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A Mössbauer study of the intermetallic compound Np₄Ru₇Ge₆

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Received 28 September 1995, in final form 29 February 1996

Abstract. An extensive ²³⁷Np Mössbauer study of the new compound Np₄Ru₇Ge₆ is reported. This material reveals only a purely axially symmetric quadrupole interaction down to 4.2 K, and its isomer shift suggests the occurrence of a Np³⁺ charge state. A careful analysis of the intensities of the hyperfine lines shows the presence of an absorber thickness effect, and an Einstein-type model fits the temperature dependence of the isotropic lattice vibrations well. The non-magnetic ground state inferred from the in-field measurements may be understood within the Kondo lattice/RKKY model. The observation of an enhanced susceptibility at low temperature may indicate a heavy-fermion behaviour.

1. Introduction

The discovery of uranium intermetallics which exhibit heavy-fermion (HF) behaviour, i.e. an enhanced value of the coefficient γ of the electronic specific heat, has led to an exponentially increasing amount of work on uranium systems in the last decade [1, 2]. In these studies, most attention has been paid to the UT_2Si_2 compounds (T is an *n*d transition metal) which crystallize with the body-centred ThCr₂Si₂-type structure, the intermetallics of special interest being those showing the presence of 'magnetic' electrons coexisting with a superconducting state. In this respect URu₂Si₂ aroused much interest as the first HF system in which coexistence of antiferromagnetism ($T_N = 17.5$ K) and superconductivity $(T_{SC} = 0.8 \text{ K})$ was established [3]. This peculiar and still puzzling behaviour initiated the search for new HF uranium systems as well as an extension to neptunium intermetallics which are isostructural to uranium compounds with HF properties. The extensive researches directed towards these ends permitted the characterization of the UPd₂Al₃ HF system [4] $(T_N = 14.5 \text{ K}, T_{SC} = 2 \text{ K})$ with U carrying a substantial ordered moment of 0.85 μ_B . This is in contrast to the case for URu₂Si₂ where the U moment of only 0.02 μ_B is considerably below the U³⁺ and U⁴⁺ free-ion values of $\sim 3 \mu_B$. So far only few HF compounds containing Np are known of. The best-established example is NpBe₁₃ ($T_N = 3.4$ K), but no superconductivity was detected down to 80 mK [5]. It is worth noticing that neither NpRu₂Si₂ [6] nor NpPd₂Al₃ [7], which are complex antiferromagnets, present properties characteristic of HF systems; however, specific heat data are not yet available. In the course of the study of the formation of ternary neptunium transition metal germanides some of us discovered the existence of the compound Np₄Ru₇Ge₆ [8]. It is isostructural to U_4 Ru₇Ge₆ [9]. The physical properties of $U_4Ru_7Ge_6$ investigated in detail by the Leiden group [10]

0953-8984/96/234259+10\$19.50 © 1996 IOP Publishing Ltd

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indicated that this compound exhibits interesting characteristics. Indeed, U₄Ru₇Ge₆ can be classified as a dense Kondo lattice which orders ferromagnetically at $T_C = 6.8$ K with small ordered moments of 0.2 μ_B per uranium atom. These findings prompted us to examine the physical properties of the parent Np₄Ru₇Ge₆ compound.

In the present paper we report on extensive 237 Np Mössbauer measurements on Np₄Ru₇Ge₆ with emphasis on gaining information on its electronic and magnetic properties, which are expected to shed light on the ground state of the Np ions.



Figure 1. The nearest-neighbour configuration of the Np atoms in $Np_4Ru_7Ge_6$, and the principal axis of the electric field gradient of axial symmetry.

2. Experimental procedure

The Np₄Ru₇Ge₆ sample was prepared by arc melting the pure constituents in stoichiometric ratio in a purified argon atmosphere. The sample was characterized by x-ray powder diffraction (Cu K α radiation) and the structural investigation was carried out on a small single crystal isolated from the fragmented button by using an Enraf–Nonius CAD fourcircle diffractometer with Mo K α radiation ($\lambda = 0.71073$ Å). The x-ray diffraction did not reveal any impurity phase, and the diffractogram showed that Np₄Ru₇Ge₆ is isotypic with U₄Ru₇Ge₆ which crystallizes in the U₄Re₇Si₆ type of cubic symmetry (space group $Im\bar{3}m$) [8, 11]. Figure 1 shows that the neptunium atoms are surrounded by a cuboctahedron formed by six Ru(1) atoms, all in the same plane, and six Ge atoms. The lattice parameter a = 8.3083(9) Å is comparable to that found for U₄Ru₇Ge₆ (8.293 Å, [10]) and for $U_4Os_7Ge_6$ (8.302 Å, [9]). Notice that the interactinide distance, equal to a/2, is well above the Hill limit, ~3.5 Å and ~3.2 Å for U and Np, respectively. Thus, 5f delocalization cannot proceed via direct f-f overlap.

The ²³⁷Np Mössbauer measurements using the 59.5 keV gamma-ray transition between I = 5/2 excited and ground states were performed on a powder absorber with a thickness of 102 mg Np cm⁻². The Mössbauer source of 108 mCi of ²⁴¹Am metal was kept at 4.2 K and the temperature of the absorber was varied from 4.2 K up to 104 K. The in-field experiments were carried out by application of a uniform magnetic field up to 8 T normal to the plane of the absorber and along the direction of propagation of the gamma rays. The spectra were recorded with a sinusoidal drive using conventional methods, and the 59.5 keV gamma rays were detected with an intrinsic Ge detector.



Figure 2. The ²³⁷Np quadrupole interaction spectrum of Np₄Ru₇Ge₆ at 4.2 K. Curve a represents a fit with the line intensities proportional to the Clebsch–Gordon coefficients. For curve b the data were fitted allowing deviations from the Clebsch–Gordon intensities which could be accounted for by assuming either anisotropic lattice vibrations or absorber thickness effects.

3. Results and discussion

3.1. ²³⁷Np Mössbauer experiments in zero applied magnetic field

The 4.2 K spectrum of Np₄Ru₇Ge₆ presented in figure 2 reveals a well resolved pure quadrupole interaction pattern with zero asymmetry parameter η , in agreement with the local symmetry of the neptunium atoms (figure 1). A close examination of the spectral shape shows, however, deviations in the intensities of certain hyperfine components from those predicted by the Clebsch–Gordon coefficients (figure 2, curve a). The analysis of the line positions yields a quadrupole coupling constant $|e^2qQ| = 69.4(2) \text{ mm s}^{-1}$ (or 3332 MHz) and an isomer shift δ_{IS} of $-8.3(5) \text{ mm s}^{-1}$ relative to NpAl₂. The principal component of the electric field gradient (V_{zz}) inferred from the e^2qQ -value amounts to $+3.36 \times 10^{22} \text{ V m}^{-2}$. Notice that its positive sign can only be deduced from the in-field Mössbauer measurements (see below). The above-mentioned intensity anomalies indicate partial saturation of the $\Delta m = 0$ transitions. Such a behaviour could originate from either preferential orientation of crystallites, anisotropic lattice vibrations (the Goldanskii–Karyagin effect, GKE), or the absorber thickness effect [12, 13] (figure 2, curve b).



Figure 3. ²³⁷Np quadrupole interaction spectra for Np₄Ru₇Ge₆ at different temperatures. The solid curves represent fits with the transmission integral formalism without any GKE.

The ²³⁷Np quadrupole interaction spectra for Np₄Ru₇Ge₆ recorded at different temperatures ranging from 4.2 K up to 104 K are presented in figure 3. An analysis of the data shows that: (i) e^2qQ is almost *T*-independent, e.g. it amounts to 69.0(2) mm s⁻¹ at 104 K; and (ii) the deviations from Clebsch–Gordon intensities decrease with increasing temperatures and are vanishingly small in the 104 K spectrum. This latter behaviour is an indication for the absence of a GKE in Np₄Ru₇Ge₆. Indeed, if one assumes an isotropic thermal expansion of the lattice, the anisotropy of the lattice vibrations is expected to increase when raising the temperature. The smallness or even the absence of a GKE in Np₄Ru₇Ge₆ is further supported by the transmission integral fits discussed below. Moreover our data indicate that texture effects do not play a significant role.

Therefore, the absorber thickness effect might be responsible for the intensity discrepancies observed in the quadrupolar spectra. Before considering this mechanism we will first discuss the information on the lattice dynamics gained from the temperature dependence of the spectral area, which is proportional to the Lamb-Mössbauer factor f(T). Its thermal variation, f(T)/f(4.2 K) versus T, is represented in figure 4. The f-factor



Figure 4. The temperature dependence of the Lamb–Mössbauer factor for Np₄Ru₇Ge₆: full circles: experimental data; continuous line: the theoretical curve obtained using an Einstein-type model for the phonon spectrum with $\theta_E = 130$ K; dashed lines: theoretical curves obtained using Debye-type phonon dispersion laws with θ_D ranging from 320 K (upper curve) to 240 K (lower curve) in steps of 20 K.

or resonant fraction has a form quite analogous to the well known Debye-Waller factor $f = \exp\{-k^2 \langle x^2 \rangle\}$. In practice one usually evaluates the resonant fraction in terms of either the Debye or Einstein models for the phonon dispersion laws [14]. The Debye model is better suited to probing the acoustic phonon branch while the Einstein model is often used to approximate part of the phonon spectrum, particularly the contribution of optical phonons of frequency ω_E . The different theoretical curves displayed in figure 4 indicate that the thermal variation of f(T)/f(4.2 K) is better accounted for by an Einstein-type law with $\hbar\omega_E = k_B \theta_E = 130$ K than by Debye-type laws computed with θ_D ranging from 320 K to 240 K. This leads to an f-factor value of \sim 0.49 at 4.2 K which sounds reasonable. It should, however, be emphasized that the Einstein model, although representing rather satisfactorily the experimental results, is a very crude model for a real solid. The actual phonon frequency distribution $G(\omega)$ is much more intricate. Nevertheless our data suggest the occurrence of a sharp peak at ω_E in $G(\omega)$, pointing to the possible existence of soft optical phonons with energy ~11 meV. Similar results were recently found for YbN, and the existence of soft optical phonons was demonstrated by neutron inelastic experiments [15]. It is worth mentioning here that the analysis of the specific heat data of the parent $U_4Ru_7Ge_6$ compound, in the temperature range 14–30 K, yields a Debye temperature $\theta_D \approx 276$ K [10]. This is in good agreement with the data of figure 4 when considering only the lowtemperature region (T < 60 K).

The effective absorber thickness of a Mössbauer absorber is generally defined by the expression [16]

$$T_A = n\sigma_0 f a. \tag{1}$$

Here *n* is the number of atoms per unit area of the Mössbauer element and *a* is the isotopic abundance (=1 in our case); σ_0 is the maximum resonance cross section and *f* the Lamb-Mössbauer factor. The expression for T_A in equation (1) assumes the natural linewidth for the resonance absorption cross section. For the ²³⁷Np resonance the thickness parameter

has to be reduced by the ratio between the true absorber linewidth ($\Gamma_a \simeq 1.5 \text{ mm s}^{-1}$) and the natural width ($\Gamma_0 \simeq 0.07 \text{ mm s}^{-1}$). Thus equation (1) becomes

$$T_A = n\sigma_0 f \frac{\Gamma_0}{\Gamma_a}.$$
(2)

 T_A can be computed at different temperatures since the resonant fraction f(T) can be evaluated, as shown above, in terms of the Einstein model:

$$f(T) = \exp\left\{-\frac{E_R}{k_B \theta_E} \left(\frac{2}{e^{\theta_E/T} - 1} + 1\right)\right\}.$$
(3)

Here E_R is the recoil energy associated with the absorption process of the 59.5 keV gamma rays. Using $k_B \theta_E = 130$ K and equations (2) and (3), T_A was found to range between 1.87 and 1.05 for T = 4.2 K and 104 K, respectively.

The quadrupole interaction spectra of ²³⁷Np in Np₄Ru₇Ge₆ were then analysed taking into account the influence of the absorber thickness on the lineshape. For this purpose we used the so-called transmission integral (TI) formalism [17]. The solid curves in figure 3 represent the fits obtained with the TI analysis. They reproduce perfectly the experimental data and give a straightforward explanation for the line intensity discrepancies observed when a standard analysis is used. It should be emphasized that we used a *self-consistent fitting procedure*; the *T_A*-values were fixed to the values computed from equation (2) and the free parameter proportional to the resonant fraction of the source was found to be independent of the temperature of the absorber, as expected.



Figure 5. 237 Np Mössbauer spectra for Np₄Ru₇Ge₆ taken at 4.2 K with applied magnetic fields.

3.2. ²³⁷Np Mössbauer experiments in an applied magnetic field—the ground state of the Np ions

As shown above, $Np_4Ru_7Ge_6$ reveals, at least down to 4.2 K, only a pure quadrupole interaction. This indicates that the compound is either a paramagnet with an onset of

magnetic order below 4.2 K or that the Np ions do not carry a local magnetic moment. In order to test the presence (or absence) of a permanent magnetic moment we performed measurements in applied fields of 5 T and 8 T. Figure 5 shows that application of an external field produces essentially a broadening and distortion of the lineshapes. This already indicates that the quadrupole interaction is dominant. Best fits to the data were obtained by assuming the effective hyperfine field (H_{eff}) to be parallel to the applied field (H_{ext}) which is along the gamma-beam direction, and the principal component V_{zz} of the electric field gradient (fixed to its zero-field value) to be positive and randomly directed with respect to the direction of H_{eff} . The solid curves in figure 5 result from the superposition of ten subspectra corresponding to the different angles α between the directions of H_{eff} and V_{zz} . The relative weight of each subspectrum is proportional to sin α [18].

The effective magnetic field $H_{eff} = H_{hf} + H_{ext}$ (the demagnetizing field is negligible) was found to be 9.8(2) T and 15.4(2) T, for H_{ext} equal to 5 T and 8 T respectively. This indicates that the hyperfine field H_{hf} is proportional to the applied field. It is worth noticing that our best-fit model, with H_{eff} and H_{hf} directed along H_{ext} (taken as the zdirection), rules out any strong anisotropy of H_{hf} which would result in a distribution of the effective magnetic field [19]. Using the well established relationship ($H_{hf}/m = 215 \text{ T}/\mu_B$) [20] between the Np hyperfine field and the Np local magnetic moment, which are both proportional to $\langle J_z \rangle$, one deduces a local moment of ~0.034 μ_B for $H_{ext} = 8$ T which corresponds to a local susceptibility of 2.4×10^{-3} emu per mole of Np at 4.2 K. It is worth noticing that the proportionality constant is valid even for itinerant and strongly hybridized 5f systems. Furthermore, it was shown in many instances that the so-deduced local susceptibility compares well with the bulk susceptibility evaluated from magnetization measurements [21–23]. It may thus be concluded that Np₄Ru₇Ge₆ is a 'non-magnetic' compound, i.e. that the Np ions do not carry a permanent magnetic moment.

Additional information on the ground state of Np₄Ru₇Ge₆—although it is intricate in conducting systems—is provided by the assignment of a charge state to the Np ions via the isomer shift value. Actually δ_{IS} for Np₄Ru₇Ge₆ (-8.3(5) mm s⁻¹) lies between the values corresponding to the most ionic Np³⁺:5f⁴ (+37 mm s⁻¹) and Np⁴⁺:5f³ (-9 mm s⁻¹) compounds [24, 25]. But, from the isomer shift systematics in conducting materials, it was shown that the presence of conduction electrons always increases the electron density at the nuclear origin compared to its free-ion value and shifts δ_{IS} towards the next higher charge state (e.g. $Np^{3+} \rightarrow Np^{4+}$). This means that the shift observed in a conducting compound will always be more negative when compared to that in an insulating material containing Np in the same ionic form. An additional and most informative clue as to the electronic structure of the Np ions was drawn from a plot of the hyperfine field versus the isomer shift for various series of intermetallic compounds [25, 26]. The observation of monotonic correlations between H_{hf} and δ_{IS} , even when the latter crosses the Np⁴⁺ ionic line, leads to the claim that an isomer shift larger than -9 mm s^{-1} (the isomer shift of fully ionic Np⁴⁺) may correspond to a compound containing trivalent neptunium. According to these systematics a Np³⁺ charge state was tentatively assigned to the Np ions in Np₄Ru₇Ge₆. Its isomer shift represents the low end of the large range of possible values for Np³⁺ ions in conducting systems. It is tempting to assign the large shift of δ_{IS} towards the Np⁴⁺ free-ion value to a delocalization of the 5f electrons through f-d or f-p hybridization which acts in the same direction as that for conduction electrons of s character. Indeed, the contribution of one 7s electron was estimated to be slightly less than the screening effect of one 5f electron. It must, however, be emphasized that δ_{IS} ranges from -2.3 up to +20 mm s⁻¹ in the NpT₂Ge₂ or NpT₂Si₂ series of intermetallic compounds, although the 5f electrons, as shown by high-pressure measurements, were considered to be well localized [25, 26].

In any case, one should be aware that the notion of a definable charge state is somewhat dubious in hybridized systems.

The other hyperfine interaction parameter related to the electronic structure of the Np ions is the electric field gradient (efg). It is customary to express the efg as a sum of two terms [26]: a lattice contribution arising from the ionic charges of the lattice, and a local contribution due to the non-spherical charge distribution of the electrons near the probe site. For a non-S-state ion like Np³⁺, the local efg is caused by the open 5f shell as well as by the asphericity of the local charge distribution of conduction electrons and electrons involved in covalent bonds through hybridization effects with ligand orbitals. The evaluation of the non-f electronic contribution requires rather elaborate band-structure calculations [27]. In contrast, the efg due to the 5f shell can be evaluated if the wavefunctions and energies of the thermally populated crystal-field levels, characterized by different values of J_z , are known. The 5f efg is proportional to the thermal average $(3J_z^2 - J(J+1))_T$. For $J_z = 4$, the Np³⁺ free-ion value, it amounts to -1.32×10^{22} V m⁻². The lattice contribution can be computed using the point charge model but its application to metallic systems raises the question of the effective charge to be assigned to the various ions in the lattice. For a polycrystalline sample subjected to an external magnetic field, both the lattice efg and the contribution of non-f electrons are expected to be randomly oriented with respect to H_{ext} . In contrast, owing to the isotropic behaviour of the induced hyperfine field $(\propto \langle J_z \rangle)$ it is expected that the associated 5f efg ($\propto (3J_7^2 - J(J+1))$) will align along H_{ext} too. As shown above, the in-field data are well represented assuming that the principal component of the efg is random with respect to H_{ext} . Thus, we may conclude that the 5f contribution to the efg is vanishingly small, i.e. that the measured V_{zz} of $+3.36 \times 10^{22}$ V m⁻² is essentially due to the lattice and to the contribution of the non-f electrons, in agreement with its nearly temperature-independent behaviour. It is worth noticing that comparable values of V_{zz} were measured in Gd intermetallics where the f contribution to the efg is negligible. The observed large values of V_{zz} were in part attributed to the asphericity of the Gd charge distribution [28].

The occurrence of a non-magnetic ground state for the Np ions in Np₄Ru₇Ge₆ is at first sight surprising, as the Np compounds were shown to be generally more magnetic than their uranium counterparts ($U_4Ru_7Ge_6$ is magnetically ordered!) Nevertheless this is not a unique situation. Recently we have found that Np₂Rh₂Sn is non-magnetic although U₂Rh₂Sn exhibits magnetic order at low temperature [29]. One possible reason for this behaviour is the possible existence of a singlet crystal-field ground state for the Np^{3+} ions (5f⁴, ⁵I₄) with the restriction that the ground-state wavefunction of the 5f shell: $|5f\rangle = a_0|J\rangle + a_1|J-N\rangle + a_2|J-2N\rangle + \dots$, from which one deduces $J_z^2 = a_0^2 J^2 + a_1^2 (J-N)^2 + a_2^2 (J-2N)^2 + \dots$, should fulfil the condition $(3J_z^2 - J(J+1)) \cong 0$. This model is, however, unable to explain the enhanced susceptibility observed at 4.2 K. Another more straightforward explanation which may be invoked is the Kondo effect which is able to screen or even destroy the local Np moment [1]. This implies that $Np_4Ru_7Ge_6$ should be classified among the Kondo lattice systems. The ground-state properties of these systems are dictated by the strength of the exchange interaction J_f between localized 5f electrons and conduction electrons and thereby the competition between the intersite (RKKY) and intrasite (Kondo) interactions with characteristic temperatures $T_{RKKY} \propto J_f^2$ and $T_K \propto \exp\left(-1/|J_f|\right)$ respectively. This delicate balance is theoretically described by Doniach [30] in a magnetic phase diagram depending on J_f . Above a critical value $(J_f)_c$ of the exchange interaction the Kondo effect dominates and leads to a non-magnetic ground state. In contrast if $J_f < (J_f)_c$, long-range order with somewhat reduced moments develops at low temperatures. The heavy-fermion state is observed close to the critical region,

i.e. close to a magnetic instability. The Doniach model has already been considered to explain the reduced moment observed for U₄Ru₇Ge₆, a moderately HF compound [10], and the absence of magnetic order in U₄Os₇Ge₆ [9, 10]. In the latter compound, with larger unitcell volume, the Kondo effect dominates the RKKY interaction and causes a non-magnetic ground state. The measurement of a large susceptibility of about 2.4×10^{-3} emu/mol Np at 4.2 K, roughly 1000 times that of simple metals, suggests that Np₄Ru₇Ge₆ is close to a magnetic instability, i.e. in a HF regime. Indeed, according to the Jones plot [31], which displays on a log–log scale the low-temperature coefficient of the specific heat (γ) versus the low-temperature susceptibility (χ (0)) for a number of mixed-valence and heavy-fermion materials, it may be anticipated that γ could reach 200 mJ K⁻²/mol Np. Notice that an even larger γ -value is found if one accepts the linear correlation between the isomer shift δ_{IS} and the γ -value presented for some heavy-fermion Np intermetallics [32]. At this stage Np₄Ru₇Ge₆ appears to be a very suitable candidate for further studies aimed at searching for HF behaviour.

4. Conclusion

In the course of the study of ternary neptunium-transition metal germanides a new intermetallic phase, Np₄Ru₇Ge₆, isotypic with the cubic U₄Ru₇Ge₆ compound was characterized. Np4Ru7Ge6 reveals at low temperatures Mössbauer patterns with well resolved axially symmetric quadrupole interaction. No onset of magnetic interaction could be detected down to 4.2 K. The observed deviations in the intensities of certain hyperfine components from those predicted by the Clebsch-Gordon coefficients were shown to arise from the absorber thickness effect. A possible existence of soft optical phonons was deduced from the temperature dependence of the Lamb-Mössbauer factor which can be accounted for within an Einstein model. Application of an external field of 8 T induces a Np moment of only 0.034 μ_B at 4.2 K and indicates that the Np³⁺ ions in Np₄Ru₇Ge₆ are in a non-magnetic ground state. This is in contrast with the counterpart U4Ru7Ge6 which orders ferromagnetically, with a reduced moment, at low temperature. The non-magnetic ground state of $Np_4Ru_7Ge_6$ as well as the behaviour of $U_4Ru_7Ge_6$ can be understood in terms of the competition between the Kondo effect and the RKKY exchange interaction, $Np_4Ru_7Ge_6$ being close to a magnetic instability. The enhanced susceptibility of $Np_4Ru_7Ge_6$ measured at 4.2 K points towards this interpretation; the use of the Jones plot suggests a γ -value of about 200 mJ K⁻²/mol Np. Future work on Np₄Ru₇Ge₆ should concentrate on resistivity measurements, including high-pressure experiments (Np₄Ru₇Ge₆ is expected to become magnetic under pressure) and on specific heat data. In addition, the study of the (U/Np)₄Ru₇Ge₆ solid solutions should shed some light on the development of the heavyfermion state.

Acknowledgments

The high-purity Np metal required for the fabrication of this compound was made available through a loan agreement between Lawrence Livermore National Laboratory and EITU, in the framework of a collaboration involving LLNL, Los Alamos National Laboratory, and the US Department of Energy.

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