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Pulsed-beam study of ^{111}Cd nuclear quadrupole interaction with recoil-induced lattice defects in cubic palladium

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The quadrupole interaction of excited ^{111}Cd nuclei with electric field gradients due to lattice defects in fcc palladium has been studied by the technique of time-differential observation of perturbed angular distribution of γ rays. The lattice defects were induced by the recoiling ions following the nuclear reaction ^{108}Pd(α, n)^{111}Cd. A sharp frequency of 87(2) MHz is assigned to trapped interstitials. A broad temperature-dependent frequency distribution arising from various other lattice defects was found. A migration energy of 0.10(2) eV for free interstitials is deduced.

In DPAD measurements (time-differential observation of perturbed angular distribution of γ rays following a nuclear reaction) the recoiling probe nuclei are usually assumed to come to rest at regular lattice sites. However, several investigations on magnetic and electric hyperfine interactions (HFI) exhibit perturbing interactions due to radiation damage created during the stopping process. These damage effects usually affect the magnitude of the modulation amplitude of the time spectra and its damping behavior,[1-4] and even the interaction frequency may be slightly changed.[4] At higher temperatures and especially above the melting point of the specimen under consideration the influence of the damage is weak or vanishes completely.[1,4] At lower temperatures, the radiation-induced lattice defects may be frozen in, and their interaction with the probe nuclei often yields the dominant contribution to the total HFI. The different types of defects will successively anneal when the temperature is raised, and the observation of the different recovery stages may yield information about the nature of the defects involved.

Conventional macroscopic methods for studying lattice defects, such as the observation of the change of the residual electric resistivity with temperature, give only global and time-averaged information. Earlier reported perturbed-angular-correlation (PAC) and DPAD measurements[5,6] are also of time-averaged character. In contrast, the DPAD method offers the opportunity of investigating the perturbation of the neighborhood of the probe nuclei during the microscopic time interval immediately following the nuclear reaction. Such a DPAD experiment will be described in this article.[7]

We have chosen the nuclear reaction ^{108}Pd(α,n)^{111}Cd. Nonmagnetic Cd impurities were recoil-implanted in the cubic (fcc) palladium metal, thus avoiding HFI's originating from a noncubic crystal structure or internal magnetic fields. Only the electric quadrupole interaction of the nuclear quadrupole moment with electric field gradients (EFG's) due to radiation-induced lattice defects should therefore be present.

The $\frac{5}{2}^+$ isomeric state of the probe nucleus ^{111}Cd (τ=123 nsec, E=247 keV) was populated using a pulsed α-particle beam from the 7-MV Van de Graaff accelerator at the Hahn-Meitner-Institut Berlin (E_α=13 MeV, pulse width 8 nsec, repetition time 1 μsec, ion current 20–50 nA). The target consisted of an 98% isotopically enriched palladium metal disk of about 50 mg/cm^2. The experiments were carried out at temperatures ranging from 20 up to 1200 K. For the measurements below room temperature the target was attached to the cold finger of an electronically regulated continuous-flow cryostat. At 20 K the increase in temperature due to the ion beam was measured to be 0.2 K/nA. For further details of this target arrangement see Ref. 8. In the experiments above room temperature, the target was heated electrically as described in Ref. 9. The intensity of the delayed γ rays, $I(\theta, t)$, was detected by two 1.5×1.5-in.^{2} NaI(Tl) scintillators placed at angles $\theta=0°$ and 90° with respect to the beam axis.

The measured time spectra were analyzed by inserting $I(\theta,t)$ in the ratio $R(t)=I(0°,t)/I(90°,t)$ and fitting this ratio with the expression $R(t)=1+\frac{3}{2}A_2G_2(t)$. The perturbation factor $G_2(t)$ (see, e.g., Frauenfelder and Steffen[10]) was taken as[11]

$$G_2(t)=\sum_{n=0}^{3} s_{2n}[f\cos(n\omega_0 t)+(1-f)e^{-n\delta t}]. \qquad (1)$$

It includes two contributions to the perturbation: (i) A fraction f of the probe nuclei interacts with a unique electric field gradient (EFG) giving rise to a sharp frequency ω_0; (ii) the residual $(1-f)$ fraction of the nuclei experiences EFG's broadly distributed around zero. A Lorentzian distribution was used rather than a Gaussian one since it gave better fits in all cases.

Some experimental time spectra together with the obtained fits are shown in Fig. 1. The measurements led to the following results:

(i) For the anisotropy coefficient we measured $A_2 = +0.15(3)$, independent of temperature in the entire range investigated. Apparently in this case there are no temperature-dependent effects which reduce the initial nuclear alignment, as observed in some other DPAD experiments (e.g., Ref. 2).

(ii) The strong damping of the perturbation amplitude $A_2G_2(t)$, which decreases with increasing temperature, indicates a temperature dependence of the distribution of the EFG's. The width of the corresponding interaction frequency distribution $\Delta(e^2Qq/h) = 20\delta/6\pi$ [see Eq. (1)] is plotted as a function of temperature in Fig. 2.

A sharp quadrupole interaction frequency ω_0 is observed, similar to that found in Ref. 6 for ^{111}Cd in Ag by DPAC. No significant dependence of ω_0 on temperature was measured. The averaged value was $e^2Qq/h = 20\omega_0/6\pi = 87(2)$ MHz. Below $T = 410$ K, the fraction f of the nuclei interacting with the unique EFG was found to be constant within the limits of error. Averaging over several measurements the fraction $f = 0.035(15)$ was obtained. At $T = 410$ K, however, the sharp frequency disappeared completely.

The results were shown to be reproducible on varying the experimental conditions and using targets which were prepared differently. The time spectra also appeared to be independent of the

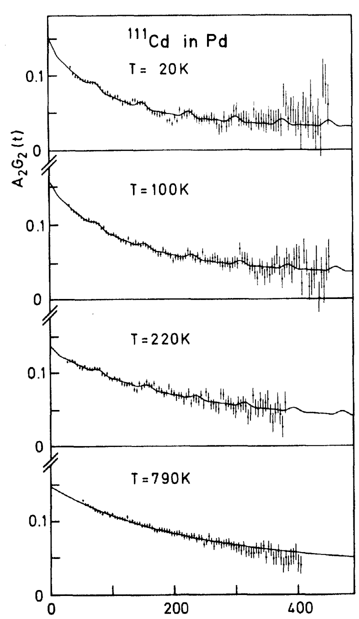

FIG. 1. DPAD spectra of ^{111}Cd in fcc palladium metal for different temperatures.

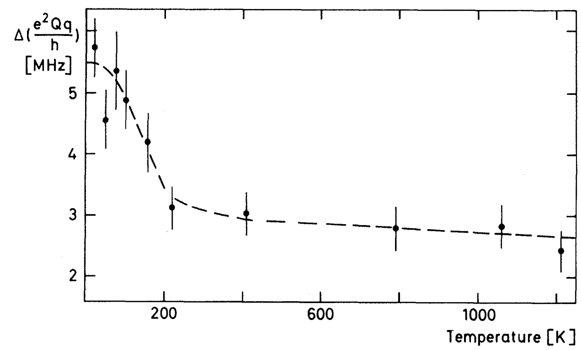

FIG. 2. Measured half-width of the frequency distribution as a function of temperature.

irradiation dose. Since the α particles come to rest far away from the stopped recoils as a consequence of their longer range, damage effects in the immediate surrounding of the probe nuclei due to accumulation of helium should be negligible.

Using the approximation of a static random distribution of point charges on lattice sites, we calculated the perturbation factors by a Monte Carlo technique, including effects of EFG asymmetry. Applying this model, we obtained a sharp frequency due to a point charge at a nearest-neighbor position plus a broad distribution around zero frequency, in agreement with the assumptions of our fitting function of Eq. (1). Typical results of the Monte Carlo calculation are shown in Fig. 3. They reveal a sensitive dependence of the damping on the concentration of point defects. The closest resemblance of the calculated results to the curves measured below $T = 410$ K was obtained for a concentration of 0.5(2)%.

A more realistic interpretation of the results may be attempted in the light of the present under-

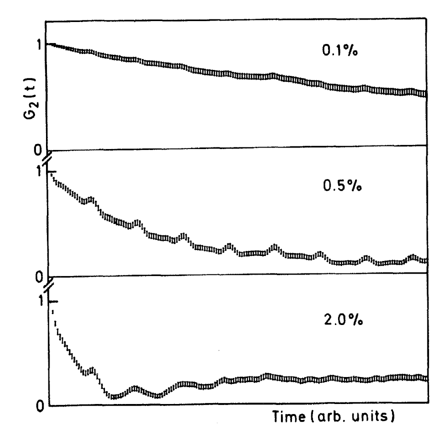

FIG. 3. Calculated G_2 functions from the statistical model for different concentrations of point charges.

standing of defects in fcc metals. At the lowest temperatures employed in this experiment interstitials and vacancies do not show thermally activated motion. They would be expected to give rise to static EFG's at the probe nuclei broadly distributed around zero. This corresponds to our $1-f=0.965$ distribution of random defects. A defect at the nearest-neighbor position of the probe nucleus may be expected to produce an EFG much larger than the others. Using the estimated quadrupole moment of $Q=0.5$ b,[12] the interaction frequency of 87(2) MHz yielded an EFG of 7×10^{17} V cm^{-2} for such a point defect. This value of the EFG is indeed of the order of magnitude expected.

The sharp drop in the width of the frequency distribution (Fig. 2) at about $T=150$ K indicates that above this temperature one contribution to the perturbation disappears within a time less than $\Delta t=10^{-8}$ sec. We may identify this drop with the annealing stage I, generally attributed to free interstitial migration. This stage was observed at $T'=35$ K in a macroscopic study of damage in Pd,[13] i.e., above this temperature the interstitials anneal in a time shorter than $\Delta t'=10^3$ sec, the typical annealing time in resistivity measurements. Combining these data with ours ($\Delta t=10^{-8}$ sec at $T=150$ K) we can estimate the activation energy for free interstitial migration as $E=k(\ln\Delta t-\ln\Delta t')/(T'^{-1}-T^{-1})\approx0.10(2)$ eV.

Since Cd strongly distorts the Pd lattice, as evidenced from the change in lattice constants,[14] some interstitials may be trapped by the Cd impurities and thus will anneal at higher temperatures. As the point defects giving rise to the unique quadrupole frequency remain above 150 K but disappear at about 400 K, it is tempting to identify them with interstitials trapped by Cd impurities. The axially symmetric EFG assumed in our fits[11] is in agreement with such a picture. Above 150 K we observed no enhancement of the fraction f attributed to trapped interstitials, i.e., thermally activated trapping found, for example, in a recent Mössbauer experiment[15] does not give a significant contribution. A binding energy of 0.2(1) eV may be extracted from our data.

In this description the frequency distribution observed at temperatures above 400 K is due to defects with lower mobility. These should then also be stable at room temperature and may lead to damping effects similar to those observed by Behar and Steffen.[6]

In a further DPAD experiment on the reaction ^{108}Pd$(\alpha,n)^{111}$Cd, performed with the purpose of determining the Knight shift and its temperature dependence of Cd in palladium metal, the magnetic spin rotation spectra revealed a temperature-dependent damping behavior. This could be nicely fitted by applying the results shown in Fig. 2.

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