Magnetic properties of NdRu$_2$Si$_2$ revisited

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Abstract

Magnetic properties of ternary NdRu$_2$Si$_2$ are analyzed according to resistivity and specific heat studies at very low temperatures. The compound exhibits two magnetic transitions at 9 and 23 K. The latter is the Néel point, whereas the former one was attributed to a change of magnetic ordering from antiferromagnetic to a complex phase with predominant ferromagnetic component. The specific heat measurements reveal an enhancement of electronic specific heat coefficient $\gamma = 86(1)$ mJ/mol K$^2$. The Debye temperature was estimated based on resistivity studies to be 273.1(5) K.

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1. Introduction

The tetragonal ternary RT$_2$X$_2$ (R—rare earth, T—d electron metal, X—p electron element) compounds exhibit a great variety of interesting magnetic properties stimulating the interest of researchers during the past few decades [1,2]. The NdRu$_2$Si$_2$ compound crystallizes in the ThCr$_2$Si$_2$-type structure (I4/mmm) [3].

Former magnetic and neutron diffraction studies on polycrystalline samples revealed the existence of two magnetic phases below the ordering temperature of 23 K [4,5]. At $T_N = 23$ K Nd magnetic moments become ordered antiferromagnetically with incommensurate propagation vector $\mathbf{k} = (0.13; 0.13; 0)$. With decreasing temperature a third harmonic of $\mathbf{k}$ was observed indicating a squaring of the primary structure below 16 K. At about 9 K magnetic ordering changes from antiferromagnetic to coexisting ferromagnetic and antiferromagnetic ones with domination of the ferromagnetic phase [4].

Neutron diffraction on single crystals reveals the squaring of the AF phase as well (existence of both third and fifth harmonics was reported), however below 9 K a ferrimagnetic phase with small ferromagnetic component was suggested [6].

Transport and specific heat measurements were reported in Refs. [7,8], however neither the Debye temperature nor the electronic specific heat $\gamma$ coefficient were estimated.

The above measurements confirmed the multiple magnetic transitions in NdRu$_2$Si$_2$ compound. In the present paper our former data are reanalyzed bringing new light on NdRu$_2$Si$_2$ properties.

2. Results and discussion

A polycrystalline sample was obtained by melting of stoichiometric amounts of the constituent elements (Nd of the 3N purity, Ru and Si of the 4N purity) in an induction furnace. Subsequently the sample was annealed in an evacuated quartz tube at 800 °C for 100 h. Quality of the product was checked by X-ray powder diffraction at room temperature. The sample turned to be single phase without impurities [7].

2.1. Electrical resistivity

Temperature dependence of electrical resistivity $\rho(T)$ in the 1.8–300 K temperature range is shown in Fig. 1(a). The quasi-linear character of $\rho(T)$ curve above the ordering temperature
As one can see at temperatures lower than 0.5 K the resistivity is relevant for anisotropic ferromagnet, whereas simple relation \( \rho \sim T^2 \) is typical of simple metals. Above the ordering temperature, \( \rho(T) \) was analyzed using Bloch–Grüneisen–Mott formula:

\[
\rho(T) = (\rho_0 + \rho_0^\infty) + 4RT \left( \frac{T}{\Theta_D} \right)^4 \times \int_0^{\Theta_D} \frac{\alpha^2}{(e^\alpha - 1)(1 - e^{-\alpha})} - KT^3
\]

(1)

where \( \rho_0 \) — the residual resistivity, \( \rho_0^\infty \) — the spin-disorder resistivity. The second term is due to electron–phonon scattering and finally the third term accounts for s–d interband scattering processes (Mott scattering). Fitting formula (1) to the \( \rho(T) \) curve above 24 K allowed us to estimate free parameters. The sum of residual and spin-disorder resistivities was found to be 19.9(2) \( \mu \Omega \) cm. Assuming that \( \rho_0 \) can be roughly estimated to be 12.3 \( \mu \Omega \) cm (see inset to the Fig. 1) the \( \rho_0^\infty \) may be evaluated to be about 7.6 \( \mu \Omega \) cm which is similar to the value reported earlier [8]. The \( R \) parameter was found to be 0.221(8) \( \mu \Omega \) cm/K and the Debye temperature is equal to 273.1(5) K. Due to good linearity of \( \rho(T) \) at high temperatures the Mott coefficient \( K \) was found to be negligibly small and was not taken into account. It is worth understanding that, so derived, Debye temperature may be affected by electron–electron correlations and it may be treated as a rough estimation of \( \Theta_D \) [9].

The Fig. 1(b) shows the low temperature part of \( \rho(T) \). The distinct kink at \( T_N = 23 \) K is visible whereas the transition into ferromagnetic phase at about 9 K is far less pronounced. At temperatures of about 6 K the \( \rho(T) \) curve flattens, which was observed in Ref. [8], however with subsequent lowering of the temperature, the resistivity lowers as well. Calculated \( d\rho/dT \) derivative shows similar behavior to that measured in Ref. [8], however two crucial differences are visible. At first, plateau in the 12–16 K temperature range is clearly visible. The upper limit seems to be connected to the onset of squaring of the incommensurate AF structure observed in neutron diffraction [6]. Finally, the reported earlier measurements carried out down to 3.7 K exhibited saturation of resistivity and its derivative [8]. In our measurements we observe further lowering of resistivity and consequently a rise of \( d\rho/dT \) derivative below 4 K. As there are some differences in interpretation of the neutron diffraction data for polycrystalline and single crystal NdRu\(_2\)Si\(_2\) [4,6] below 8 K it is hard to interpret the above mentioned resistivity lowering. Still, we suggest minimizing of the modulated phase below 4 K for powder sample.

The onset of the first anomaly at about 9 K was analyzed using formula for ferromagnetic spin waves [10]:

\[
\rho = C + a\Delta T e^{\frac{\Delta}{T}}\left(1 + 2\frac{T}{\Delta}\right)
\]

(2)

where \( C \) and \( a \) are constants, \( \Delta \) — anisotropy gap in magnon dispersion relation. The resulting fit is presented in Fig. 2. The \( \Delta \) parameter was estimated to be 121.9(7) K. The contribution from the electron–phonon scattering was not considered as it is surpassed by residual resistivities denoted as \( C \) (see Fig. 1(a)). Formula (2) is relevant for anisotropic ferromagnet, whereas simple relation \( \rho \sim T^2 \) is expected for fully isotropic ferromagnetic system with \( \Delta = 0 \).

2.2. Specific heat

Low temperature part of the specific heat is given in Fig. 3. As one can see at temperatures lower than 0.5 K the contribution from nuclear hyperfine splitting is visible. Within the 0.8–4 K temperature range specific heat can be considered
In the tetragonal symmetry the crystal field splits the Nd$^{3+}$ multiplet into five Kramers doublets. In the right side diagram of the Fig. 4 the splitting of four Kramers doublets corresponding to the solid line is presented. The fifth doublet is not shown as the temperature range of specific heat data is limited to 45 K consequently levels above 150 K will not give noticeable contribution. In isostructural compounds depending on T element the overall splitting of the ground multiplet ranges from 135 K (NdCu$_2$Si$_2$ [14]) to 280 K (NdFe$_2$Si$_2$ [15]).

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### References