Tunneling relaxation of a mixed dumbbell in an Al-Zn solid solution

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A low-frequency ($\sim$ 1-kHz) acoustic method has been used to measure the kinetics of the reorientation of mixed dumbbells in Al-Zn solid solutions. The reorientation is accompanied by diffusive jumps of Zn atoms between equilibrium positions within the unit cell. At $T < 20$ K, the relaxation rate τ^{-1} increases with decreasing temperature, as would be expected in the case of coherent tunneling diffusion.

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Impurity atoms in weak substitutional solutions such as Al-Fe and Al-Zn combine with interstitial atoms to form pairs which are stable at $T < 200$–250 K (Ref. 1). Geometrically, the pairs are dumbbells oriented along $\langle 100 \rangle$ directions (Fig. 1). Since the diffusive mobility of impurity atoms is vanishingly low at low temperatures, these mixed dumbbells are localized and are constrained to undergoing reorientations among the six directions along $\langle 100 \rangle$ axes. The impurity atom (the black circle in Fig. 1) undergoes transitions between six equilibrium positions within the unit cell, while the lattice atoms (the white circles) are simply displaced from their lattice sites. The energy barrier between equilibrium positions of the dumbbell is ≈ 15 meV high (Refs. 2 and 3).

In the present experiments we used a low-frequency ($\sim$ 1-kHz) acoustic method to study the kinetics of the reorientation of dumbbells in the alloy Al + 150 ppm Zn. This method was necessary because the symmetry of the deformation of the lattice around the dumbbells is lower than cubic, so that the dumbbells interact with the elastic fields. In an alternating field, the reorientation of the "elastic dipoles," accompanied by diffusive transitions between wells, causes a dissipation of elastic vibration energy. The frequency dependence of the elastic susceptibility of a crystal containing elastic dipoles is described by the Debye formula $\chi(\omega) = \chi(0)/(1 - i\omega\tau)$, where ω is the frequency, τ is the relaxation time, and $\chi(0)$ is the susceptibility corresponding to $\omega = 0$. By measuring the imaginary (χ'') and real (χ') parts of the elastic susceptibility (in other words, the absorption and the dispersion), we can determine the temperature dependence $\tau(T)$.

The test samples were small plates with dimensions of $0.3 \times 1 \times 10$ mm, pressed on one side into a massive block. The natural bending vibrations of the sample (the first quarter-wave mode) were excited by a regenerative system.[4] We measured the absorption of the acoustic vibrations and the elastic modulus (expressed below in units of the reciprocal quality factor, Q^{-1}, and the square of the resonant frequency, ν^2, respectively, of the acoustic vibrator).

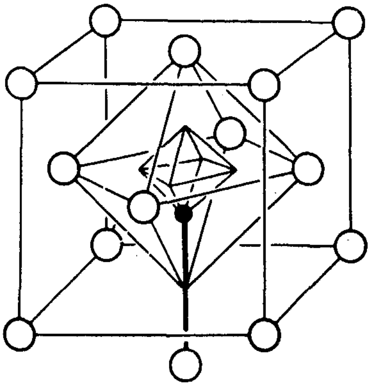

FIG. 1.

After bombardment by 4-MeV electrons in a dose of 6×10^{18} e/cm^2 at $T\approx 77$ K, the samples were placed in a helium cryostat (they were not allowed to rise above ≈ 100 K). The procedure used to bombard and transfer the samples was designed to preserve the mixed dumbbells formed as a result of the capture of impurity zinc atoms by interstitial atoms, which are mobile at $T\gtrsim 40$ K. In addition to bombarding the Al-Zn alloy, we bombarded pure Al (99.9996%) under identical conditions as a control.

In samples which were not bombarded and also in the bombarded pure aluminum we observed a monotonic decrease in the elastic modulus and an increase in the sound absorption with increasing temperature. The increased absorption can be attributed to the anharmonicity of the lattice and to the motion of dislocations. The temperature dependence of ν^2 and Q^{-1} of the electron-bombarded alloy is shown in Fig. 2. The elastic modulus decreases at $T<20$ K, and the absorption has a maximum at $T\approx 11$ K. These anomalous features can be erased by raising the samples to $T\approx$ 200 K for a few minutes (this procedure anneals out the dumbbells). These results suggest that the absorption maximum at $T\approx 11$ K and the anomalous decrease in the elastic modulus at $T<20$ K are due to the mixed dumbbells.

The only type of motion which could result from the introduction of the dumbbells at low temperatures and which could be responsible for the sound absorption and the dispersion of the elastic modulus is the reorientation of these dumbbells among the six equilibrium positions (Fig. 1). Precisely this motion has been observed at 10–30 K in the electron-bombarded Al-Fe alloy by acoustic absorption[2] and Mössbauer spectroscopy.[3]

The Debye dispersion of the elastic modulus for a sample containing elastic dipoles can be described by

$$(G_0 - G(T))/G_0 = A/(1+\omega^2\tau^2(T)). \qquad (1)$$

Here G_0 is the elastic modulus of the crystal in the absence of relaxation, $G(T)$ is the measured "dynamic" elastic modulus, and the coefficient A is proportional to the concentration of elastic dipoles. In all cases of the relaxation of elastic dipoles which

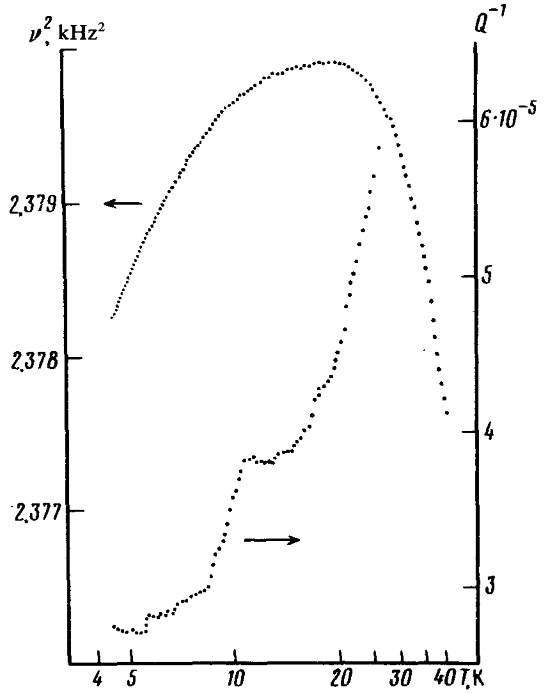

FIG. 2.

have been studied to date, the modulus $G(T)$ decreases with increasing temperature in a certain dispersive region near the $\omega\tau = 1$ relaxation resonance in accordance with (1), since the relaxation time τ always decreases in accordance with the classical Arrhenius law $\tau(T) = \tau_0 \exp(U/T)$, where U is the activation energy, representing the height of the barrier between the potential wells. We see that the observed inverse dispersion of the elastic modulus [the increase in $G(T)$ with increasing temperature] could be caused by only a decrease in τ with decreasing temperature. A $\tau(T)$ dependence of this type is characteristic of coherent tunneling diffusion.[5–7]

Quantum diffusion is usually associated with low temperatures, light particles, and quantum crystals. In the present case, the situation favors tunneling of a heavy atom because of such particular characteristics of the mixed dumbbell as the small effective thickness of the barrier wells (< 1 Å) and the existence of a soft librational mode,[8] $\omega_0 \approx \omega_D/10$.

Comparison of the tunneling kinetics of the mixed dumbbells with the well-studied case of the quantum diffusion of He^3 and He^4 crystals[9] shows that the relaxation rate of the dumbbells is about a hundred times lower at $T \approx 10$ K. It has been found possible to observe this slow tunneling relaxation because of the use of a low acoustic frequency, $\omega \approx 10^4$ s^{-1}, for which the dispersion band coincides with the region in which $\tau(T)$ changes in the tunneling regime.

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