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Hyperfine Interactions of ^{181}Ta in Zr_2Ni Observed Using PAC Spectroscopy

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Abstract. We have measured nuclear electric–quadrupole interactions (EQI) at ^{181}Ta impurities substituted as Hf atoms into the Zr site in Zr_2Ni. Using perturbed-angular-correlation (PAC) spectroscopy, we measured the EQI over temperatures ranging from 10 to 1200 K. Over the entire range of temperature, the Zr_2Ni crystal has a bct Al_2Cu structure that includes a single Zr site. The crystal field symmetry surrounding this site is rather low, giving rise to a highly asymmetric electric-field gradient tensor. At 10 K, the EQI is characterized by an angular frequency $\omega_0 = 601(3)$ Mrad s^{-1}, and an asymmetry parameter $\eta = 0.835(2)$. At 1200 K, ω_0 decreases to 516(3) Mrad s^{-1}, and η also decreases to 0.790(4). Although weak, the temperature dependence of ω_0 is consistent with a $(1 - BT^{3/2})$ power law, in which $B = 6 \times 10^{-6}$ K$^{-3/2}$. The EQI also manifests a very narrow linewidth. We observed no evidence either for magnetic ordering or for structural phase transitions in the temperature range covered by this experiment. Moreover, the sharpness of the EQI indicates that the samples as prepared are remarkably free of strain and defects. These results indicate that the Zr_2Ni structure does not promote the formation of defects and that the power-law dependence of ω_0 on T is insensitive to the asymmetric nature of the crystal.

Key words: perturbed angular correlations, electric–quadrupole interactions, ^{181}Hf, defect structure of Zr alloys.

1. Introduction

In contrast to ordinary alloys, binary intermetallic compounds such as Zr_2Ni consist of crystals having two distinct sublattices, and thus exhibiting long-range chemical order. The atomic-scale environments around the atoms occupying these sublattices often differ significantly, because these structures usually have low symmetry. A consequence of this long-range order and low symmetry is that the physical properties of these compounds are potentially very sensitive to the presence of defects, foreign atoms, and deviations from stoichiometry. These effects have been the subject of several recent investigations [1–5].

Because intermetallic crystals have much lower symmetry than the crystals of pure metals and of common binary alloys, the question arises concerning a possible correspondence between the features of the crystal structure and corresponding

space group, and the tendency for defects to form in the crystal. For example, in the Laves-phase magnetic intermetallic, $ZrFe_2$, we have observed effects of Fe antisite defects near Zr sites that enhance the transferred magnetic hyperfine field at ^{181}Ta impurity probes residing on Zr sites [5]. Likewise, in a recent investigation [6] on the intermetallic Zr_3Fe, the electric-field-gradient (EFG) parameters show different temperature dependences for the two Zr-sites, and a band-theory calculation of these parameters shows significant deviation from the experimental measurements.

To investigate the relationship between defects and crystal symmetry, we have measured the temperature dependence of the electric-field gradient at the Zr-site in the metallic compound Zr_2Ni. Zr_2Ni crystallizes in the $CuAl_2$ (C16) structure, which is body-centered tetragonal (bct) with a c/a ratio of 0.8671. For this purpose we used ^{181}Hf–^{181}Ta perturbed-angular-correlation (PAC) spectroscopy, and we exploit the chemical and metallurgical similarity of Zr and Hf in order to introduce the radioactive ^{181}Hf probe nuclei into the Zr site. Over the temperature range from 10 to 1200 K, we observed very sharp line shapes that indicate the presence of few point defects. Thus, the Zr_2Ni intermetallic crystal represents a paradigm of the so-called "perfect crystal", against which the features of other intermetallic crystals having different symmetries can be compared. Furthermore, the temperature dependence of the quadrupole interaction frequency, although weak, is consistent with a power-law of $T^{3/2}$.

2. Experiment

We prepared samples of the intermetallic compound Zr_2Ni by arc-melting high-purity Zr and Ni under an argon atmosphere. The pure metals were doped concurrently with 0.5 at.% Hf that had been neutron-irradiated to produce the ^{181}Hf tracer activity. Hf goes into the crystal substitutionally for Zr, on account of the chemical and metallurgical similarity of Zr and Hf. We observed γ–γ perturbed angular correlations (PAC) in the well-known 133–482-kev γ cascade in ^{181}Ta produced by the β^--decay of 43-d ^{181}Hf. We carried out measurements at 17 temperatures between 10 and 1200 K. Temperatures below laboratory temperature were produced using a closed-cycle helium refrigerator with a microprocessor-controlled temperature controller. The PAC spectrometer used in this temperature regime employed three BaF_2 scintillation detectors, with a time resolution of 0.7 ns fwhm. Above laboratory temperature, we used an alumina tube furnace with thermocouple temperature control, and a four-detector spectrometer that has two BaF_2 and two CsF scintillators with a time resolution of 0.9 ns fwhm. The data was analyzed using the DEPACK analysis program developed by Lindgren [7].

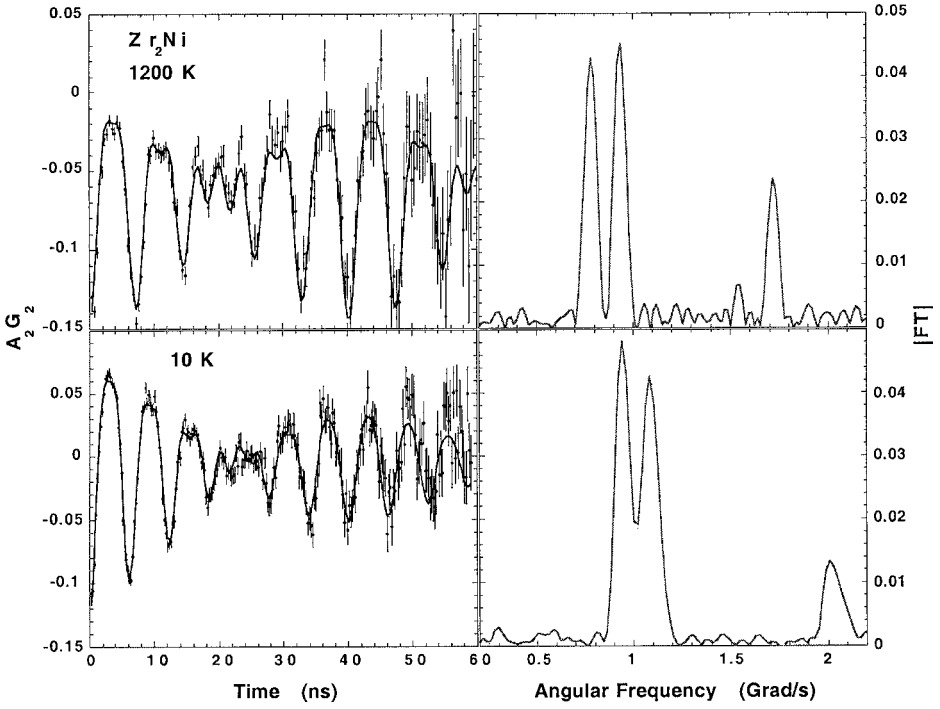

Figure 1. Measured PAC perturbation functions (*left*) and corresponding Fourier trasforms (*right*) for Zr_2Ni at 1200 K (*upper set*) and at 10 K (*lower set*).

3. Results and discussion

Representative PAC perturbation functions and corresponding Fourier transforms are shown in Figure 1. The observed perturbations show a unique electric–quadrupole interaction, characteristic of a single site for the Hf probe over the entire temperature range. The magnitude of the electric-field gradient tensor is relatively large for a metallic system, and possesses a large anisotropy. The linewidth found in all three samples is very narrow. The best fits are obtained using a Lorentzian frequency distribution function, given by $\exp(-\delta\omega_i t)$. The value of the Lorentzian linewidth parameter δ extrapolated to $T = 0$ K varies from 0.015(1) to 0.019(1), depending on sample. δ decreases smoothly to a value consistent with zero at temperatures above 1000 K. Over the entire temperature range there is no evidence either for magnetic order or for a change in crystal structure.

As seen in Figure 2, both the magnitude of the EFG as represented by the quadrupole angular frequency $\omega_0 = 6eQV_{zz}/4\hbar I(2I - 1)$, and its anisotropy decrease slowly with increasing temperature. The fitted curve in Figure 2(a) represents a functional dependence $\omega_0(T) = 601(2)(1 - BT^m)$ Mrad/s, where $B = 6(4) \times 10^{-6}$ K^{-m}, and the exponent $m = 1.42(11)$. The very weak temperature dependence of ω_0, and thus of the EFG, is consistent with the empirical $T^{3/2}$ power-law dependence on temperature found in many metals and alloys. However,

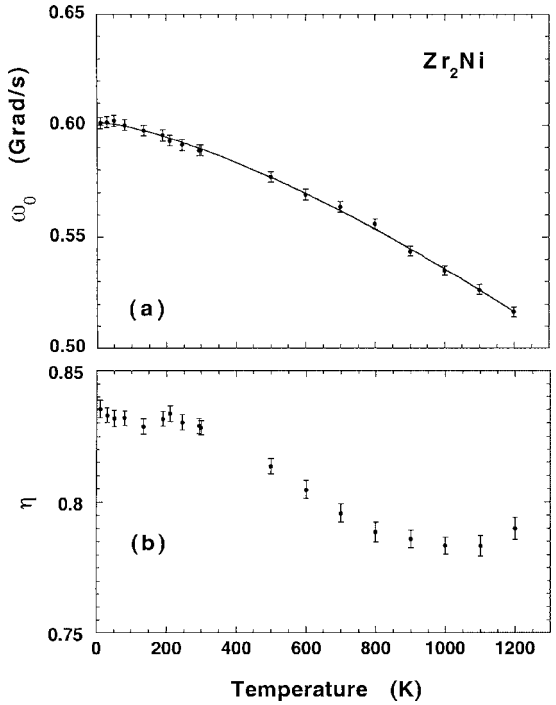

Figure 2. Temperature dependence of (a) electric–quadrupole interaction frequency and (b) asymmetry parameter for Zr_2Ni. The curve in (a) is a fit to the power law discussed in the text.

the magnitude of the constant B is considerably smaller than is typically found in metals and alloys. Although the empirical $T^{3/2}$ dependence on temperature appears to be followed in many systems, there is no general theory that supports it. Indeed, in some cases theoretical calculations suggest that this temperature dependence may be an approximation resulting from the dominance of different power laws in different temperature regimes, at least in the case of pure metals [8]. However, a test of fitting our data separately in low- and high-temperature regimes results in the same exponent within experimental error.

4. Conclusion

We have determined the dependence on temperature of the electric-field gradient at the Zr-site in Zr_2Ni, from 10 to 1200 K. The measured perturbation functions represent a single well-defined EQI characterized by very sharp lines. This result indicates that few point defects are present near the probe sites. Thus the C16 structure appears not to favor the formation of point defects.

Note added in proof: It has been called to the authors' attention that this system was previously investigated in the temperature range above 300 K by Wodniecka *et al.* [9]. Within this temperature range, our results are in agreement.

Acknowledgements

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