TIME-DEPENDENT RELAXATION OF ALIGNED NUCLEI DUE TO RADIATION-INDUCED VACANCIES IN SOLID GERMANIUM

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We observe the temperature dependence of the alignment of an isomeric state in ^{73}As (426 keV, $\frac{9}{2}^+$, $\tau = 8\,\mu$sec), produced by the reaction ^{72}Ge$(d,n)^{73}$As in solid Ge. The relaxation process can be interpreted with a diffusion-controlled time-dependent quadrupole interaction due to radiation-induced lattice damage. A diffusion energy of $E = 1.3 \pm 0.2$ eV is assigned to the activation energy for excess vacancy diffusion. This result yields, for the first time, quantitative insight into relaxation mechanisms associated with radiation damage in solids, which may occur when implantation techniques and nuclear reactions are employed.

A recently published method, the stroboscopic observation of the nuclear Larmor precession in connection with pulsed beams, was announced to be suitable for precise hyperfine measurements on isomeric states with lifetimes in the 10^{-6}-10^{-2}-sec range.[1] Particular difficulty for such investigations arises from the requirement of preserving alignment during the lifetime τ of the nuclear state. Problems related to the stopping process of the recoil, its final position, and radiation damage of the lattice are largely unsolved.

In the present work the 8-μsec, 426-keV state of ^{73}As is produced in a solid Ge target by the reaction ^{72}Ge$(d,n)^{73}$As at deuteron energies $E_d = 5$ MeV. The rotation of the resultant anisotropy of the de-exciting γ-ray distribution in an applied magnetic field is observed as a function of temperature, where the stroboscopic method is used to detect the nuclear Larmor precession. The g factor obtained by this method is $g = 1.163 \pm 0.003$.[2]

The variations of the anisotropy are interpreted with the time-dependent electric quadrupole relaxation theory of Abragam and Pound, where the correlation time is assumed to be governed by vacancy-diffusion processes in the radiation-damaged Ge lattice.

The target was cubic (diamond structure) ^{72}Ge metal, molten onto a cylindrical tungsten backing, 6 mm in diameter and 10 mm long. It was indirectly heated and placed inside a chamber connected to the vacuum system of the accelerator. The target construction provided temperature stability of ± 1°C, indicated by a thermocouple which was introduced into the tungsten cylinder.

If relaxation occurs during the lifetime of the excited states that can be described in the sense of Abragam and Pound with a time-differential-attenuation factor $G_k(t) = e^{-\lambda_k t}$, where only $k = 2$ occurs in this experiment, the linewidth and the amplitude of the resonance curve are affected as follows[1]:

$$\Delta\omega_{1/2} = \Delta\omega_{1/2}{}^0 + \lambda_2 \quad \text{(linewidth)},$$

$$R = R^0 G(T) \quad \text{(amplitude)}, \tag{1}$$

$$G(T) = \left[1 + \frac{\tau\lambda_2}{1 + \frac{1}{4}A_2(1)A_2(2)}\right]^{-1}. \tag{2}$$

$A_2(1)A_2(2)$ are angular-correlation parameters, describing the nuclear reaction mechanism and the γ-ray transition.

Several resonance curves at different target temperatures (Fig. 1) illustrate the line broadening and variation of the amplitude. Below 600°C no measurable resonance occurred, indicating a relaxation time $T_{\text{rel}} \equiv 1/\lambda_2 < 500$ nsec. To determine the relaxation behavior over the entire temperature range accessible, the attenuation $G(T)$ of the resonance amplitude was measured as a function of temperature; this is shown in Fig. 2. In Fig. 1 the amplitude is given in the form of the double ratio $(Z_1/Z_2)(Y_2/Y_1)$, associated with a two-counter experiment (Z, Y) at different counting times (indices 1, 2), stated in detail in Ref. 1.

The experimental data in Fig. 2 can be explained with an exponential dependence of λ_2 on temperature. This dependence of the perturbation mechanism on temperature suggests a model of lattice defects, which diffuse near the final-site positions of the precessing nuclei. Several authors find evidence that recoils in solids are subjected to considerable quadrupole fields, where radiation damage might be responsible for lattice defects.[3] Diffusion then would introduce a time-dependent electric quadrupole inter-

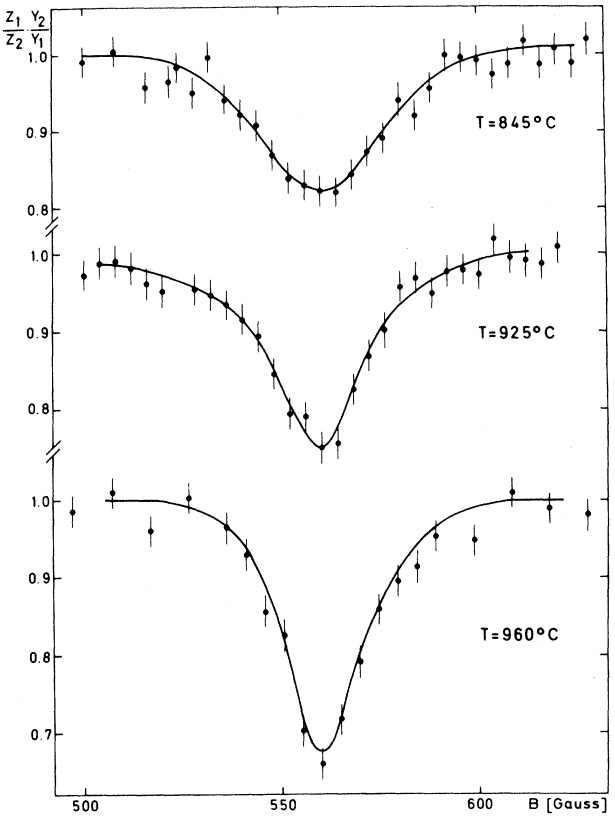

FIG. 1. Stroboscopic resonance curves of the γ-ray intensity at different target temperatures. At $T=845$°C and $T=925$°C the target is solid Ge; at $T=960$°C it is in the liquid phase.

action, and $\lambda(k=2)$ is expressed as[4]

$$\lambda=\frac{1}{T_{rel}}=\frac{9}{40}\frac{(eQ)^2}{h^2}\langle V_{zz}^2\rangle\frac{4I(I+1)-7}{I^2(2I-1)^2}\tau_c$$
$$=\langle\omega_Q^2\rangle\tau_c, \qquad (3)$$

where angle brackets denote averages. Every change in the lattice configuration surrounding the recoil, caused by a jump of an atom to a different lattice site, will change the perturbing configuration considerably; the inverse of this jump frequency may be interpreted as the correlation time τ_c.

Taking $\tau_c\sim e^{E/kT}$, and therefore $\lambda\sim e^{E/kT}$, a least-squares fit using Eq. (2) yields $E=1.3\pm0.2$ eV. The whole temperature dependence in this model is assigned to τ_c. The activation energy $E=1.3$ eV fits well with several experimental and calculated diffusion data, which associate this value with the activation energy for migration of a vacancy E_V^M.[5] The fact that this activation energy can be observed in this experiment is based on the following reasons: (1) The recoil affects excess vacancies near its final-site

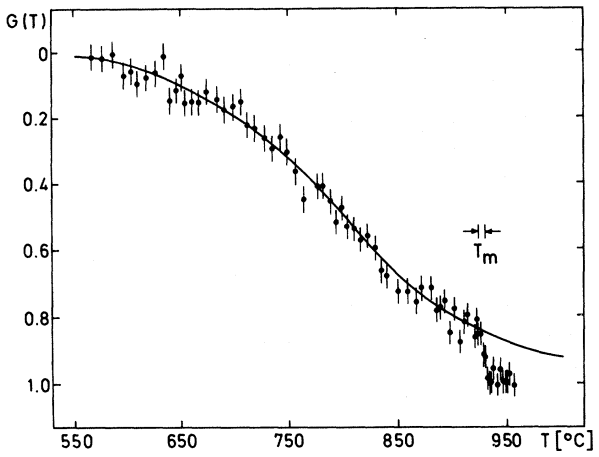

FIG. 2. Attenuation factor $G(T)$ of the stroboscopic amplitude as a function of temperature; the solid line is a least-squares fit using Eqs. (2), (3), and (5).

position; (2) the precessing nucleus is sensitive to the dynamic behavior of its enviornment on a microscopic time scale (about 10^{-5} sec).

Introducing the concept of enhanced diffusion, which in metals generally occurs via excess vacancies, the diffusion coefficient can be written as[6]

$$D=D_V(c_{th}+c_{irr})$$
$$\equiv D_V^0\exp(-E_V^M/kT)(c_{th}+c_{irr}), \qquad (4)$$

where D_V is the vacancy-diffusion coefficient, c_{th} the atomic fraction of vacancies in thermal equilibrium, and c_{irr} the radiation-induced atomic fraction of vacancies near the final-site position of the recoil nucleus. c_{th} is not larger than 10^{-6} even at high temperatures. It is given by $\exp(-E_V^F/kT)$, where E_V^F is the energy required to form a vacancy and $E_V^F+E_V^M\equiv E^{SD}=3$ eV is the self-diffusion energy in Ge.[5] c_{irr} may easily be larger by several orders of magnitude; then, if c_{irr} does not change considerably during the lifetime τ of the excited nucleus, i.e., the lifetime τ^V of the vacancy is larger than τ, the first term of Eq. (4) can be neglected.

The temperature dependence is given by E_V^M. A constant fraction of vacancies c_{irr} during the lifetime of the nucleus is assumed for the following reasons: The lifetime of an excess vacancy can approximately be calculated to be not shorter than about 10^{-3} sec (at 900°C), which is computed by using data of excess vacancy diffusion in proton-bombarded Ge.[7] On the other hand, overall radiation effects can be neglected because of the rapid annealing of the lattice defects.

The correlation time τ_c now is defined as the

inverse of the jump frequency of a vacancy in a region near the recoil times the number f of vacancies in this region, because each of them contributes independently to a change of the lattice configuration. This can be expressed in terms of the vacancy diffusion coefficient

$$\tau_c = a^2/D_V f, \tag{5}$$

where a is the distance between adjacent lattice sites, and f can be related to c_{irr} by $f = nc_{irr}$, where n is the number of atoms near the recoil position.

The quadrupole interaction [Eq. (3)] is a function of c_{irr}, $\langle\omega_Q^2\rangle = \langle\omega_Q^2\rangle(c_{irr})$; in general $\langle V_{zz}^2\rangle$ will increase with increasing radiation damage.

To calculate the order of magnitude for τ_c and the mean quadrupole coupling constant, we take $D_V = 2(-1.3/kT)$ [5] and $f \approx 1$. Then, $\tau_c \approx 10^{-10}$ sec (900°C), and inserting this value in Eq. (3), we obtain $(eQ/\hbar)[\langle V_{zz}^2\rangle(c_{irr})]^{1/2} \approx 10^8$ MHz, where $\langle\omega_Q^2\rangle\tau_c$ results from the least-squares fit using Eq. (2). The quadrupole interaction frequency reflects the radiation-induced charge asymmetry round the recoil nucleus; a Sternheimer antishielding factor may significantly contribute to the effective field gradient.

The requirements of Abragam and Pound's theory, $\tau_c \ll \tau$ and $\langle\omega_Q\rangle\tau_c \ll 1$, are satisfied over the entire temperature range observed. The lower temperature limit is given by the ratio τ/T_{rel} [Eq. (2)]; the remaining amplitude R is no longer observable for $\tau/T_{rel} \gtrsim 20$. At high temperatures, Fig. 2 clearly indicates the influence of melting on the relaxation time. The lattice collapses, and no measurable relaxation is present any longer; however, a lower limit can be given from the width of the stroboscopic resonance, $T_{rel} > 80$ μsec.

From the well-defined activation energy which is obtained in this experiment, it is concluded that radiation damage near the final-site position in this temperature region results in point defects rather than in extended amorphous zones. Radiation-created interstitials are highly mobile compared with vacancies ($E_I^M = 0.4$ eV)[5]; thus they do not contribute to the relaxation mechanism.

Concerning the value of 1.3 eV, some remarks seem to be of interest: Some evidence is given that this energy might be the energy for migration of a divacancy, bound together by an attractive potential of about 2 eV.[8] The formation time of this divacancy would be less than 1 μsec as follows from this measurement. This method seems to provide a possibility of getting some insight into diffusion mechanisms on a microscopic time scale.

Experiments on the 160-μsec, 473-keV state in ^{77}As are in progress: Values concerning the relaxation time in liquid Ge and information on the ratio of the quadrupole moments of ^{73}As/^{77}As seem attainable.

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