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The time differential perturbed angular correlation study of the Ni–5 at.% Hf alloy

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ABSTRACT

The time differential perturbed angular correlation technique was used to observe the magnetic dipole and electric quadrupole interactions of ^{181}Ta probe in the Ni–5 at.% Hf alloy from 78 to 1131 K. The presence of the Larmor precession frequency of ^{181}Ta in the Ni(Hf) solid solution phase confirms the Ni dominant concentration for the onset of ferromagnetism in the Ni–Hf alloys. A very weak quadrupole interaction originates from imperfections in the $HfNi_5$ phase formed during melting procedure. The third observed quadrupole interaction corresponds to the Ta probe in the HfO_2 contamination due to internal oxidation of the sample.

1. Introduction

The Ni-based alloys are formed in various binary and ternary systems. Recently, these alloys have come to the focus of numerous investigations due to their wide application characteristics, very high strength at high temperature and superior qualities: smaller grain structure, increased strength and corrosion resistance and increased stability of weld joints. The hyperfine interactions of ^{181}Ta probe in the nickel–5 at.% hafnium alloy were observed using the time differential perturbed angular correlation (TDPAC) method in the temperature range 78–1131 K.

2. Experimental

The sample preparation and the structure analysis of the nickel–5 at.% hafnium alloy employing X-ray diffraction (XRD), scanning electron microscopy (SEM) and extended X-ray absorption fine structure spectroscopy (EXAFS) are described in refs. [1,2]. These earlier investigations on structural properties and alloy formation in the Ni–5 at.% Hf alloy showed that the composition of this alloy is in accordance with the Ni–Hf phase diagram [3]. The TDPAC apparatus and annealing procedure of the polycrystalline Ni–Hf alloy are described in ref. [4]. Two forms of perturbations by static interactions of ^{181}Ta have been encountered during the investigation: pure magnetic dipole interaction (MDI) and pure electric quadrupole interaction (EQI). The magnitude of EQI at the ^{181}Ta site is described with the electric quadrupole frequency $\omega_Q = (eQV_{zz})/[4I(2I-1)\hbar]$, and with the frequency distribution width δ describing a broadening of the electric field gradient (EFG) due to the lattice imperfections. The principal axes of the EFG tensor are defined in such a way that V_{zz} is the largest component of the EFG tensor and $\eta = (V_{xx} - V_{yy})/V_{zz}$ is its asymmetry parameter. $Q = 2.36 \times 10^{-24}$ cm^2 is the quadrupole moment for the ^{181}Ta intermediate level $I = 5/2$ of the measured γ–γ cascade. The strength of the MDI corresponding to the hyperfine magnetic field H_{hf} acting on ^{181}Ta probe is characterized by the Larmor frequency $\omega_L = g\mu_N H_{hf}/\hbar$, with the Lorentzian frequency distribution width parameter Λ. The least-square fitting procedure to the experimental TDPAC spectra was made using the DEPACK program [5].

3. Results and discussion

The selected TDPAC delayed coincidence spectra of ^{181}Ta probe in the Ni–Hf alloy are shown in Fig. 1. Previous SEM and XRD analysis of the nickel–5 at.% hafnium alloy revealed the presence of two phases: the Ni(Hf) solid solution and the defective $HfNi_5$ phases [1]. The majority of Hf atoms appeared to be incorporated in the $HfNi_5$ phase as demonstrated by EXAFS investigation on this alloy [1,2].

The magnetic dipole frequency of 541 Mrad/s at room temperature (RT), was found in the experimental spectra and it disappeared from the spectra above the Curie point of nickel. According to the known interaction frequency values for Ta impurities in nickel [4,6],

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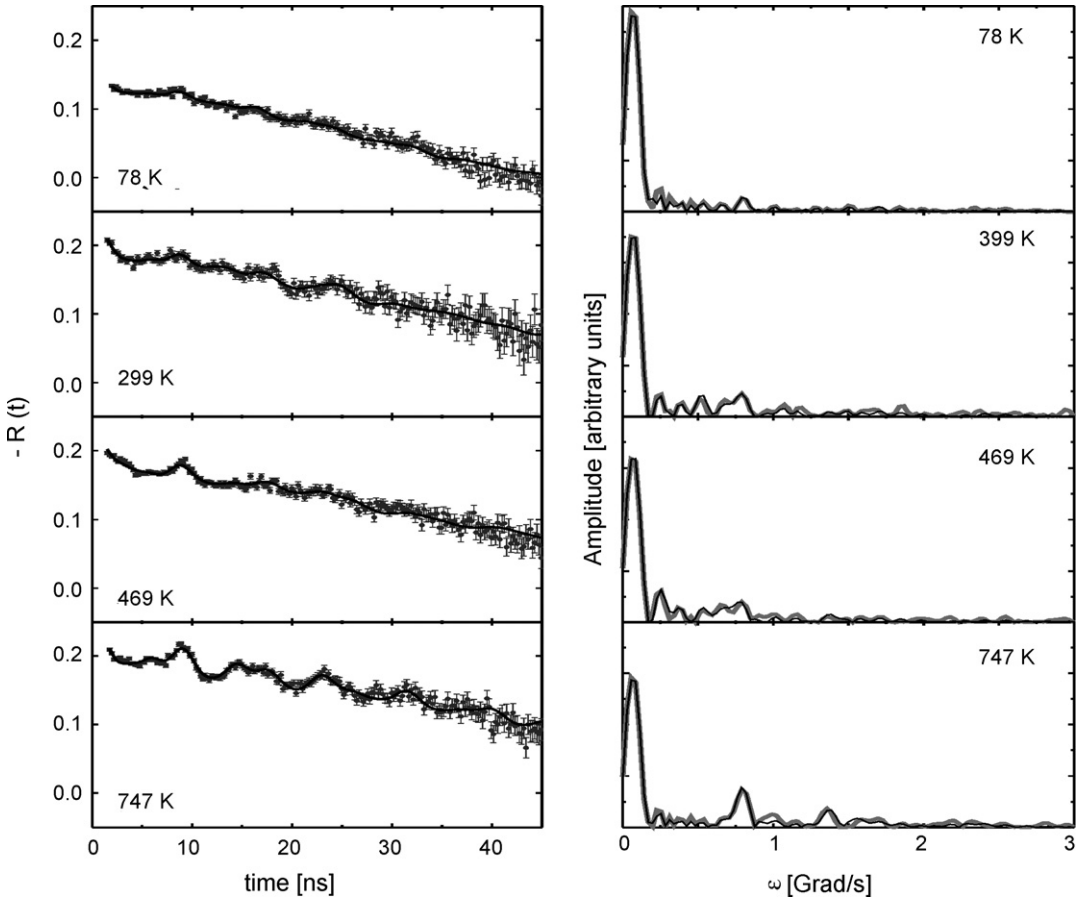

Fig. 1. TDPAC spectra of ^{181}Ta probe in the nickel–5 at.% hafnium alloy and their corresponding Fourier transforms (grey line—data, black line—fit).

it was assigned to the substitutional position of Ta in the ferromagnetic Ni(Hf) solid solution phase. The quadrupole frequency of 117 Mrad/s at 1131 K, was also evident in the experimental spectra above the Curie point of nickel. The best fit to the TDPAC experimental data on the nickel–5 at.% hafnium alloy (as it is similarly found for the nickel–2 at.% hafnium alloy [7]) revealed the existence of three hyperfine interactions: one MDI and two EQIs.

The parameters of the magnetic dipole interaction, ascribed to the probe site in the Ni(Hf) solid solution are given in Table 1. As expected, the hyperfine magnetic field H_{hf} is decreasing with increasing temperature. Existence of the Larmor precession frequency in the nickel–5 at.% hafnium alloy is in accordance with the work of Bakonyi et al. [8] where the limiting concentration of Ni for the onset of ferromagnetism in the Ni–Hf alloys is deduced to be about 89.5 at.% Ni.

Table 1
Relevant MDI parameters for ^{181}Ta probe in the Ni(Hf) solid solution phase in the nickel–5 at.% hafnium alloy.

T (K)	$\omega_L^{(1)}$ (Mrad/s)	Λ_1 (%)	H_{hf} (T)	f_1 (%)
78	603(15)	0(3)	9.68(26)	2(1)
299	541(7)	0(9)	8.69(11)	6(1)
362	480(6)	0(1)	7.71(9)	5(1)
469	295(6)	13(3)	4.74(10)	13(2)
492	238(8)	21(6)	3.83(13)	16(2)
296	539(5)[a]	0(3)	8.66(9)	5(1)

[a] Remeasured at RT after the whole temperature range has been completed.

On the other hand, the significant fraction of Ta atoms (88% at 78 K) experienced weak EQI (3.5 Mrad/s at 78 K) with anomalous temperature dependence (Fig. 2). Regarding our earlier EXAFS and XRD results on this alloy [1,2], it was attributed to the majority of Ta atoms in the paramagnetic $HfNi_5$ phase having the defect

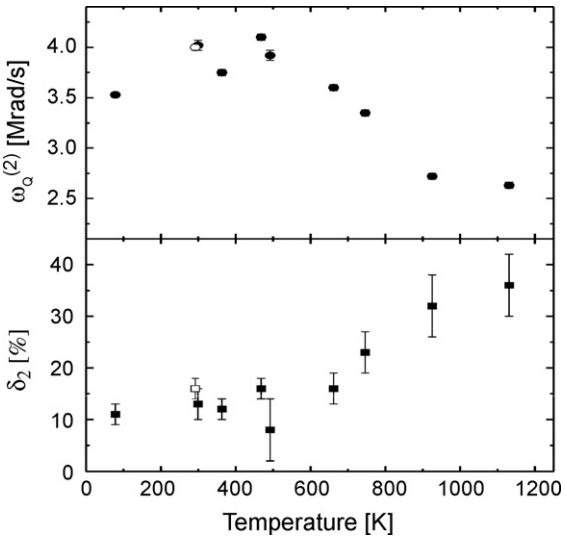

Fig. 2. Temperature dependence of the electric quadrupole frequency $\omega_Q^{(2)}$ and frequency distribution width δ_2 for ^{181}Ta in the defective $HfNi_5$ phase in the nickel–5 at.% hafnium alloy; (□) remeasured at RT after the whole temperature range has been completed.

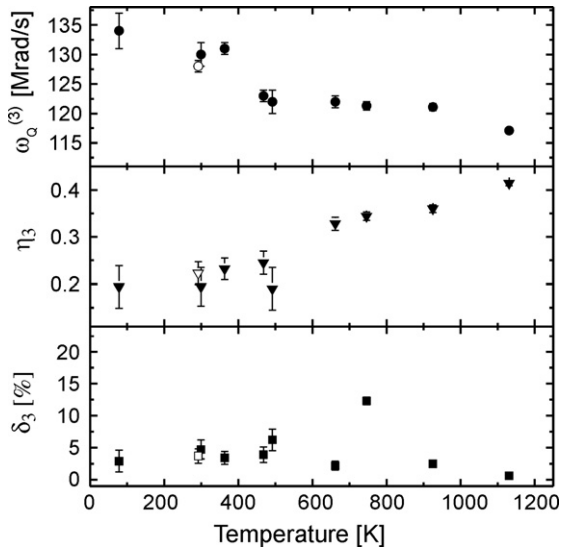

Fig. 3. Temperature dependence of the EQI parameters for ^{181}Ta in the HfO_2 contamination in the nickel–5 at.% hafnium alloy; (□) remeasured at RT after the whole temperature range has been completed.

cubic structure. The temperature independent EFG asymmetry parameter $\eta_2 \approx 1$ fitted well the experimental spectra in the whole measured range. The similar was observed for the nickel–2 at.% hafnium alloy [7].

The third hyperfine interaction with quadrupole frequency of 117 Mrad/s at 1131 K, originated from the dielectric HfO_2 contamination. The measured hyperfine parameters are presented in Fig. 3, and are in very good agreement with the known one for ^{181}Ta in the monoclinic HfO_2 [9,10]. The internal oxidation of Hf-intermetallic alloys may occur at high temperatures, $T > 900$ K, as seen in the work of Bedi and Forker [11].

4. Conclusions

The TDPAC experimental data revealed that the ^{181}Ta probe is situated at three nonequivalent crystallographic Hf sites in the nickel–5 at.% hafnium alloy: Hf substituting in the nickel lattice, $HfNi_5$ and HfO_2 phases. The magnetic dipole interaction of Ta in the Ni(Hf) solid solution exhibited decrease with increasing temperature. The weak electric quadrupole interaction of Ta in $HfNi_5$ originated from imperfections in its cubic structure. The high quadrupole frequency found for Ta in HfO_2 confirmed the oxidation process in the sample.

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