a collective ordering at 32 K with spin-reorientation effects with decreasing temperature. The most noteworthy findings in the plots shown in figure 1 are: (i) the upturn

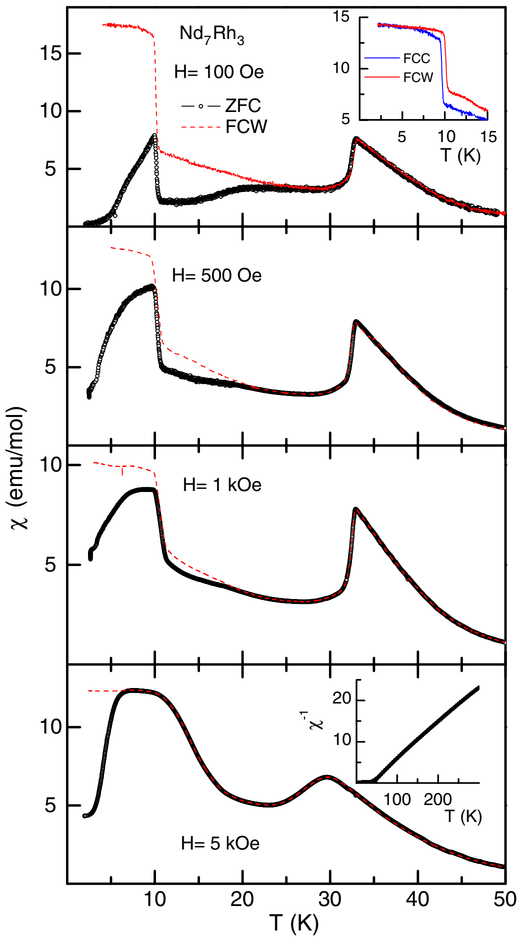

Figure 1. The dc magnetic susceptibility (χ) as a function of temperature (T) for Nd_7Rh_3 below 50 K, measured in the presence of various magnetic fields, for zero-field-cooled (ZFC, from 50 K) and field-cooled (FCW) conditions of the specimens. The data shown were collected while warming. The top inset shows the profiles of FCC and FCW (field-cooled-warming) curves taken with $H = 100$ Oe to show hysteresis. The bottom inset shows inverse χ as a function of T below 300 K, obtained in a dc field of 5 kOe.

in $\chi(T)$ at the onset of 10 K transition is quite sharp for a low field of 100 Oe, as though the transition is first order; however, there is a gradual broadening of the transition with increasing H; (ii) the FCW curve for $H = 100$ Oe does not track the ZFC curve and there is a dramatically sharp rise below 10 K, followed by saturation at lower temperatures; *to our knowledge, such behaviour of the ZFC–FCW* $\chi(T)$ *curves has not been noted in the literature*; as the magnetic field is increased, however, these two curves tend to track each other, eventually merging above 7 K for $H = 5$ kOe. Considering that the upturn below 10 K for the FCW curve for $H = 100$ Oe is sharp, one is tempted to believe that the magnetic transition is first-order in its character in the presence of low fields. We have also obtained $\chi(T)$ curves while cooling in the presence of a magnetic field of 100 Oe and we found a very weak hysteresis (see the top inset in figure 1), as though this transition is first-order at low fields; we would, however, like to view this hysteretic effect with some degree of caution, as it was rather difficult to control

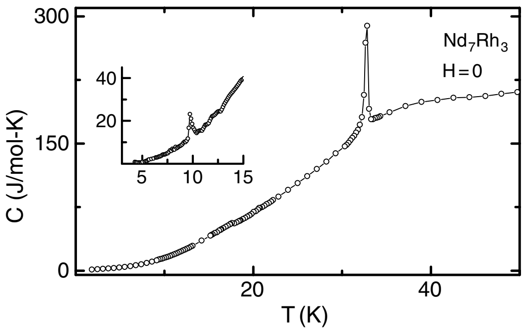

Figure 2. Heat capacity as a function of temperature taken in the absence and in the presence (10 kOe) of a dc magnetic field, obtained by a relaxation method with PPMS, for Nd_7Rh_3. The inset shows the data obtained with a semi-adiabatic heat-pulse method using a home-made calorimeter to highlight the existence of a transition at 10 K.

the temperature to the desired accuracy while cooling the sample. The origin of the broadening of the transition at higher fields is at present puzzling to us.

3.2. Heat capacity

In figure 2, we show the results of heat capacity measurements to look for further evidence for magnetic transitions. The $C(T)$ plot obtained with PPMS shows a well-defined anomaly at 32 K, whereas there is no feature at 10 K. In this connection, it is to be remembered [6] that the relaxation method employed in PPMS to measure heat capacity suppresses sharp features due to first-order transitions. In such cases, the adiabatic heat-pulse method should be preferred to measure heat capacity. In fact, we have performed heat capacity measurements using a home-made set-up by the semi-adiabatic heat-pulse method. The results are shown in the inset of figure 2, and reveal the existence of a well-defined anomaly at 10 K. This comparison of the heat capacity data obtained by these two methods appears to favour the first-order nature of this 10 K-transition. Finally, there is no prominent peak, but a weak one around 20 K in $C(T)$. Considering this, it is possible that the 20 K transition results in the low entropy change, though a possible contribution from magnetic impurities cannot be ruled out.

3.3. Electrical and magnetoresistance

The results of temperature dependent electrical resistivity studies are shown in figure 3. The values of ρ are typically much less than 1 mΩ cm and $d\rho/dT$ is positive down to 60 K over the entire temperature range of investigation. There is a drop at 32 and 10 K (for the data taken in zero field). All these findings are consistent with the previous report [2]. It should be noted that the drop at 10 K in our case is quite sharp, as though the transition is first-order, whereas in [2] the transition is found to be broad. We have also attempted to probe the influence of magnetic field on the magnetic transitions and on the $\rho(T)$ behaviour, the results of which are shown (while warming) in figure 3 for the ZFC condition of the specimen. It is apparent from figure 3 that while the transition around 32 K gets broadened by the application of H, say 50 kOe, the feature at 10 K is washed out completely; this is due to the stabilization of ferromagnetism at high fields. In addition, the values of electrical resistivity are suppressed dramatically in the magnetically ordered state, resulting in negative magnetoresistance, defined as $\mathrm{MR} = [\rho(H) - \rho(0)]/\rho(0)$ (see figure 3, bottom). It is interesting to see that the magnitude of MR is as large as about 60% below 10 K, say for $H = 50$ kOe, arising from

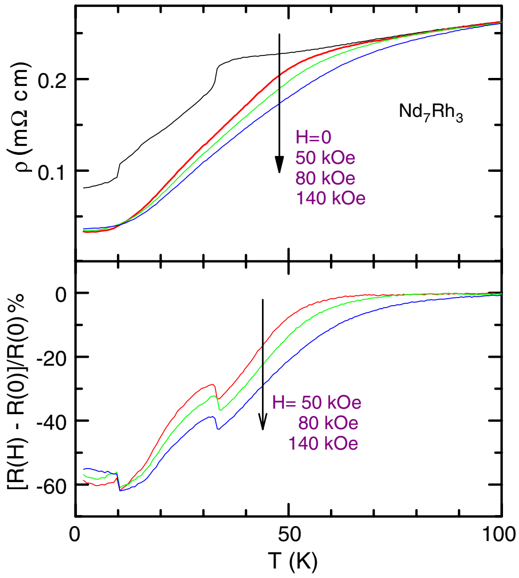

Figure 3. Electrical resistivity as a function of temperature below 100 K in the presence of various dc magnetic fields, for Nd_7Rh_3 (top). The magnetoresistance obtained from these data are plotted in the bottom figure.

percolative conduction through ferromagnetic clusters as explained in [1]; above 10 K also, values of MR are large, arising from the field-induced ferromagnetism [1]. The magnitude of magnetoresistance peaks at the two (10 and 32 K) magnetic transitions. In fact, the suppression of ρ begins at a temperature which is nearly three times the magnetic ordering temperature as in the case of heavy rare-earths of this series, the implications of which have been discussed at length in [7–9]. Finally, there is no prominent peak close to 20 K in $\rho(T)$ or in MR(T), but the change of slopes in the curves (noticeable in MR(T)) could possibly indicate a transition.

4. Summary

The temperature dependence of magnetic susceptibility, magnetoresistance and heat-capacity of the compound Nd_7Rh_3, which was recently reported to show a novel ‘phase co-existence’ phenomenon following field-cycling at low temperatures, is presented in this letter. The results establish that there are at least two magnetic transitions, one at 32 K and the other at 10 K. The most interesting observation made here is that the transition at 10 K in low magnetic fields is found to be very sharp with an unusual bifurcation behaviour of ZFC–FCW dc χ curves. It appears that the transition is first order. It is not clear whether there are any structural changes associated with this transition. Therefore, careful neutron and x-ray diffraction studies as a function of temperature and magnetic field will be quite rewarding.

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