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Abstract - Ultrasonic velocity measurements were made after thermal neutron irradiation of single-crystal Cu specimens containing 700 or 3700 appm Be. Ultrasonic attenuation was measured in similar specimens after 3 MeV electron irradiation. Three anelastic relaxation processes due to self-interstitial-Be complexes were observed. Cu-Be 1 was found to occur near liquid helium temperature; the temperature dependence of the velocity change suggests that reorientation of the Cu-Be 1 complex may involve quantum mechanical tunneling. Cu-Be 2 produced an attenuation peak at ~2.5 K at a frequency of 10 MHz. Cu-Be 3 appeared simultaneously with 2 as a shoulder on the high temperature side of the 2.5 K attenuation peak.

I - INTRODUCTION

Many examples of very stable complexes formed when an irradiation-induced self-interstitial-atom (SIA) becomes trapped near a solute atom (SA) can be found in the literature. Anelastic relaxation has been used to investigate the symmetry and migration behavior of several of these SIA-SA complexes /1,2/. Since they can "fingerprint" individual defect types /3/, anelastic relaxation studies are highly complementary to investigations, e.g. those employing electrical resistivity, x-ray scattering, or ion channeling, which average over all defects.

II - EXPERIMENTAL

Two different single crystals of Cu-Be were used. One was purchased from the Monocrystals Company in Cleveland, Ohio and contained 700 appm Be. A second crystal was kindly furnished by L. M. Howe and M. L. Swanson of Chalk River Nuclear Laboratories; it contained 3700 appm Be.

Thermal neutron irradiation, which produces primarily isolated Frenkel pairs because of the low recoil energies involved, was performed in the low temperature

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irradiation cryostat at the CP-5 reactor formerly located at Argonne National Laboratory. Since SIA's become mobile in pure copper at temperatures near 40 K, specimen temperatures were maintained between 60 and 65 K during irradiation to enhance the probability of producing single SIA-Be complexes. Following thermal neutron irradiation, a visual pulse-echo overlap technique at a carrier frequency of approximately 10 MHz was used to measure the relative changes in the shear elastic constants ($\Delta C/C$) as a function of temperature with an accuracy of $\pm 5 \times 10^{-5}$.

Electron irradiations were performed at 1 MeV using the Van deGraaf accelerator in the Materials Research Laboratory of the University of Illinois at Urbana. Again, the temperature was maintained between 60 and 65 K during irradiation. No resistivity specimens were included in the electron irradiation runs. From previous studies in other alloys, we estimate the defect production to be 1 appm Frenkel pairs per 2×10^{17} e^{-}/cm^{2} /4/.

III - RESULTS AND DISCUSSION

The crystal containing 3700 appm Be was irradiated with thermal neutrons to a total measured resistivity change of 11 nΩ-cm, which corresponds to a Frenkel pair concentration of 40 appm. Measurements of the C_{44} mode revealed that a SIA-Be complex is produced during the 60 K irradiation which undergoes stress-induced reorientation and which disappears at annealing temperatures between 100 and 150 K. The effect on C_{44}, which we label as Cu-Be 1, is quite large. A total Frenkel pair concentration of only 40 appm produces a 2×10^{-3} change in C_{44} at 5 K.

The net temperature dependence, i.e., the difference between the pre and post irradiation results, introduced into C_{44} by Cu-Be 1 is shown in an Arrhenius plot in Fig. 1. Classically /3/, a thermally activated reorientation process produces no effect on the elastic constant at low temperatures where defect reorientation occurs

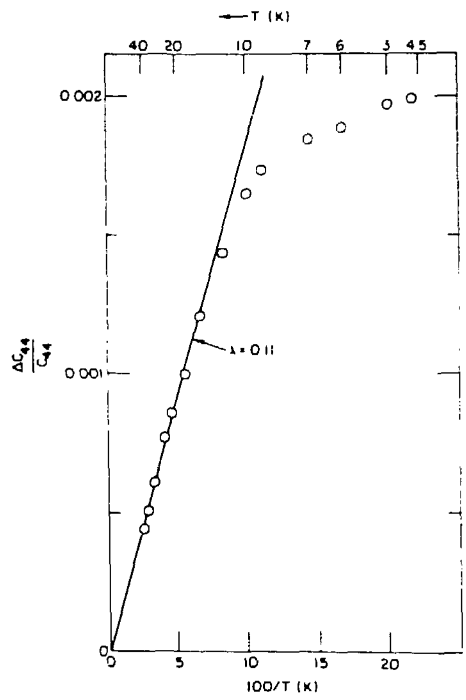

Fig. 1 - The temperature dependence of Cu-Be 1 plotted in an Arrhenius fashion.

much more slowly than the measuring frequency, exhibits a sharp dispersion at the temperature for which the reorientation and measuring frequencies are equal, and follows a T^{-1} dependence at temperatures for which the defect reorients faster than the measuring frequency. From Fig. 1, we see that at the lowest investigated temperature, 4.6 K, the change in C_{44} is still increasing with decreasing temperature. Hence, the defect reorients at a rate greater than $2\pi \times 10^7$ s^{-1} even at 4.6 K. Ultrasonic techniques, such as employed in the present study, are advantageous for studying defects which reorient so rapidly without annealing. Torsion pendulum (~1 Hz) or resonant bar (kHz) methods would require temperatures < 1 K in order to observe equivalent effects.

The crystal containing 3700 appm Be was irradiated with 3 MeV electrons to a dose of 2.4×10^{18} e^-/cm^2. Velocity measurements of the C_{44} mode after irradiation confirmed the existence of Cu-Be 1. After a 145 K anneal, a clear peak in the attenuation was observed near 2.5 K (Fig. 2). This attenuation peak, which we label as Cu-Be 2, was accompanied by a shoulder on the high temperature side, which we tentatively label as Cu-Be 3. As can be seen from Fig. 2, the shoulder disappeared after a 160 K anneal. Cu-Be 2 remained unchanged after the 160 K anneal, but disappeared completely after a 200 K anneal. No additional attenuation peaks or significant velocity changes were found in either the C_{44} on C' modes.

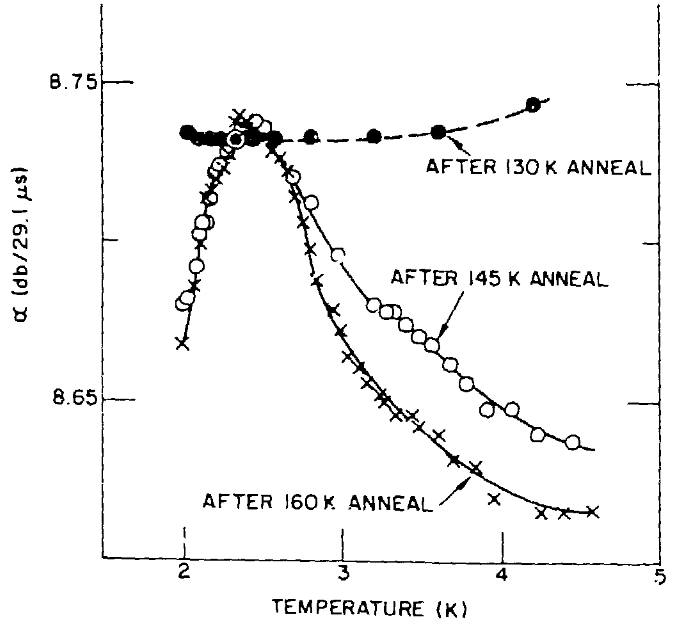

Fig. 2 - Attenuation in the C_{44} mode as a function of temperature measured in a Cu-3700 appm Be crystal irradiated with electrons then annealed at temperatures of 130 K (filled circles), 145 K (open circles), and 160 K (crosses).

The second-rank tensor λ_{ij} used to describe an elastic dipole is the strain component ε_{ij} per unit mole fraction c of defects /4/:

$$\lambda_{ij} \equiv \frac{\partial \varepsilon_{ij}}{\partial c}. \qquad (1)$$

For axially symmetric defects there are only two independent elements of this tensor, λ_1 and λ_2, and only the absolute value of the difference between these two, the so-called "shape factor" $|\lambda_1 - \lambda_2| \equiv \lambda$, can be obtained from measurements of the elastic moduli. A trigonal defect (<111> axis) possesses the highest symmetry consistent with the observation of a large anelastic effect in C_{44} and no effect in C'. For <111> symmetry, Eq. 1 becomes

$$- \Delta C_{44}/3C_{44}^2 = \frac{4}{27} (c_o V_0/kT) \lambda^2, \qquad (2)$$

where c_o represents the molar concentration of defects and V_0 is the molecular volume /4/. Assuming that the total measured defect concentration of 40 appm Frenkel pairs contributes, a fit of Eq. 2 to the T^{-1} dependence observed for T > 10 K in Fig. 1 yields $\lambda = 0.11$ for Cu-Be 1.

Below 10 K, a nonclassical temperature dependence is observed for Cu-Be 1. At least two possible explanations exist for such behavior. First, at these low temperatures defect reorientation may occur via quantum mechanical tunneling rather than by thermal activation. Experimental evidence for tunneling of SIA-SA complexes in irradiated Al-Zn has been obtained previously by Granato and coworkers /5/. Alternatively, the assumption that a large fraction of the required lattice vibrational modes are excited, which is necessary to obtain Eq. 2, may be invalid at these very low temperatures. This assumption has been discussed in detail by Flynn /6/.

The 2.5 K attenuation peak anneals completely at a temperature of 200 K, just slightly below the onset of vacancy migration in pure copper under similar irradiation and annealing conditions. Hence, both the binding and migration enthalpies of the SIA-Be complex responsible for process 2 are $\gtrsim$ 0.6 eV. Barteis et al. /7/ have used electrical resistivity to investigate radiation-induced segregation in dilute Cu-Be alloys during electron-irradiation slightly above room temperature. They conclude that a SIA-Be complex with a migration enthalpy of ~0.6 eV produces the very efficient removal of Be from solution that they observe. The strong binding and similar annealing behavior of process 2 observed here suggests that the Cu-Be 2 defect is responsible for the observed segregation effects.

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