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Magnetic properties of NpNiGa₅

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Received 19 January 2007, in final form 21 January 2007

Published 17 May 2007

Online at stacks.iop.org/JPhysCM/19/246202

Abstract

We have investigated a polycrystalline sample of NpNiGa₅ by magnetization, specific heat and ²³⁷Np Mössbauer spectroscopy. We confirm that a ferromagnetic order sets in at $T_C \approx 30$ K and that NpNiGa₅ undergoes a second transition at $T_N \approx 18$ K. Mössbauer data indicate that the transition at T_N is accompanied by a steep increase of the Np magnetic moment, which saturates to a value of $0.94 \mu_B$, in good agreement with the recently published neutron diffraction results. The Sommerfeld specific heat coefficient $\gamma = 114 \text{ mJ mol}^{-1} \text{ K}^{-2}$ is the highest among the NpTGa₅ compounds. It compares to the ones observed in the PuTGa₅ superconductors. The trend of the isomer shift along the NpTGa₅ (T = Fe, Co, Ni, Rh) series points to a tendency towards localization of the 5f electrons when increasing the number of d electrons of the transition metal T. Neither fully localized nor fully itinerant models are able to reproduce the whole experimental data. Realistic models should include the dual nature of the 5f electrons, i.e. consider the difference in the degree of itinerancy among 5f electrons in the NpTGa₅ compounds.

1. Introduction

The study of isostructural families provides valuable information that allows identifying trends and key parameters that clarify general mechanisms driving the physical properties of intermetallic compounds. Such systematic comparisons require detailed knowledge of physical properties of the isostructural compounds.

The isostructural AnTGa₅ series is of particular interest because on one hand it represents a large family (An = U, Np, Pu; T = column VIII transition metals) and on the other

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hand it exhibits an exceptional diversity of magnetic and electronic properties, ranging from Pauli paramagnetism to unconventional superconductivity through antiferromagnetism, ferromagnetism and metamagnetism. Moreover, these phenomena occur at temperatures and magnetic fields fairly accessible to transuranium compounds for which safety concerns and the self-heating property drastically limit the accessible range of temperature and experimental facilities.

The $UTGa_5$ compounds with T from the Fe or Co columns are Pauli paramagnets [1], whereas $UNiGa_5$, $UPdGa_5$ and $UPtGa_5$ order in simple antiferromagnetic structures ($\mathbf{k} = [1/2\ 1/2\ 1/2]$ for T = Ni and $\mathbf{k} = [0\ 0\ 1/2]$ for T = Pd, Pt) with reduced magnetic moments (0.75, 0.33 and 0.32 μ_B , respectively) pointing along the *c*-axis [2, 3].

$PuCoGa_5$ and $PuRhGa_5$ exhibit bulk superconductivity below $T_c = 18$ K and 9 K, respectively [4, 5], whereas $PuNiGa_5$ and $PuFeGa_5$ are simple paramagnets. It is worth mentioning that $PuTGa_5$ compounds with T = Co and Rh, in the normal state, and T = Ni, are Curie–Weiss paramagnets [6].

The $NpTGa_5$ compounds (T = Fe, Co, Rh, Ni) all order magnetically. $NpCoGa_5$ is a type I antiferromagnet ($T_N = 47$ K, propagation vector $\mathbf{k} = [0\ 0\ 1/2]$) with Np magnetic moments $\mu_{Np} \approx 0.84\ \mu_B$ oriented along the *c*-axis and a metamagnetic transition to an induced ferromagnet at $B_c = 4.5$ T (at $T = 5$ K) [7–9]. $NpRhGa_5$ also develops a type I antiferromagnetic order below $T_N = 37$ K with magnetic moments $\mu_{Np} \approx 0.96\ \mu_B$ that point along the *c*-axis but rotate into the basal plane at $T^* = 32$ K [10–14]. $NpFeGa_5$ orders antiferromagnetically ($\mathbf{k} = [1/2\ 1/2\ 0]$) at $T_N = 118$ K and undergoes a second transition at $T^* = 78$ K. Both Np and Fe carry magnetic moments that are aligned along the (110) axis in the high temperature phase. Below T^* an antiferromagnetic component ($\mathbf{k} = [1/2\ 1/2\ 1/2]$) appears and the moments ($\mu_{Np} \approx 0.86\ \mu_B$) rotate (25°) out of the basal plane [11, 14–16].

$NpNiGa_5$ is so far the only $AnTGa_5$ compound that exhibits ferromagnetic order. The Curie temperature is $T_C = 30$ K, but below $T_N = 18$ K a basal plane antiferromagnetic component ($\mathbf{k} = [1/2\ 1/2\ 1/2]$) appears and leads to a canted antiferromagnetic structure [11, 17, 18]. The saturated magnetization at low temperature reaches 0.6 μ_B/Np , whereas the total Np moment amounts to 0.87 μ_B .

Note that $NpNiGa_5$ also displays the largest specific heat coefficient ($\gamma \approx 100$ mJ mol⁻¹ K⁻²) in $AnTGa_5$ (An = U, Np) systems, comparable to those of $PuCoGa_5$ [19] and $PuRhGa_5$ [20].

In order to complement previous, above-mentioned, investigations, we have measured a polycrystalline sample of $NpNiGa_5$ by ²³⁷Np Mössbauer spectroscopy, in-field specific heat and extended magnetization measurements up to 7 T.

2. Experimental details

A polycrystalline ingot of $NpNiGa_5$ was obtained by arc melting stoichiometric amounts of the constituent elements under an atmosphere of high purity argon on a water cooled copper hearth, using a Zr getter. Starting materials were used in the form of 3N5 nickel and 3N7 gallium shot as supplied by A D Mackay Inc., and 3N neptunium metal. Homogeneity of the sample was ensured by turning over and re-melting the button several times. Weight losses were below 0.5%.

Similarly to the isostructural cobalt and rhodium compounds [7, 10], the arc melted sample was annealed at 750 °C for a week to obtain the $NpNiGa_5$ phase. The phase purity of the sample which crystallizes with the $HoCoGa_5$ structure type was checked by x-ray powder diffraction data (Cu K α radiation) collected on a Bragg–Brentano Siemens D500 diffractometer using a 2θ step size of 0.02°. The diffraction patterns were analysed by a Rietveld-type profile

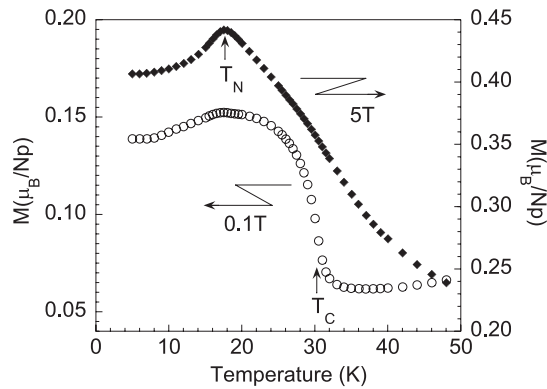


Figure 1. Magnetization of NpNiGa₅ in 0.1 T (left axis) and 5 T (right axis) applied fields.

Table 1. Structural parameters a , c , z_{Ga} and tetragonality t of the Np–Ga layer (see text).

	a (Å)	c (Å)	z_{Ga}	$t \times 100$
NpFeGa ₅ ^a	4.2578	6.7616	0.301	4.40
NpCoGa ₅ ^b	4.2377(1)	6.7871(3)	0.3103(4)	0.60
NpNiGa ₅ ^c	4.2353(2)	6.7858(3)	0.3061(4)	1.91
NpRhGa ₅ ^d	4.2943(1)	6.8392(2)	0.2987(4)	4.86

^a Reference [30].

^b Reference [7].

^c This work.

^d Reference [10].

refinement method using the Fullprof program [21]. The lattice parameters and the adjustable positional parameter thus obtained are presented in table 1 and compared to other isostructural Np compounds. A small amount of an impurity phase could be detected but does not affect significantly the physical property data and/or could be accounted for in the data processing.

DC-magnetization measurements were carried out on a Quantum Design SQUID magnetometer (MPMS-7) in magnetic fields up to 7 T on a 62.57 mg piece of polycrystalline sample. The specific heat experiments were performed using a 1.45 mg polycrystalline sample by the relaxation method in a Quantum Design PPMS-9 within the temperature range 4.2–300 K and in magnetic fields up to 9 T. The ²³⁷Np Mössbauer spectra were recorded using a sinusoidal drive motion of a ²⁴¹Am metal source kept at 4.2 K. The temperature of the absorber containing 219 mg Np/cm² was varied from 4.2 to 40 K. The velocity scale of the spectrometer was calibrated with reference to an NpAl₂ absorber ($B_{\text{hf}} = 330$ T at 4.2 K).

3. Results

3.1. Magnetization

Figure 1 shows the magnetization versus temperature recorded for $B = 0.1$ and 5 T. The ferromagnetic ordering is revealed by the sharp increase of $M(T)$ around $T_C \approx 30$ K, whereas the maximum at $T_N \approx 18$ K indicates an antiferromagnetic-like transition, in good agreement with previous work [11].

The unsaturated magnetization (figure 2) reaches $0.46 \mu_B$ at $T = 5$ K and $B = 7$ T, slightly below the single crystal value along the c -axis ($0.57 \mu_B$ at $T = 5$ K and $B = 5.5$ T [11]).

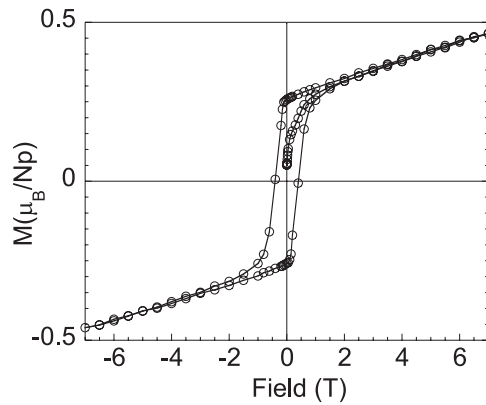


Figure 2. Magnetization of NpNiGa₅ at $T = 5$ K. The line is a guide for the eye.

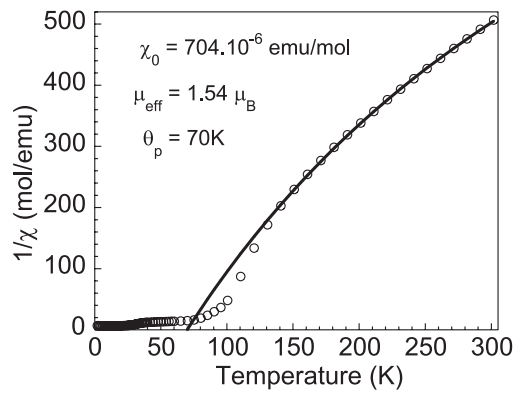


Figure 3. Inverse magnetic susceptibility of NpNiGa₅. The symbols represent the experimental points and the solid line the modified Curie–Weiss fit.

In the paramagnetic state, the magnetic susceptibility can be accounted for by a modified Curie–Weiss law (figure 3):

$$\chi = \chi_0 + C/(T - \theta_p) \quad (1)$$

with an effective moment $\mu_{\text{eff}} \approx 1.54 \mu_B$, a paramagnetic Curie temperature $\theta_p \approx 70$ K and a constant term $\chi_0 \approx 704 \times 10^{-6} \text{ emu mol}^{-1}$. The value of the effective moment is strongly reduced compared to the free ion Np^{3+} ($2.68 \mu_B$), similarly to what is observed in NpCoGa₅ [7] and NpRhGa₅ [10]. The strongly positive paramagnetic Curie temperature reveals the presence of ferromagnetic interactions, in agreement with the magnetic structure inferred from neutron diffraction [18]. Note that Aoki *et al* [11] reported an effective moment ($\sim 2.4 \mu_B$) closer to the Np^{3+} free ion value by adjusting the magnetic susceptibility above 250 K by a Curie–Weiss law.

3.2. Specific heat

Figure 4 shows the two magnetic phase transitions observed at $T_C = 30.4$ K and $T_N = 18.8$ K in zero field. Application of a magnetic field tends to slightly increase both T_N and T_C and strongly decrease the intensity of the related anomalies. In 9 T, an anomaly indicating the onset

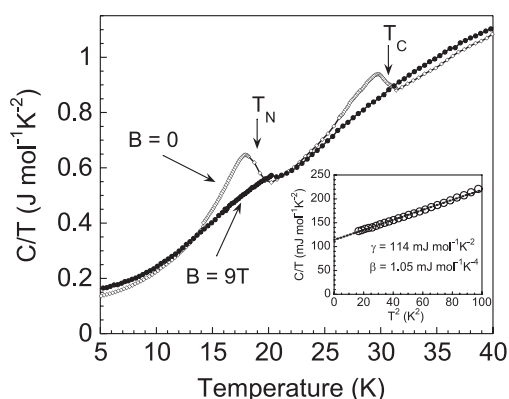


Figure 4. Specific heat of NpNiGa₅ around T_C and T_N in zero field (empty diamonds) and in $B = 9$ T (full circles). The inset shows the linear dependence of C/T on T^2 .

of the antiferromagnetic component is still visible, whereas the anomaly around T_C results only in an additional (compared to zero field) entropy, observed up to ~ 40 K.

The inset of figure 4 shows the low temperature part of C/T versus T^2 . Below $T \sim 10$ K, the specific heat can be accounted for by the linear law:

$$C/T = \gamma + \beta T^2,$$

that reflects the electronic and phonon contributions and yields $\gamma = 114$ mJ mol⁻¹ K⁻², which is slightly higher than the estimation from the previous work [11] and comparable to the values obtained for PuCoGa₅ (95–130 mJ mol⁻¹ K⁻²) and PuRhGa₅ (80–150 mJ mol⁻¹ K⁻² [20]). The value of $\beta = 1.05$ mJ mol⁻¹ K⁻⁴ allows a rough estimate of the Debye temperature $\theta_D \approx 123$ K.

3.3. ²³⁷Np Mössbauer spectroscopy

Figure 5 shows typical Mössbauer spectra recorded at 35 K (paramagnetic phase), 22 K (ferromagnetic phase), 10 and 4.2 K (canted antiferromagnetic phase). At 35 K, the spectrum consists of a single narrow line ($W = 3.3(1)$ mm s⁻¹) indicating the absence of quadrupole splitting despite the non-cubic symmetry of the Np site. The isomer shift $\delta_{IS} = 8.3(4)$ mm s⁻¹ versus NpAl₂ of the single line falls into the range of the previously studied NpTGa₅ compounds (T = Fe, Co, Rh): see table 2. The linewidth of the single line starts to broaden at $T \approx 29$ K, indicating the onset of magnetic order, in agreement with other measuring techniques. At 4.2 K, the spectrum shows a large symmetrical magnetic hyperfine splitting that reflects a vanishing quadrupolar interaction as already observed in the paramagnetic phase. The value of the hyperfine field ($B_{hf} = 202(2)$ T) allows us to infer the ordered magnetic moment carried by the Np ions $\mu_{Np} \approx 0.94(5)$ μ_B through the relation $B_{hf}/\mu_{Np} = 215$ T/ μ_B [22]. This moment value agrees well with the moment (0.87 μ_B) deduced from the neutron diffraction experiments.

When increasing the temperature above 4.2 K, dynamical effects develop. They can be well accounted for by using the Wegener relaxation model (see e.g. [7]).

Figure 6 represents the thermal variation of the Np magnetic moment in NpNiGa₅. The steep increase of μ_{Np} at T_N , due to the onset of an in-plane antiferromagnetic component as shown by the neutron diffraction results [18], is clearly evidenced.

The value of the isomer shifts in the NpTGa₅ series strongly suggests the occurrence of a Np³⁺(5f⁴) charge state. Indeed, it is well established that in conducting materials the

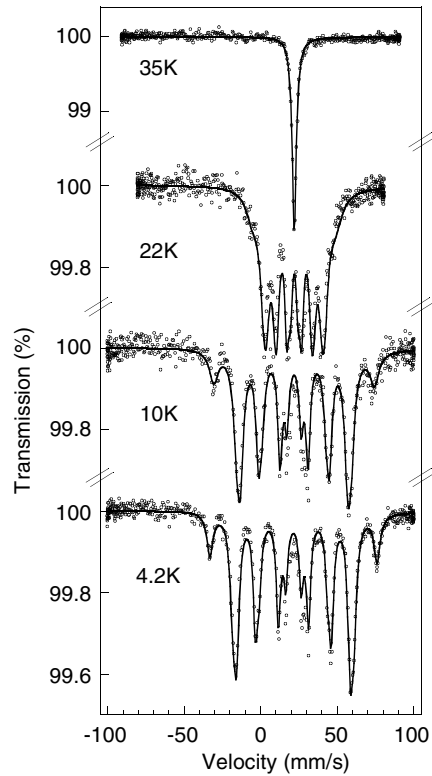


Figure 5. Typical ^{237}Np Mössbauer spectra of NpNiGa_5 recorded at $T = 35, 22, 10$ and 4.2 K.

Table 2. Np saturated moment (μ_{Np}), isomer shift (δ_{IS}) and quadrupole coupling constant (e^2qQ) deduced from the ^{237}Np Mössbauer study of the NpTGa_5 ($T = \text{Fe, Co, Ni, Rh}$) compounds.

	$\mu_{\text{Np}} (\mu_{\text{B}})$	$\delta_{\text{IS}} (\text{mm s}^{-1})$ versus NpAl_2	$e^2qQ (\text{mm s}^{-1})$
NpFeGa_5 ^a	0.86	5.5	-14.6°
NpCoGa_5 ^b	0.84(5)	6.8(2)	$-3.5(5)$
NpNiGa_5 ^c	0.94(5)	8.3(2)	~ 0
NpRhGa_5 ^d	0.96(5)	9.4(2)	$-13.4(5)$

^a Reference [15].

^b Reference [7].

^c This work.

^d Reference [10].

^e Calculated from $(1/2)e^2qQ(3\cos^2\theta - 1) = +3.4 \text{ mm s}^{-1}$ [15] and taking $\theta = 65^\circ$ as deduced from the neutron diffraction results [16]. θ is the angle between the direction of the Np moment and the main principal axis of the electric field gradient, which is along the tetragonal c -axis.

presence of conduction electrons increases the electron density (decreases δ_{IS}) at the Np nucleus compared to the free ion Np^{3+} value ($\delta_{\text{IS}} = +37 \text{ mm s}^{-1}$ versus NpAl_2) and shifts δ_{IS} to smaller values. The rather narrow range of δ_{IS} values agrees with the fact that the electronic structures of the NpTGa_5 compounds are rather similar. The observed increase of δ_{IS} with the number of d electrons of the T element (i.e. from Fe to Ni) and within the same column with the size of T (Co to Rh) points to a stronger tendency towards a localization of the $5f$ electrons within the series. This trend coincides well with the deviations of the ratio $|\mu_L/\mu_S|$ of the orbital to spin

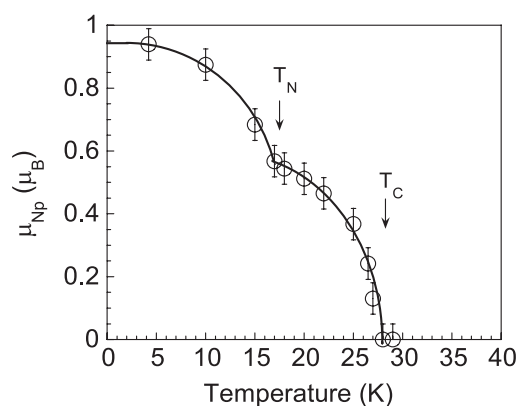


Figure 6. Thermal dependence of the Np magnetic moment inferred from the ^{237}Np hyperfine magnetic field by the relation given in the text.

Np moments (1.71 and 1.49 for NpRhGa_5 [13] and NpNiGa_5 [14], respectively) from the Np^{3+} free ion value (1.75). These observations would be rather consistent with the use of crystal electric field (CEF) models of localized 5f electrons to explain the magnetic properties of the NpTGa_5 series. Recently, a doublet–singlet CEF-model including dipolar and quadrupolar intersite interactions was shown to be able to describe the main features of the magnetic orders in NpTGa_5 [23].

However, the low temperature ordered moments found experimentally (see table 2) are significantly lower than the one ($1.23 \mu_B$) calculated from the ground doublet wavefunction ($0.8733|3 > +0.4872|1 >$) [8] using the Np^{3+} Russel–Saunders Landé factor value (0.6). The 5f electrons probably have a dual nature in the NpTGa_5 series. Indeed, it was concluded from the de Haas–van-Alphen measurements that the 5f electrons contribute significantly to the conduction bands [17, 24, 25]. Band structure calculations assuming that all 5f electrons are itinerant should nonetheless be reconsidered, although they capture some of the experimental observations.

In contrast to the magnetic moments, the quadrupole coupling constant (e^2qQ) exhibits a rather large range of values (see table 2). According to the above mentioned doublet–singlet CEF model [23], it is expected that the 5f contribution to e^2qQ should be small ($\sim -1.3 \text{ mm s}^{-1}$ and -1.0 mm s^{-1} for the doublet and singlet respectively). Thus, the large negative e^2qQ values observed in NpFeGa_5 and NpRhGa_5 could be attributed mostly to the lattice contribution to the electric field gradient. This is in line with the increase of the local tetragonality $t = 1 - (2cz_{\text{Ga}}/a)$ [2] of the Np–Ga layers with the sequence $\text{Rh} > \text{Fe} > \text{Ni} \sim \text{Co}$ (a and c are the lattice parameters and z_{Ga} the positional parameter of the Ga (4i) site) (see table 1).

4. Discussion

NpNiGa_5 exhibits unique behaviours within the AnTGa_5 family.

- From the magnetic point of view, it is the only ferromagnet of the series and its low-temperature antiferromagnetic component displays a propagation wavevector $\mathbf{k} = [1/2 \ 1/2 \ 1/2]$ unusual in AnTGa_5 compounds (it was found only in UNiGa_5).
- From the electronic point of view, it displays the highest γ -value among UTGa_5 and NpTGa_5 compounds, comparable to the specific heat coefficients observed in

superconducting PuTGa₅ compounds, and it is isoelectronic to PuCoGa₅ and PuRhGa₅ with a valence electron count amounting to the ‘magic’ number 32 [6]. Lastly, unlike in NpCoGa₅ [24] and NpRhGa₅ [25], no cylindrical Fermi surface was observed in NpNiGa₅ [17].

The possible link between the magnetic wavevector $\mathbf{k} = [0\ 0\ 1/2]$ observed in the antiferromagnetic NpCoGa₅ and NpRhGa₅ compounds and the occurrence of magnetically mediated superconductivity [26] in the Pu analogues has been recently pointed out [12]. Thus, by comparison with its isostructural compounds, one may speculate that the electronic properties of NpNiGa₅ would qualify it as a potential superconductor, but its magnetic properties are not suitable for the formation of Cooper pairs. Moreover, it was shown, although the theoretical Fermi surfaces of NpNiGa₅ and PuCoGa₅ are quite similar, that their two-dimensionality in NpNiGa₅ becomes worse compared with those of PuCoGa₅. This would be another reason why NpNiGa₅ is not superconducting [27].

5. Conclusion

We have shown by magnetization, specific heat and ²³⁷Np Mössbauer spectroscopy that NpNiGa₅ undergoes a double magnetic transition at $T_C \approx 30$ K and $T_N \approx 18$ K. The latter transition is accompanied by a jump of the Np magnetic moment, which saturates at a value of $0.94 \mu_B$, in good agreement with recent neutron diffraction data. In contrast to NpCoGa₅ and NpRhGa₅, the vanishing quadrupolar interaction in NpNiGa₅ prevents us from gaining any information concerning the Np moment directions. From the observed trend of the isomer shift along the NpTGa₅ series it is suggested that the tendency to localization increases with the number of d electrons of the transition metal T. The nature of the 5f electrons still remains controversial. Localized as well as itinerant models were invoked to explain the experimental work. The search for crystal field excitations in NpCoGa₅ is foreseen in the near future. If successful, this would be a strong argument in favour of localized models. But it is expectable that the 5f electrons in the NpTGa₅ compounds could have a dual nature, as shown in UPd₂Al₃ [28] and by photoemission in the PuCoGa₅ superconductor [29].

Acknowledgments

The authors are grateful to D Aoki, Y Homma, F Honda, K Kaneko, Y Haga and N Metoki for fruitful discussions. The high purity Np metals required for the fabrication of the compound were made available through a loan agreement between Lawrence Livermore National Laboratory and ITU, in the frame of a collaboration involving LLNL, Los Alamos National Laboratory and the US Department of Energy.

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