# Measurement of the Intrinsic Thermal Expansion of Irradiation Defects in Aluminum at Low Temperature

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A first measurement of the intrinsic thermal expansion coefficient of Frenkel defects produced by a low-temperature neutron irradiation in aluminum leads us to a value 12 times larger than the coefficient of thermal expansion for the pure metal.

In order to account for their self-diffusion experiments under pressure at various temperatures, Gilder and co-workers[1-3] suppose the activated vacancy at high temperature to have an intrinsic thermal expansion coefficient, $\beta_A = \partial v_A / v_A \partial T$ ($v_A$ is the activation volume for self-diffusion), much higher than that of the host crystal ($\beta_0$). The ratio $\beta_A/\beta_0$ is about 15 in zinc and cadmium. The strong curvature in the Arrhenius plot is then explained quite naturally. The temperature dependence of vacancy-formation parameters such as energy, volume, etc., has become a matter of renewed theoretical interest.[4-6] Audit and Gilder's calculation[7] predicts a ratio $\beta_d/\beta_0$ ($\beta_d = \partial v_f / v_f \partial T$) varying from 2.5 at 60 K to 4 at 890 K in aluminum and from 5 at 93 K to 8 at 296 K in sodium.

However, no direct measurement of the thermal expansion of intrinsic point defects has ever been reported. Only the thermal expansion of solute atoms has been measured in some rare cases.[8] We present a first measurement of the intrinsic thermal expansion of point defects (vacancies and interstitials) produced by neutron irradiation of aluminum at liquid-hydrogen temperature.

We use a differential dilatometer described elsewhere by Asty.[8] As a function of temperature, we measure the variation $\Delta\delta L$ of the difference in length $\delta L$ of a test sample made of commercial aluminum (purity 99.99%).

The two samples are coaxial tubes of equal length (50 mm) with three small "feet," defining reference planes, at each end. The test sample (inner tube) and the reference sample (outer tube) each carry an optical flat with semireflecting parallel planes. The image of the interference fringes produced by the incident beam from a He-Ne laser on the upper plane is recorded by means of an optical assembly composed of a telescope, a microscope, and a magnetoscope. The length resolution is 0.01 $\mu$m. During every measurement the temperature of both samples is stabilized to within 0.01 K. For all experiments the following temperature program was run: 2-h holding time for measurement alternating with 2-h temperature increases or decreases at a rate of 2 K/h.

After machining, chemical polishing, and annealing in a high vacuum furnace (4 h at 400°C under $10^{-6}$ Torr) the sample was irradiated in liquid hydrogen with fast neutrons (fluence $10^{18} n/\mathrm{cm}^2$, $E > 1$ MeV) in the low-temperature irradiation facility Vinka of the reactor Triton at Fontenay-aux-Roses described elsewhere.[9] After transfer in a cryostat, the sample was stored in liquid nitrogen, where it disactivated for two weeks. It was then mounted into the dilatometer in cold condition: Its temperature, monitored simultaneously, reached 160 K. After a slow cooling down to 77 K, we observed a positive slope $(\Delta\delta L/\Delta T)_a$ of the $\delta L$ vs $L$ plot between 80 and 160 K [Fig. 1(a)], which was confirmed by a complete cycle (decrease in temperature down to 80 K and second increase). When temperature increased beyond 160 to 260 K the annealing of the radiation-induced defects in the test sample resulted in its contraction with respect to the reference sample. Between 260 and 280 K, $\delta L$ approached a constant value (Fig. 2). In order to restore the initial state, we unmounted the sample for a second anneal in the ultravacuum furnace (4 h at 400°C, $10^{-6}$ Torr). The same test sample was then remounted to run the reference experiment, which yielded a positive but much weaker slope $(\Delta\delta L/\Delta T)_b$ [Fig. 1(b)] and showed no contraction phenomenon.

Unmounting and remounting the test sample produces two independent values with a data spread altogether comparable to those obtained for two successive temperature cycles without intermediate handling. Therefore we assume the slope $(\Delta\delta L/\Delta T)_b$ in the reference experiment to represent the null effect due to imperfections of the apparatus.

After irradiation and mounting into the dilatometer at 160 K the sample contains an overall concentration $c$ of radiation defects in the meta-

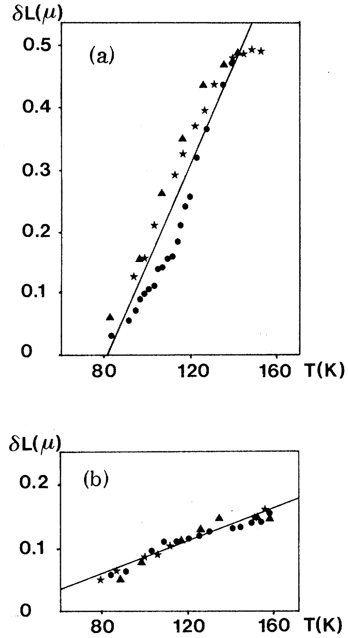


FIG. 1. Length difference $\delta L$ between test and reference sample vs temperature. (a) After irradiation at 20 K and mounting at 160 K. (b) Reference experiment after annealing at 400°C. Triangles, first run (increase in temperature); circles, second run (temperature decrease); stars, third run (temperature increase).

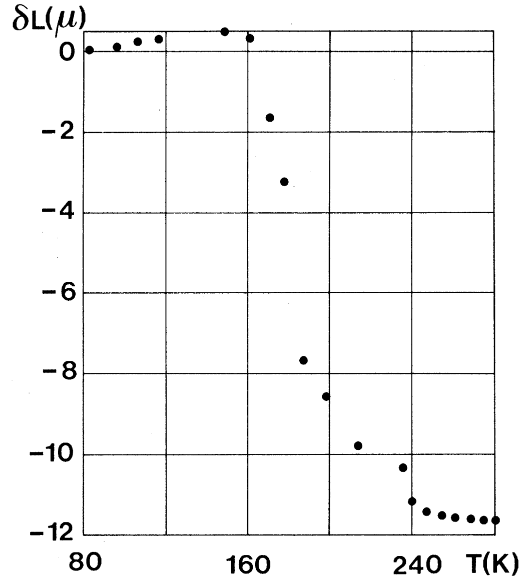


FIG. 2. Length difference $\delta L$ between test and reference sample vs temperature up to 280 K, showing the annealing through state III.

stable state at the end of the so-called stage II of annealing, i.e., isolated vacancies plus small interstitial clusters.[10] The effect of the defects on the thermal expansion of the sample is small compared to the total expansion due to pure aluminum, about 0.8%. The measurement technique enhances this small difference to produce Fig. 1. From 160 to 260 K the annealing through stage III resulted in a total relative contraction of the test sample with respect to the reference sample $\Delta\delta L_A = -12$ $\mu$m (Fig. 2). We verified that another sample, which had been irradiated to the same dose and then kept at room temperature, underwent no length change after each of the two successive treatments (4 h at 460°C, 16 h at 460°C), by comparison with the reference sample using a micrometer (reproducibility 1 $\mu$m).

Since both the length change $\Delta\delta L_A$ due to annealing through stage III, and the difference between the slopes in the two experiments $(\Delta\delta L/\Delta T)_a - (\Delta\delta L/\Delta T)_b$ are proportional to the mean formation volume of the defects $\langle v_f\rangle$, and the temperature derivative of this volume $(\partial v_f/\partial T)_p$, respectively, we can readily evaluate the intrinsic coefficient $\beta_d = \partial_f/v_f\,\partial T$. Indeed we can write $L = L_0(1 + cv_f/3\Omega_0)$, where $L_0$ is the length of the test sample without defects (reference experiment), $L$ the length of the sample containing an overall concentration $c$ of radiation defects, and $\Omega_0$ the mean atomic volume.

The derivative is

$$\frac{d\ln L}{dT} = \frac{d\ln L_0}{dT} + \frac{cv_f}{3\Omega_0}\left(1 + \frac{cv_f}{3\Omega_0}\right)^{-1}\left(\frac{d\ln v_f}{dT} - \frac{d\ln\Omega_0}{dT}\right).$$

Since

$$\frac{d\ln v_f}{dT} = \beta_d, \quad \frac{d\ln L_0}{dT} = \frac{\beta_0}{3}, \quad \frac{d\ln\Omega_0}{dT} = \beta_0$$

we get

$$\frac{dL}{dT} = \frac{L_0\beta_0}{3} + \frac{L_0 cv_f}{3\Omega_0}\frac{\beta_0}{3} + \frac{cv_f}{3\Omega_0}L_0(\beta_d - \beta_0)$$

$$\frac{dL}{dT} - \frac{dL_0}{dT} = \frac{cv_f}{3\Omega_0}L_0(\beta_d - \tfrac{2}{3}\beta_0).$$

But $cv_f L_0/3\Omega_0$ is just the length change due to the defects $|\Delta\delta L_A|$, and $\langle dL/dT - dL_0/dT\rangle$ is the difference between the slopes in our experiments, $(\Delta\delta L/\Delta T)_a - (\Delta\delta L/\Delta T)_b$, both explicitly measured. Thus,

$$\langle\beta_d - \tfrac{2}{3}\beta_0\rangle = \frac{1}{|\Delta\delta L_A|}\left[\left(\frac{\Delta\delta L}{\Delta T}\right)_a - \left(\frac{\Delta\delta L}{\Delta T}\right)_b\right].$$

With $\langle\beta_0\rangle = 0.5\times10^{-4}$ taken from the literature[11] and with $|\Delta\delta L_A| = 12 \pm 1$ $\mu$m,

$$(\Delta\delta L/\Delta T)_a = (0.79 \pm 0.11) \times 10^{-2} \ \mu\text{m/K},$$

$$(\Delta\delta L/\Delta T)_b = (0.13 \pm 0.02) \times 10^{-2} \ \mu\text{m/K},$$

resulting from the irradiation and the reference experiments, we find $\langle \beta_d/\beta_0 \rangle = 12 \pm 4$.

A second experiment has been done recently with another test sample of zone defined aluminum. In this case two reference experiments were run, one before irradiation, one after irradiation, both yielding the same result. It led to $\beta_d/\beta_0 = 14$, a value consistent with the previous one.

In conclusion we have shown that in aluminum defects such as isolated vancancies and small interstitial clusters have an intrinsic thermal expansion larger by about one order of magnitude than that of the matrix ($\beta_d/\beta_0 = 12 \pm 4$).

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# Impedance-Matched Grazing-Incidence Films: Pure Nuclear Reflections, Resonant Filtering of Synchrotron Radiation, and X-Ray Interferometry

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The purpose of the Letter is to point out the possibility of a new interference technique in x-ray and $\gamma$-ray optics—the simple idea of impedance-matched grazing-incidence films—and to discuss applications to pure nuclear coherent reflections, resonant filtering of synchrotron radiation, and x-ray interferometry.

In x-ray optics, the small index-of-refraction change ($N_0 = 1 \rightarrow N_1 = 1 - 2\pi\lambda^2 nzr_0 \approx 1 - 10^{-5}$, where $n$ is the atomic density, $z$ is the atomic number, and $r_0 = e^2/mc^2$) on entering a medium of *higher* electron density produces near total reflection of x rays near grazing incidence on a flat surface, with the critical angle being $\varphi_c = (n\lambda^2 z r_0/\pi)^{1/2}$. For $\varphi > \varphi_c$, there is a rapid dropoff in reflected intensity with increasing $\varphi$, and a rapid increase in the penetration depth $l_\perp(\varphi)$. For $Fe^{56}$, and 14.4-keV radiation, $\varphi_c = 3.8$ mrad, and for $\varphi = 3.0$ mrad, then $|R|^2 \simeq 0.9$ and $l_\perp \simeq 29$ Å, while at $\varphi = 4.5$ mrad, then $|R|^2 \simeq 0.1$ and $l_\perp \simeq 472$ Å.

For $\varphi > \varphi_c$, where deep penetration occurs, it should be possible to strongly suppress the x-ray reflection with an impedance-matched quarter-wave film, just as in optical coating of lenses. That is, the mirror is coated with a film of proper impedance such that the reflections at the upper and lower interfaces are equal and with the thickness chosen so the two waves emerge 180° out of phase.

The quarter-wave-plate condition gives $l_F(\varphi) = \frac{1}{4}\lambda(\varphi^2 - \varphi_c^2)^{-1/2}$, which is typically $\approx 50$–$100$ Å. A quarter-wave plate for 1-Å radiation is only possible because grazing incidence augments the