line is a theoretical plot of an exponential decay $\exp(-t/6\times10^{-4})$, while the dashed curve represents the level to which the energy stored in the line under observation decays by cross-relaxation. The energy of the three lines then decays simultaneously from this level with a time constant equal to T_1. No attempt was made to fit a curve to the experimental points of the echo signal amplitudes, but the smaller values obtained at the start of the decay can be justified by the method used to measure the echos.

It is of importance to note that the spin-echo decay continues at the cross-relaxation rate even after the three lines of calcite have come to energy equilibrium. This is to be expected on the basis of the model proposed by Bloembergen et al.[1] Since the balance of energy between the three lines has the characteristics of a dynamic equilibrium and the very nature of cross-relaxation allows mutual spin flips between electrons with somewhat different resonant frequencies to transfer energy between spectral lines, the frequency storage of the spins is destroyed by the cross-relaxation process. The stimulated echo depends on the Z-axis storage of this frequency dependence and the echo signal consequently decays exponentially to zero with the cross-relaxation time constant.

This same process can occur within a single inhomogeneously broadened line and might make a very interesting study, since the stimulated spin-echo technique effectively separates the decay due to cross-relaxation from that due to spin-lattice relaxation when the time constants are not comparable.

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[1]N. Bloembergen, S. Shapiro, P. S. Pershan, and J. O. Artman, Phys. Rev. 114, 445 (1959).

[2]J. P. Gordon and K. D. Bowers, Phys. Rev. Letters 1, 368 (1958).

[3]E. L. Hahn, Phys. Rev. 80, 580 (1950).

[4]J. Kemp, J. Chem. Phys. 33, 1269 (1960).

[5]S. Fernbach and W. G. Proctor, J. Appl. Phys. 26, 170 (1955).

DISLOCATION RELAXATION SPECTRA OF COLD-WORKED BODY-CENTERED CUBIC TRANSITION METALS*

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During the course of a low-temperature acoustical study of the defect structure introduced into bcc transition metals by cold work, peaks have been found in the internal friction vs temperature curves of Nb, Ta, Mo, and W. The peaks appear only with prior plastic deformation and have a detailed shape which is controlled by the annealing history of the specimen.

Figure 1 shows the internal friction vs temperature curves for fine-grained vacuum-melted Nb, Ta, Mo, and W specimens given a 3% tensile prestrain at 300°K (~600°K for W). An internal friction spectrum of a typical fcc metal (Cu) prestrained 3% is included for comparison. Most of the acoustical measurements were made by the longitudinal resonant bar method at 15 kc/sec and oscillating strain amplitudes of $\sim10^{-7}$; several measurements on Mo were made at 6 cps, at a strain amplitude of 2×10^{-7} in a torsion

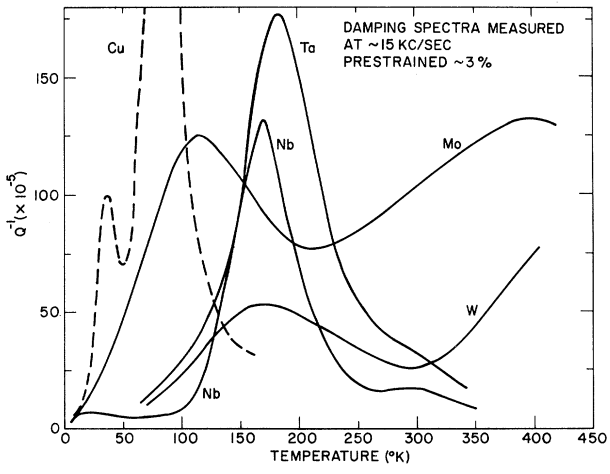

FIG. 1. Damping spectra of polycrystalline Cu, Nb, Ta, Mo, and W measured at ~15 kc/sec. All specimens were given ~3% tensile prestrain at 300°K (~600°K for W).

pendulum. It can be seen that at 15 kc/sec the Nb and Ta spectra have a large peak at 170°K and 180°K, respectively, and a much smaller one near 310°K. The Mo spectrum at 15 kc/sec is divided into two broad peaks, a low-temperature peak (α) at 115°K and a higher temperature peak (β) near 400°K; the β peak is located at 240°K for measurements made at 6 cps. The 15-kc/sec W spectrum consists of an α peak at 170°K and a β peak which appears to exist somewhere above 450°K.

Figure 2 shows evidence of structure in both the α and β peaks of fine-grained Mo prestrained in tension one percent. The observed structure resulted from a series of one-hour vacuum anneals at successively increasing temperatures. The β peak begins to break up into at least three subpeaks located at about 260°K, 320°K, and 400°K for a 300°C, one-hour anneal, then virtually disappears after a 400°C, one-hour anneal. The α peak shifts to lower temperatures and shows evidence of structure with a slight shoulder on its high-temperature side. The lower peaks in both Nb and Ta shift to lower temperatures on low-temperature annealing.

There is additional evidence for believing that the observed internal friction spectrum in Mo is the sum of a number of subpeaks. The ratio of the quantity $(Y - Y')/Y_0$, where Y' is the relaxed component, Y the elastic component of the Young's modulus, and Y_0 the elastic component of Young's modulus measured at 5°K after a 800°C anneal, to Q_m^{-1}, where Q_m^{-1} is the height

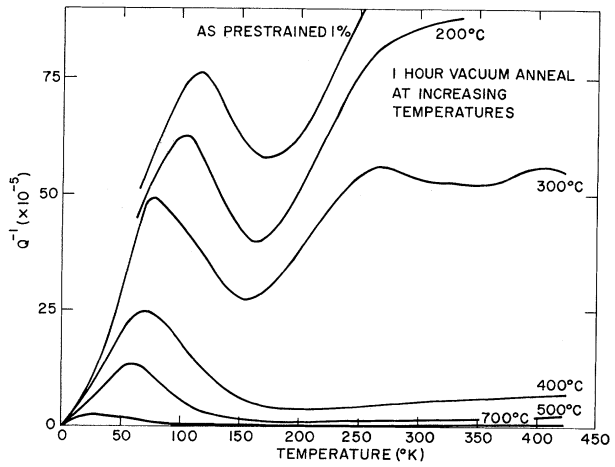

FIG. 2. Effect of successive one-hour vacuum anneals on the damping spectra of polycrystalline Mo measured at ~15 kc/sec. The specimen had received ~one percent prestrain at 300°K.

of the internal friction peak, is about 8; whereas a single relaxation process would yield a ratio of 2.[1] Also, careful examination of the Young's modulus vs temperature curve shows the existence of a number of slight but distinct discontinuities in the slope of the curve at temperatures corresponding to the location of the suspected subpeaks. These discontinuities are similar to those found in cold-worked Cu single crystals by Thompson and Holmes.[2]

All four of these metals also show the strong amplitude dependence in the internal friction and Young's modulus characteristic of pinned dislocation motion.[3] It is observed that the annealing of the peaks is always accompanied by a strong (10- to 50-fold) increase in the breakaway amplitude. This effect was first noted in earlier work on Nb by the authors[4] and attributed to the pinning of dislocations by the diffusion of point defects (gaseous interstitials) to dislocations. The present data indicate that this mechanism can be extended to include Ta, Mo, and W.

The peaks observed here are similar in several respects to those produced by cold-working fcc metals (the Bordoni peaks)[5]: Both possess strong dependence on cold work, have a wide distribution of relaxation times, and both occur, for a comparable measuring frequency in a similar reduced temperature range, i.e., temperature normalized to the melting point of the metal. Furthermore, the attempt frequency, f_0,[6] in the relation $f = f_0 \exp(-H/RT)$ for the β peak in Mo is $10^{10.4 \pm 0.4}$ sec^{-1}, which is comparable to that found for the Bordoni peaks in fcc metals. On the other hand, the peaks in bcc metals all anneal at a considerably lower reduced temperature than do the peaks in fcc metals; and the bcc α-peak positions show a marked shift toward lower temperatures with annealing in contrast to a similar but smaller shift on annealing found in the Bordoni peaks of fcc Cu.

It has also been observed that variations in the concentration of gaseous impurities affect principally the annealing characteristics of a given peak, while the peak temperature and height remain essentially unchanged. A more complete account of these impurity effects and their implications regarding the nature of the defect mechanisms operating here will be published elsewhere.

On the basis of the above observations, the authors consider that the defects responsible for the relaxation spectra (especially the β peaks) in cold-worked bcc metals are similar to those that

produce the Bordoni peaks in fcc metals. Furthermore, we feel that the different response of the bcc metals to annealing can be attributed to the action of efficient dislocation pinning points (gaseous interstitials and/or carbon) known to exist in bcc metals.

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[1]C. Zener, Elasticity and Anelasticity of Metals (University of Chicago Press, Chicago, Illinois, 1949), p. 47.

[2]D. O. Thompson and D. K. Holmes, J. Appl. Phys. 30, 525 (1959).

[3]A. Granato and K. Lücke, J. Appl. Phys. 27, 583 (1956).

[4]R. H. Chambers and J. Schultz, Acta Met. 8, 585 (1960).

[5]P. G. Bordoni, J. Acoust. Soc. Am. 26, 495 (1954).

[6]A. Seeger, H. Donth, and F. Pfaff, Discussions Faraday Soc. 23, 19 (1957).

ABSENCE OF AN ISOTOPE EFFECT IN SUPERCONDUCTING RUTHENIUM

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Within the accuracy of our measurements, (0.001°), naturally occurring ruthenium and its isotopes of mass 99 and 104 have identical superconducting transition temperatures, T_{sc}. Our results are presented in Fig. 1. This lack of dependence of T_{sc} on atomic mass is quite different from the results obtained for the isotopes of other superconducting elements.[1] All elements which have been previously measured, Sn, Hg, Tl, and Pb, show an isotope shift of T_{sc} which is essentially proportional to $M^{-1/2}$. However, they are all nontransition elements, having s and p electrons only. Ruthenium is the first of the transition elements to be checked with respect to an isotope shift. The reason for this lack of data may have been—in part, at least—experimental difficulties. The superconducting properties of ruthenium and osmium have been discovered and reported by Goodman[2] and Hulm and Goodman.[3] With the exception of these two metals, transition element superconductors have critical temperatures which change strongly with small amounts of dissolved nitrogen or oxygen. These variations exceed the shift that may be expected from an isotope effect. There is no good way at present to get rid of these gases in a quantitative way with the small amounts of material available.

Ruthenium isotopes were obtained in powder form from Oak Ridge National Laboratory through the courtesy of P. V. Arow. The isotope analysis furnished is given in Table I. After our initial results, a second analysis was made at the Oak Ridge Laboratory which was in good agreement with the first. Spectrographic analysis at Oak Ridge indicated an absence of all transition metal impurities which might falsify the results, with the possible exception of iron. However, a further analysis indicates less than 10 parts per

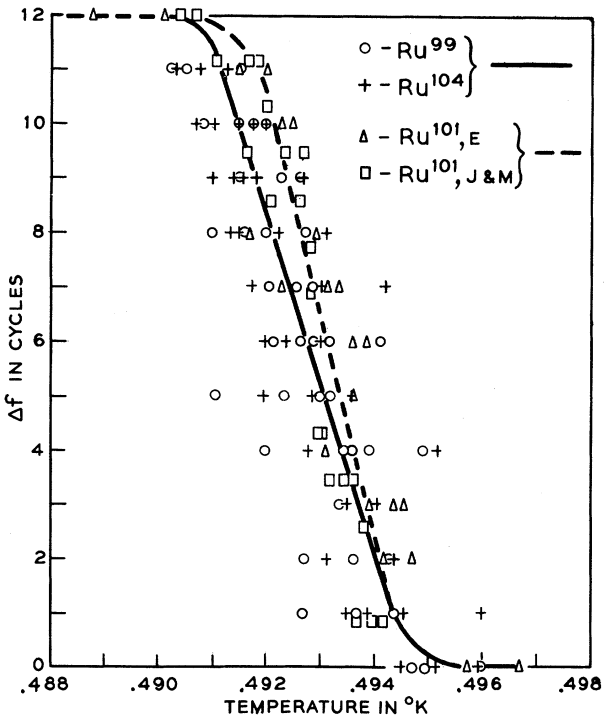

FIG. 1. Change in frequency of oscillator circuit as a function of temperature for arc-melted pellets of ruthenium. Samples Ru^{99} and Ru^{104} were made from enriched powders of Ru^{99} and Ru^{104}; sample J&M was made from Johnson-Matthey powder and sample E from Englehard Industries' natural powder.