Influence of Cold Work and Radiation Damage on the Angular Correlation of Gamma Rays from In^{111} Nuclei Imbedded in Silver*

GEORGE W. HINMAN,† GILBERT R. HOY, JOSEPH K. LEES,‡ AND JOHN C. SERIO
Carnegie Institute of Technology, Pittsburgh, Pennsylvania

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Measurements have been made to determine the effect of cold work and radiation damage on the angular correlation of gamma rays in the decay of In^{111} nuclei in silver. The correlation for well-annealed silver samples is also discussed. It is concluded that there is only slight attenuation in well-annealed pure silver specimens. Furthermore, both radiation damage and cold work influence the correlation.

THE angular correlation of the two-cascade gamma rays occurring during the radioactive decay of In^{111} is expected to be unattenuated if the nuclei occupy substitutional sites in a pure silver lattice. The reasons for this are that the magnetic fields present are too small or rapidly changing to be important in a metal-like silver, and the electric quadrupole interaction is zero at a lattice site because of the cubic symmetry. Early experiments[1] on silver samples, however, showed considerable attenuation. Recent experiments indicate that the correlation is indeed unattenuated for specimens that are annealed at sufficiently high temperatures. The present report presents some data for pure silver samples that have been annealed for several hours at 890°C. There are, in addition, results of annealing curves for silver samples that are initially heavily cold worked and radiation damaged and also curves for the case where only the radiation damage is present.

The apparatus and sample preparation used in these experiments have been described elsewhere.[2,3] Summarizing briefly, the equipment measured delayed coincidences between the 173-keV and the 247-keV Cd^{111} gamma rays occurring in the decay of In^{111}. A coincidence is recorded if a 173-keV gamma actuates one counter during the period from 155 to 335 nsec preceding a 247-keV pulse in the second one. The angle between the counters is varied alternately between 90° and 180°.

The unattenuated angular correlation reported by Steffen[4] is

$$W(\theta)=\sum_{\text{even } k} A_kP_k(\cos\theta)=1-(0.180\pm0.002)P_2(\cos\theta).$$

For the case of a polycrystalline sample, if the angular correlation is attenuated, the attenuation can be described by $\hat{G}_2$ in

$$W(\theta)=1-(0.180\pm0.002)\hat{G}_2P_2(\cos\theta).$$

The unattenuated correlation therefore corresponds to $\hat{G}_2=1$. Steffen[4] adds a term $(0.002\pm0.003)P_4(\cos\theta)$ in the angular correlation, but it is not included here.

The In^{111} nuclei are introduced into the samples by alpha-particle bombardment through the reaction $Ag^{109}(\alpha,2n)In^{111}$. The energy of the alphas is approximately 40 MeV.

Over the course of several months a number of pure (99.99% and 99.999%) silver samples have been measured, and the results are given in Table I. The annealing history of these samples is given in column 2. All of these specimens were cold rolled from billets approximately 8 mm in diameter down to foils 0.014 in. thick. They were then bombarded by 40-MeV alpha particles to produce the In^{111}. The temperature of the samples was not controlled during these bombardments but was estimated to be less than 200°C during the period of about 10 min when the beam struck the target. Column 3 shows the directly measured values of the attenuation factor for the samples. This factor is labeled $b_2\hat{G}_2$ rather than $\hat{G}_2$ because it includes the effects of finite counter size. The values of $b_2\hat{G}_2$ are evaluated from experiment with the further assumption that $A_2=-0.180$. The values of b_2 have been calculated using the method of Rose[5] and are listed in column 4.

TABLE I. Angular correlation results in pure silver samples.

Specimen	Annealing history	$b_2\hat{G}_2$ experimental	b_2	$\hat{G}_2$ corrected
1	6 h 890°C	0.78 ±0.05	0.835±0.007	0.94 ±0.06
1	12 h 890°C	0.76 ±0.05	0.835±0.007	0.91 ±0.06
1	18 h 890°C	0.75 ±0.05	0.835±0.007	0.90 ±0.06
1	24 h 890°C	0.78 ±0.14	0.835±0.007	0.93 ±0.17
2	7 h 890°C	0.17 ±0.02	0.835±0.007	0.20 ±0.03
2	13 h 890°C	0.15 ±0.03	0.835±0.007	0.18 ±0.03
2	19 h 890°C	0.76 ±0.04	0.835±0.007	0.91 ±0.04
2	30 h 890°C	0.70 ±0.06	0.835±0.007	0.84 ±0.07
Ag 1*c*,1	24 h 890°C	0.819±0.014	0.835±0.007	0.981±0.019
Ag 1*c*,2	24 h 890°C	0.790±0.014	0.835±0.007	0.946±0.018
Ag 1*c*,3	24 h 890°C	0.814±0.016	0.835±0.007	0.975±0.021
Ag 1*c*,3	24 h 890°C	0.865±0.033	0.939±0.006	0.921±0.037
Ag 1*c*,2	24 h 890°C	0.874±0.034	0.939±0.006	0.931±0.038
Ag 1*d*,1	24 h 890°C	0.935±0.031	0.939±0.006	0.996±0.036

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† Present address: General Atomic Division, General Dynamics Corporation, San Diego, California.
‡ National Science Foundation Predoctoral Fellow.
[1] R. M. Steffen, Phil. Mag. Suppl. 4, 293 (1955).
[2] C. A. Giffels, G. W. Hinman, and S. H. Vosko, Phys. Rev. **121**, 1063 (1961).
[3] G. W. Hinman, G. R. Hoy, J. K. Lees, and J. C. Serio, preceding paper, Phys. Rev. **135**, A206 (1964).
[4] R. M. Steffen, Phys. Rev. **103**, 116 (1956).
[5] M. E. Rose, Phys. Rev. **91**, 610 (1953).

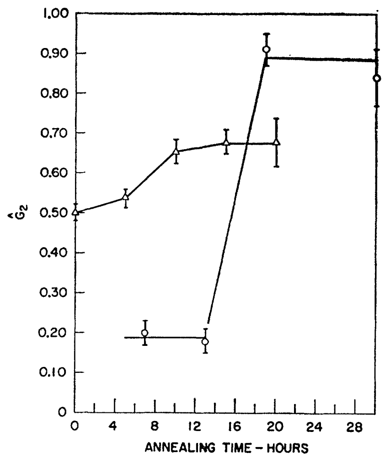

FIG. 1. $\hat{G}_2$ for sample 2(○) and for a specimen containing 0.25% Sb (Δ) as a function of annealing time at 890°C.

The correction due to finite source size has been estimated to be quite small.[6] Column 5 shows the $\hat{G}_2$ values after the counter correction has been made. It is clear from the results in Table I that the attenuation factor $\hat{G}_2$ is close to unity for well-annealed samples. There does appear to be a slight attenuation ($\hat{G}_2 \approx 0.96 \pm 0.015$) if the value $A_2 = 0.180$ given by Steffen is accepted.

On the other hand, samples often show hard-core attenuation before they are annealed as sample 2 in Table I illustrates. The $\hat{G}_2$ for sample 2 and for a specimen containing 0.25% Sb are shown in Fig. 1 as a function of annealing time at 890°C. The cold working continues to attenuate the correlation in some cases after as long as 12 h of annealing at 890°C but disappears after 24 h of annealing at this temperature.

In order to distinguish between cold working and radiation effects another series of pure silver samples was prepared. They were annealed for 24 h at 890°C before the alpha-particle bombardment in order to eliminate effects of cold work. The bombardments were carried out at liquid-nitrogen temperature but the angular-correlation measurements were made at room temperature. The values of $\hat{G}_2$ obtained in this series and the annealing treatment after bombardment are shown in Table II. Samples 1 and 2 in the table were bombarded at the same time. However, the measurements on sample 2 were made a week after those on sample 1. The difference in the two $\hat{G}_2$ values may therefore be a measure of the effect of a week's anneal at room temperature although the bombarding current of α particles was not sufficiently uniform to guarantee that the irradiation of the two foils was the same within a factor of 2. Recovery toward the unattenuated correlation is rapid at the 890°C annealing temperature and proceeds at a detectable rate even at 80°C.

It may at first seem surprising that the radiation damage is not sufficient to attenuate the correlation completely, especially in view of the trail of damage left by each In^{111} nucleus as it comes to rest. However, it is possible to explain qualitatively the observed attenuation and annealing effects using the generally accepted picture[7] of defects in a metal that has been subjected to alpha-particle bombardment. The explanation must be considered tentative because of the limited amount of data involved.

Immediately after an In^{111} nucleus comes to rest it may be in either an interstitial or a substitutional position. However, the activation energy for motion of interstitials is so low that during the 24 h at room temperature that elapse before measurements are made, the In^{111} nucleus will fall into a substitutional site. It is, of course, not necessary for the In^{111} to find a vacancy with which to combine in order for this to happen. It is sufficient for it to replace a silver ion at a substitutional site, with the silver becoming the interstitial and by a series of further interchanges moving away from the In^{111}. At the time of angular-correlation measurements, therefore, the In^{111} nuclei are presumably at substitutional sites and the effect of the alpha-particle irradiation is to leave an excess of vacancies in the sample. Most of the vacancies originally produced will have disappeared and no more than 2% to 10% will remain. Incidentally, the local vacancy cloud produced by each In^{111} as it slows to rest will have diffused to such an extent by the time measurements are made that the vacancy concentration in the wake of the In^{111} will not be appreciably higher than the general vacancy concentration produced by the alphas. The concentration of vacancies has been estimated to be 10^{-4} atomic fraction for the alpha-particle integrated flux of 0.3×10^{16} cm^{-2}. This assumes that only 5% of the vacancies originally produced are present at the time of the measurement and this concentration may easily be wrong by a factor of 4. This small concentration of vacancies is not sufficient to account for the observed attenuation if the In^{111} and the vacancies are randomly located. However, one can expect some interaction of these two and it is probably attractive in the first-

TABLE II. Radiation damage effects on angular correlation measurements.

Sample	Integrated flux of 40-MeV particles (cm^{-2})	Annealing temperature (°C)	Annealing time (h)	$\hat{G}_2$
1	0.3×10^{16}	890	0	0.41 ± 0.04
1	0.3×10^{16}	890	1	0.94 ± 0.02
1	0.3×10^{16}	890	2	1.00 ± 0.04
1	0.3×10^{16}	890	3	0.90 ± 0.03
1	0.3×10^{16}	890	5	0.91 ± 0.06
2	0.3×10^{16}	23	170	0.52 ± 0.03
3	0.1×10^{16}	80	0	0.51 ± 0.06
3	0.1×10^{16}	80	1	0.64 ± 0.06

[6] C. A. Giffels, Doctoral dissertation, Carnegie Institute of Technology, May 1960 (unpublished).

[7] F. Seitz and J. S. Koehler, in *Solid State Physics* edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1956), Vol. 2, p. 307.

neighbor position.[8] An attractive interaction will modify the distribution of vacancies and In^{111} ions and will reduce the average attenuation factor. From the experimental results it is possible to estimate the value of the attractive interaction energy in the first neighbor position. If one assumes G_2 is reduced to 0.20, the "hand core" value, for In^{111} ions with vacancies next to them, and if one assumes G_2 is unity for other In^{111} ions, the interaction energy must be about 0.20 ± 0.05 eV to explain the observed attenuation.

The annealing results at room temperature and at 80°C for this series furnish some additional information. By a fortunate coincidence the annealing at these two temperatures produces approximately the same effect. Thus, the ratio 0.52/0.41 is equal to 1.27 for the week's anneal at room temperature while the value for one hour at 80°C is $0.64/0.51=1.26$. Without going into the details of the annealing process one can evaluate the activation energy from this information using the fact that $D_1t_1=D_2t_2$, where D_1 is the diffusion coefficient at temperature T_1 and D_2 the value at T_2. It is assumed that $D=D_0e^{-Em/kT}$.

Substituting for t_1, T_1, and t_2, T_2 gives a value of $E_m=0.76$ eV. This value is very close to the activation energy for the motion of vacancies[9] and leads to a possible picture of the annealing process. The result is consistent with a gradual diffusion of the remaining vacancies in the samples to sinks, probably dislocations, with thermal equilibrium between vacancies and In^{111} ions maintained at all times.

[8] A. Blandin and J. L. DePlante, J. Phys. Radium **23**, 609 (1962).

[9] M. Doyama and J. S. Koehler, Phys. Rev. **119**, 939 (1960); **127**, 21 (1962).

There is one final check on the consistency of this picture that can be obtained from the annealing data. According to the process described above the probability that an In^{111} will have a vacancy in the nearest-neighbor position is given by $p=p_0\exp(-Ate^{-E_m/kT})$, where p_0 is the probability at time $t=0$, t is the annealing time and T is the annealing tempertaure, and $E_m=0.76$ eV is the activation energy for the diffusion process. The constant A is equal to v/a, where v is the frequency for the process $\cong10^{13}$ and a is the number of jumps a vacancy must make to reach a sink. For the present experiments the value of a, evaluated from the 80 and 23°C data, turns out to be about 5×10^6. This corresponds to a distance travelled by a vacancy to a sink of about 6×10^{-5} cm. Such a distance appears to be consistent with a distance of 10^{-4} to 10^{-5} cm between sinks. If the sinks are dislocations, the corresponding density is 10^8–10^{10} cm^{-2}, a range appropriate for a well-annealed polycrystalline sample of silver.[10]

Thus, the results of this last series of measurements appear to be consistent with the idea that the attenuation is caused by residual vacancies. However, the data are not extensive enough to regard this explanation as more than very tentative.

The principal conclusions of this work are the following:

(1) In well-annealed specimens of pure silver there is little attenuation of the angular correlation based on the A_2 value of Steffen[4] for the Cd^{111} gamma rays.

(2) Both cold working and radiation produce attenuation of the angular correlation. Of the two, severe cold work seems to produce more pronounced effects.

[10] C. Kittel, *Introduction to Solid State Physics* (John Wiley & Sons, Inc., New York, 1956), p. 554.