

Defect-Impurity Interaction and Loss of Nuclear Alignment of ^{69}Ge and ^{67}Zn in Copper Metal*

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The alignment of impurity nuclei recoil-implanted into copper has been measured as a function of temperature. At 300 K, ^{69}Ge and ^{67}Ge impurities lose 50% of the nuclear alignment within 10^{-8} sec after implantation into the copper host, while the full alignment is observed for the ^{67}Zn impurity. The temperature dependence of the alignment can be explained by a simple model which assumes static quadrupole interactions with lattice defects in the vicinity of the impurity.

With conventional methods, the study of radiation-damage-impurity interactions is made difficult by the low impurity concentration required to insure that impurity clustering does not occur. The perturbed-angular-distribution (PAD) method has recently been successfully applied to the study of radiation damage.¹ We have used excited recoil nuclei both to create damage in the host near the stopping point of the recoil, and then sensitively to probe the damage with the excited nucleus on time scales of the order of 10^{-7} sec. The sensitivity of the method allows experiments with impurity concentrations of less than 10^{-8} so that isolated impurity nuclei can be studied. We have chosen natural copper metal as a host material for the initial investigation because of the convenience of several nuclear probes that can be produced in copper, and because much knowledge is available about defects in copper from macroscopic solid-state studies.²

A pulsed beam of 32-MeV ^7Li ions from the State University of New York at Stony Brook FN tandem accelerator was incident on a thick target of 99.999% pure, natural, polycrystalline copper metal. The pulse repetition interval was either 8 or 16 μsec . The different isomeric probe nuclei which were produced are listed in Table I, with their spins, γ -transition energies, lifetimes, and g factors. Although the absolute quadrupole moments of these isomers are not known, the quadrupole interaction frequency in Zn metal gives an indication of their relative strengths and is also listed in the table.

The anisotropy of the delayed γ radiation was measured with the time-differential PAD method. A transverse magnetic field of about 9.5 kG was applied to the target, and two magnetically shielded NaI(Tl) detectors at angles of $\pm 135^\circ$ to the beam direction were used to observe the spin precession pattern of the probe nuclei. The ratio

TABLE I. Properties of nuclear probes.

Probe	I^π	E_γ (keV)	$T_{1/2}$ (nsec)	g factor	Ref.	Reaction	A_2^a	e^2qQ/h (Zn, $T=630$ K) (MHz)
^{67}Ge	$9/2^+$	734	70(7)	$-0.210(7)$	b	$^{63}\text{Cu}(^7\text{Li}, 3n)^{67}\text{Ge}$	0.124(12)	86.8(8) ^e
^{67}Zn	$9/2^+$	605	333(14)	$-0.243(2)$	c	$^{65}\text{Cu}(^7\text{Li}, \alpha n)^{67}\text{Zn}$	0.124(7)	39.3(4) ^f
^{69}Ge	$9/2^+$	398	2840(70)	$-0.224(7)$	d	$^{65}\text{Cu}(^7\text{Li}, 3n)^{69}\text{Ge}$	0.188(10)	71.1(7) ^e

^a Measured at $E(^7\text{Li}) = 32$ MeV, using a liquid $\text{Cu}_{0.8}\text{Sb}_{0.2}$ eutectic target.

^b H. Bertschat, H. Haas, W. Leitz, U. Leithäuser, K. M. Maier, H. E. Mahnke, E. Recknagel, W. Semmler, R. Sielemann, B. Spellmeyer, and Th. Wichert, J. Phys. Soc. Jpn., Suppl. 34, 217 (1973).

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$R(t) = [N_+(t) - N_-(t)] / [N_+(t) + N_-(t)]$ was formed using the background-corrected counting rates N_{\pm} . A least-squares fit to the ratio was performed with the function

$$R(t) = [3A_2G_2(\Delta t) / (4 + A_2G_2)] \times \exp(-t/\tau_d) \cos 2(\omega_L t - \theta).$$

The nuclear lifetime does not enter into this ratio and is not an important parameter provided it is long enough to observe the Larmor precession. The γ -ray anisotropy coefficient A_2 depends on the nuclear alignment produced by the reaction, and is given in Table I for the different reactions with 32-MeV projectile energy. These values were measured in a separate experiment using a molten $\text{Cu}_{0.8}\text{Sb}_{0.2}$ eutectic mixture held at about 1000 K. In solid Cu the attenuation factors $G_2(\Delta t)$, which describe the fraction of probe nuclei taking part in the Larmor precession at times greater than $\Delta t \approx 10^{-8}$ sec after the excitation, were observed to be temperature and probe dependent. This dependence is shown in Fig. 1 for the probe nuclei ^{69}Ge and ^{67}Zn . In addition, at temperatures below 750 K, the ^{69}Ge data exhibited some damping of the spin precession oscillations which could be fitted with an exponential decay constant $\tau_d = 8 \pm 3$ μsec for all tempera-

tures below 750 K. The other shorter-lived probes, ^{67}Ge and ^{67}Zn , were not sensitive at these long times.

Even though the ^{67}Zn and ^{69}Ge probes were in the same target and the temperature dependences shown in Fig. 1 were measured simultaneously, a clear difference in their behavior is observed. For the ^{67}Zn nuclei, the nuclear alignment is unattenuated in the solid, while for the ^{69}Ge and ^{67}Ge nuclei at temperatures below 700 K, $(48 \pm 3)\%$ and $(40 \pm 7)\%$ of the nuclear alignment, respectively, is lost in a time less than $\Delta t \sim 10^{-8}$ sec. Two possible mechanisms have been considered to explain this loss of alignment. Either a fast relaxation of the alignment occurs because of strong fluctuating field gradients for a short time, or a weaker static interaction occurs for a longer time. In the Abragam-Pound³ model for rapid fluctuations, one expects the strength of the relaxation to be proportional to the square of the quadrupole moment and one would expect to see 1.5 times more dealignment for ^{67}Ge than for ^{69}Ge , while the observed loss is equal within the experimental error. For the static mechanism, the loss of alignment arises from the static quadrupole interaction of the nuclear probe with the electric field gradient from a nearby lattice defect, and therefore the fraction of atoms of a given type with a nearby defect governs the fractional loss of alignment. The different loss of alignment for Ge and Zn probes indicates a different fraction of nearby defects for these different impurities.

In order to understand better the temperature dependence of the Ge impurities in copper, shown in Fig. 1, a simple model was made. After the recoil atom has dissipated its energy through atomic-collision cascades, the impurity atom sits in a limited volume of high lattice disorder. Because of the high temperature in such a collision spike, annealing takes place typically⁴ between 10^{-12} and 10^{-10} sec. If the implanted impurity presents an attraction to the lattice defects, e.g., vacancies or divacancies, a certain fraction, f , of the excited nuclei may end up with a nearest-neighbor defect after the annealing of the thermal spike. At lower temperatures this fraction of the nuclear alignment is not allowed to take part in the Larmor precession, because of the strong electric quadrupole interaction with the defect. At the higher target temperatures used, the defects are mobile and can diffuse away before significant nuclear dealignment has occurred. Assuming an activation energy E^S for

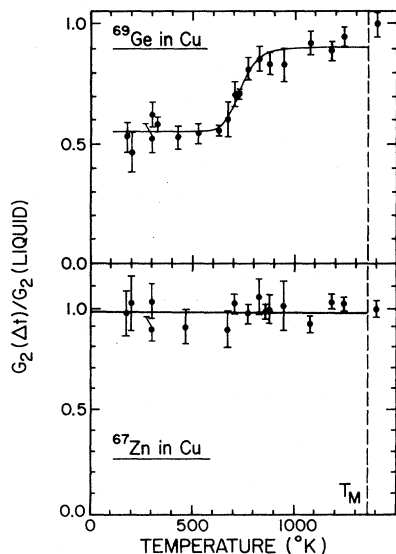


FIG. 1. Temperature dependence of the attenuation coefficient $G_2(\Delta t)$ for ^{69}Ge and ^{67}Zn isomers in copper metal. The melting temperature of copper, T_M , is indicated by the vertical line. The data shown above the melting point were taken with a liquid $\text{Cu}_{0.8}\text{Sb}_{0.2}$ eutectic mixture at 1000 K.

the defect to migrate away from the impurity, the mean jumping frequency is given by $\nu = \nu_0 \exp(-E^S/k_B T)$. The probability for a defect to jump at a time t after production is $P(t) = \nu \times \exp(-\nu t)$; taking this into account, a temperature-dependent attenuation factor G_k can be derived (the presence of the external magnetic field is neglected here):

$$G_k(t, \nu) = (1-f) + f \sum_n S_{kn} \left\{ \int_0^t P(t') \cos n \omega_0 t' dt' + [\exp(-\nu t)] \cos n \omega_0 t \right\},$$

where the S_{kn} are calculated coefficients⁵ depending on the nuclear spin I , and $\omega_0 = 3e^2 Qq/2I(2I-1)\hbar$ for half-integer I . If $\nu \Delta t \gg 1$, then most of the defects have jumped before the spin precession is observed and G_k converts to a simple form:

$$G_k(\Delta t, T) = (1-f) + f \sum_n S_{kn} \left\{ 1 + [(n\omega_0/\nu_0) \exp(E^S/k_B T)]^2 \right\}^{-1}.$$

A least-squares fit of this function is shown with the ^{69}Ge data in Fig. 1, and gives $f = 0.48 \pm 0.03$, $\nu_0/\omega_0 \approx 3 \times 10^5$, and $E^S = 0.64 \pm 0.14$ eV. The migration energy obtained here, 0.64 eV, equals the migration energy of the defect plus the defect-impurity binding energy. If we assume the binding is 0.1–0.2 eV, then the free-defect migration energy is ≈ 0.5 eV, somewhat smaller than the value of 0.8–1.0 eV usually quoted² for vacancies in Cu. This may indicate that the defect associated with the impurity is a divacancy which is known to have a smaller migration energy, and a high probability of formation in high-energy collisions. The fit is not sensitive to the quadrupole interaction frequency ω_0 or to the attempt frequency ν_0 . Since the attempt frequency² is between 10^{13} and 10^{14} Hz, the quadrupole perturbation frequency is most probably around $\omega_0 \approx 2 \times 10^8 \text{ sec}^{-1}$. The observation of a damping of the ^{69}Ge oscillations at all temperatures below 750 K is attributed to weak static quadrupole interactions with frozen defects that are more distant from the impurity probe. The damping is not observed at higher temperatures, consistent with the proposed defect migration model.

Although the quadrupole interaction of the ^{67}Zn impurity is somewhat weaker than the Ge probes, there is only a weak dependence of $G_k(t)$ on ω_0 in our model, and a dealignment should occur for the Zn probe if the damage profiles around the Zn and Ge impurities were identical. The momentum transferred in the probes in the nuclear reaction is similar, so one expects similar initial damage profiles, but the diffusion of the defects during the thermal spike may allow defect trapping at the impurity. The screening charge⁶ built up on the impurities can give rise to an impurity-vacancy attraction which would be larger for the Ge ($\Delta Z \approx 3$) than for the Zn ($\Delta Z \approx 1$). A similar

large loss in nuclear alignment for $\Delta Z \approx 3$ probes (presumably due to the mechanism described here) has been observed⁷ for ^{113}Sn in Ag and ^{200}Pb in Au.

The striking difference observed in the same target for two different probe nuclei clearly indicates a dependence of the loss of nuclear alignment on the chemical nature of the impurity. A quantitative explanation of the temperature dependence of the anisotropy indicates that a static quadrupole interaction with lattice defects is responsible for the loss of alignment.

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