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The vacancies, lubrication agents of dislocation motion in aluminium

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Abstract. Measurements of the dynamic modulus and internal friction of ultra-high-purity aluminium were performed during irradiation with fast electrons at 5.7 K (or 60 K) and subsequent anneals, using a vibrating strip system. The salient result is that the introduction of point defects on the existing dislocations during irradiation at very low temperature, when the vacancies and self-interstitials created are immobile, provokes a substantial *decrease* in the elastic modulus. This type of evolution does not originate in an elastic bulk effect but is instead shown to result from the lubrication of dislocation motion by the deposited defects. This finding substantiates the concept of an athermal lubrication mechanism proposed recently for the interpretation of cold-work results.

Further insight into the nature of the defect species responsible for this lubrication has been attained by the realisation of irradiation and anneal sequences at different temperatures. It appears that, contrary to that which happens with self-interstitials, which are found to act as dislocation pinners, as is normally the case, the vacancies facilitate the motion of dislocations down to the lowest temperatures explored.

1. Introduction

In a recent internal friction experiment carried out in ultra-high-purity aluminium, a decrease in the elastic modulus was observed after cold work at 4.2 K [1]. This effect is in good agreement with the results of another series of experiments performed in similar conditions, with use of the acoustic coupling technique [2,3], if it is assumed that point defects can activate a lubrication process for dislocation motion, operative down to very low temperatures. However, further investigation is required to ascertain that the point defects produced by the cold work are responsible for this effect, rather than changes in the dislocation network. Subsidiarily, more detailed information is needed to determine which defects, vacancies or self-interstitials are the real lubrication agents.

Irradiation with energetic particles appears to be the most appropriate approach for obtaining an unambiguous answer to this question. In fact, it enables one to vary the concentration of the elementary point defects separately, while the dislocation density and array are kept strictly unchanged, thus making the discrimination of the true role of the potential partners in the mechanism alluded to possible. Accordingly, we initiated a study of this type. It is based on the realisation of modulus and damping measurements during the course of irradiation with fast electrons and also during post-irradiation anneal cycles. Irradiation was either at temperatures at which the created defects are frozen-in or, conversely, when the self-interstitials exhibited significant mobility. As will appear in the following, comparison of the results for these two irradiation conditions, together with consideration of post-irradiation annealing effects, enabled clear separation of the respective roles of the two defect species produced, vacancies and self-interstitials.

2. Experimental procedure

The material studied was ultra-high-purity aluminium prepared by zone melting at the Centre d'Etudes de Chimie Métallurgique de Vitry sur Seine, France. Spectrometric analysis and neutron activation indicated a total impurity concentration well below 1 at. ppm [4].

The samples were rectangular plates 25 mm long, 5 mm wide, with thickness 200 μm, cut by spark erosion from a single crystal and polished mechanically. These strips were clamped in a copper block in which a heat exchanger was incorporated. A cryogenic set-up enabled cooling down to 4.7 K, the temperature at which liquid helium boils under a pressure of 1.2×10^5 Pa. A Rh–Fe resistance was then used to monitor the sample temperature. For the measurement of damping and dynamic modulus, the reeds were electrostatically driven under vacuum, in flexural vibrations with typical strain amplitude, 10^{-6}, and frequency, 10^3 Hz. Automatic data acquisition enabled direct recording of the relevant parameters, frequency, f, damping, Q^{-1}, irradiation flux, Φ, and absolute temperature, T, during the irradiations and the anneals. It is recalled that the relative variations of elastic modulus are proportional to the relative frequency variations, squared [5].

The device considered is very versatile. It can be operated in-line with a van de Graaff accelerator, in the irradiation facility of the Centre d'Etudes Nucléaires de Grenoble [6]. Cold-work and heat treatments can also be performed *in situ*, in the temperature range 4.7–370 K. For present purposes, fast electrons were used, with an energy of only 0.5 MeV (compared to values of 1–3 MeV used in most previous studies). This energy was selected in order that most of the Frenkel pairs created in the metal target were elementary pairs, not multiplets, thus making experimental conditions ideally simple. More precisely, the maximum recoil energy, R, is then about 56 eV, which is not very high compared to the displacement threshold energy of 18 eV determined in prior work [7]. On account of the fact that the Rutherford diffusion law, which governs the damage, strongly favours the energy transfers with lower energy (the differential displacement cross section, $d\sigma/dR$, is proportional to R^{-2}), it can be considered that essentially isolated pairs are produced. On the other hand, it is underlined that low flux rates were used, close to 1×10^{12} electrons cm^{-2} s^{-1} (160 nA cm^{-2}). In addition, the integrated fluxes were generally as low as 10^{16} to 10^{17} electrons cm^{-2}. Taking a displacement cross section of 35 ± 5 barn [8], these fluxes yield defect concentrations smaller than a few atomic parts per million (3 at.ppm at the highest dose). These are the most appropriate conditions for a straightforward interpretation of the results.

3. In-flux measurements: evidence of a modulus decrease due to intrinsic point defects

Figure 1 shows the modulus variations that were found to occur in response to the application of an electron flux. Prior to the beginning of the irradiation, the sample was

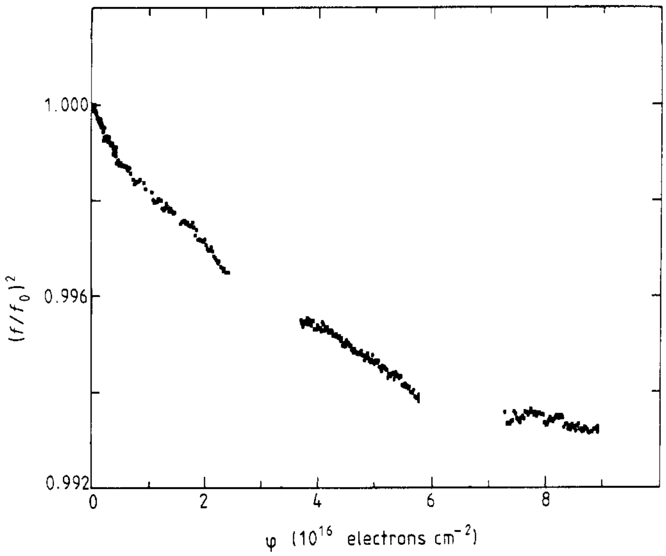

Figure 1. Modulus variations against irradiation dose, recorded during the application of a permanent electron flux, at a constant rate of 1×10^{12} electrons cm^{-2} s^{-1}. Temperature was 5.7 K and initial frequency 800 Hz. The modulus decreased monotonously during the whole course of the irradiation. Indicatively, on account of the very low irradiation dose, the total concentration of stored-in Frenkel pairs did not amount to more than a few atomic parts per million.

annealed at 370 K and its initial resonant frequency determined at 4.7 K. Subsequently, irradiation was triggered at an approximate flux rate of 1×10^{12} electrons cm^{-2} s^{-1}, which resulted in a temperature increase of about 1 K. Measurements were then performed during the course of permanent irradiation. However, from time to time irradiation was stopped momentarily so that the modulus changes could be measured at the reference temperature of 4.7 K. This was to verify that the evolution observed did not originate from an eventual temperature shift under flux but instead stemmed from the storing of the radiation defects.

The most characteristic feature of the modulus evolution is its actual sign. Surprisingly enough, a monotonous *decrease* takes place, starting from the very beginning of the irradiation. It is steady and reaches a relative value of -7×10^{-3} for an electron dose of roughly 10^{17} electrons cm^{-2}. This profile was observed reproducibly, in well annealed aluminium and also for various pre-anneal conditions, as reported in § 4.

4. Origin of the modulus decrease: elastic bulk effect versus dislocation lubrication mechanism

The basic phenomenon to be explained is the marked decrease of elastic modulus that occurs during the 5.7 K irradiations. Clearly, it is the newly produced elementary point defects that are responsible for these modifications in the elastic properties of aluminium. However, for the interpretation of the observed decrease, two very distinct processes can be envisaged. (1) The most natural explanation lies in an intrinsic effect associated with the presence of the frozen-in self-interstitials created in the bulk material by the impinging particles. That is that due to their inherent elastic polarisability, these defects

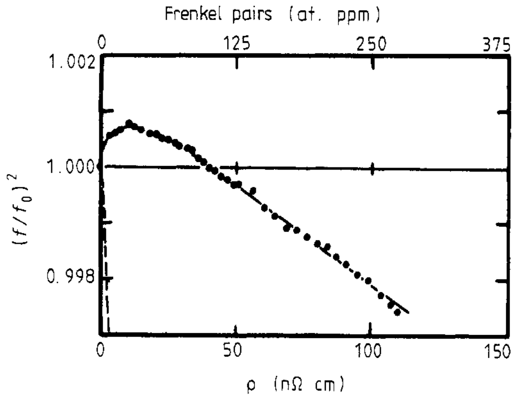

Figure 2. Dose dependence of the relative change of torsional frequency, squared, of an aluminium ⟨111⟩ crystal (after Robrock and Schilling [9]). Irradiation was at 4.5 K with 3 MeV electrons at a dose rate of 5×10^{13} electrons $\mathrm{cm^{-2}\,s^{-1}}$. The frequency variations, squared, determined in present work are sketched by the broken curve. The respective evolutions for the two cases considered are quite different, both in character and order of magnitude.

can be expected to give rise to a marked modulus decrease, on the application of a mechanical stress, as is the case in the present experiments. This evolution is generally termed the elastic bulk effect (see below). (2) Another plausible explanation lies in the idea of an enhanced dislocation mobility arising from a specific interaction between the pre-existing dislocations and the fresh radiation (point) defects, according to the scheme alluded to in the introductory presentation. This alternative is now discussed.

An interesting reference in this discussion is the work by Robrock and Schilling [9], who examined the modulus changes in aluminium during and after electron irradiation at 4.5 K. Although the data were obtained in aluminium with lower purity than ours and for more energetic (3 MeV) electrons, valuable information can be extracted from their measurements, which were performed on single crystals using an inverted torsion pendulum vibrating in the range 20–80 Hz. These measurements yielded accurate values of the variations of the elastic shear moduli c_{44} and $c' = (c_{11} - c_{12})/2$ induced by the irradiation. Following an initial increase, due to dislocation pinning, a downturn shows up before a linear decrease takes over. It is spread over the whole subsequent evolution, up to the higher defect concentrations studied (about 250 at.ppm) as is seen in figure 2. This latter effect is directly proportional to the radiation-induced defect density. On the basis of this in-flux behaviour, together with the evolution during subsequent temperature pulses, the authors interpreted the data in terms of a softening of the lattice via a dia-elastic polarisation of the strain fields associated with the self-interstitial defects present in the bulk. Incidently, this is clearly distinguished from the stress-induced reorientation relaxation associated with the elastic dipole moment characteristic of the split interstitials. The reorientation gives rise to an additional decrease in the elastic constants but, unlike the dia-elastic polarisation, the reorientation process is thermally activated, so that the corresponding decrease in modulus is observed only above a characteristic temperature, namely 30 K, in aluminium [10]. The absolute magnitude of the elastic bulk effect was found to be rather large, since it amounts to about 20–25% per atomic per cent of Frenkel pairs. Indicatively, similar values were obtained in other FCC metals, in particular copper (for a review, see Rehn and co-workers [11]).

This information is of considerable assistance in interpreting our data. In the present case, although the dose rate and integrated dose are much smaller, no upward transient is detected at the beginning of the irradiation. Further, the decrease rate is larger by a factor of 40 than the one characteristic of the bulk effect. We are then led to the unequivocal conclusion that we are dealing at present with a phenomenon absolutely distinct from the one(s) reported earlier in the literature to explain the lowering of the low-temperature modulus consequent upon irradiation. Additional information is

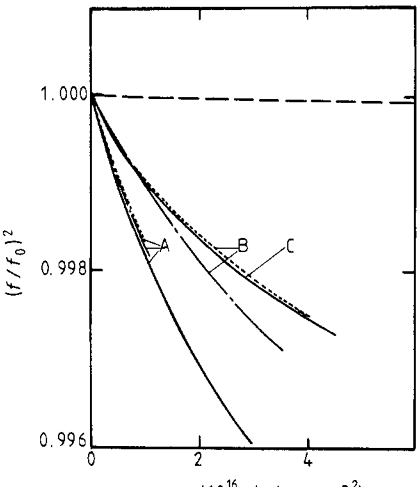

Figure 3. Dose dependence of the relative change of flexural frequency, squared, of polycrystalline aluminium samples, for different structural states: A, preannealed at 370 K; B, the same, followed by slight cold work (0.2%) at 4.7 K and a further anneal at 150 K or 200 K; C preannealed at 370 K and preirradiated at 5.7 K and 60 K. The broken curve refers to previous data on the elastic bulk effect in aluminium [9].

obtained from consideration of the effect of the state of anneal of the samples on the result, as illustrated in figure 3. The rate of decrease appears to exhibit a significant curvature and also slope dependence on the existing dislocation density and arrangement, as produced by various heat treatments after deformation. Neither of these features is expected for a bulk effect. On the contrary, these remarks suggest that dislocations are involved. One is then brought back to our initial suggestion that the mobility of dislocations is enhanced by the presence of defects interacting with them. This concept of a dislocation lubrication mechanism by the point defects located in the core of dislocations has been proposed for the first time by Benoit, Bujard and Gremaud [3, 12] on basis of employing acoustic coupling measurements to monitor changes in dislocation mobility. In this original scheme the point defects located on the stacking fault ribbon between dissociated dislocations were supposed to make the motion of these defects easier (with the result of a lowering in the elastic modulus) by allowing a very easy kink pair formation at temperatures well below the ones where thermally activated formation of kink pairs takes place in the absence of lubricating point defects.

5. Identification of the defect species responsible for dislocation lubrication

From the in-flux behaviour of the elastic modulus, it appears that the point defects produced in the vicinity of the dislocation cores and interacting elastically with them indeed act as lubricants of the dislocation motion. Deeper insight into the nature of the point defects that are the actual lubricators—vacancies or self-interstitials—is obtained from consideration of the effect of the irradiation temperature on the in-flux evolution sign of the modulus and by analysis of the general behaviour of this physical parameter during post-irradiation annealing.

After every irradiation, a sample was given successive linear heating runs at progressively increased temperatures, followed by a rapid return to 4.7 K and a modulus measurement at this temperature. Two examples of the evolution profiles of modulus with temperature are presented in figures 4 and 5. A common feature is that the initial modulus decrease produced by the irradiation is not reinforced by the anneal but, on the contrary, the modulus continuously increases on annealing at progressively increased temperatures. More precisely, two regimes are distinguished for this evolution: a progressive, slow enhancement spread from about 20 K to 45 K, followed by a drastic

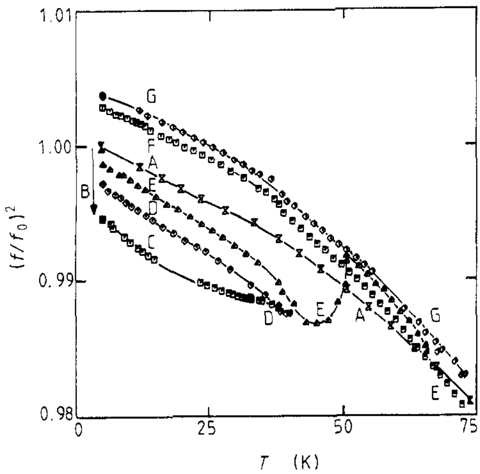

Figure 4. Evolution of modulus as a function of temperature recorded during heating runs following irradiation at 5.7 K at a dose of 5×10^{15} and 0.5×10^{15} electrons cm^{-2}, respectively. The consecutive runs are labelled by the letters A to G: A, preirradiation reference; B, irradiation; C to G, measurements during heating up to 35 K, 40 K, 65 K, 80 K and 200 K, respectively. The average heating rate was about 1 K min^{-1}.

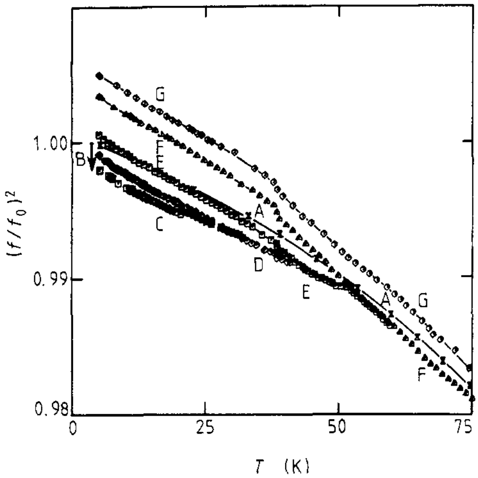

Figure 5. As for figure 4, but C to G taken up to 30 K, 40 K, 60K, 80 K and 100 K, respectively.

increase just above this temperature, specially for the higher electron flux. This abrupt variation is located exactly in the temperature interval in which the interstitial defects undergo correlated or uncorrelated random diffusion, beginning typically at 40 K, as inferred from prior work on aluminium (stages $I_D + I_E$ in the usual terminology) [13]. One is then led to conclude that the freely migrating self-interstitials that reach the dislocation lines provoke their pinning. This is the customary pinning effect reported in a variety of metals under irradiation conditions. Now, to interpret the modulus increase observed for anneal temperatures well below the ones at which self-interstitials diffuse freely, several reasons can be invoked. First, although the elastic bulk effect is much smaller than the lubrication effect evidenced, its weakening due to the progressive annihilation of the close pairs, which is expected to occur between 20 K and 40 K in aluminium [14], which is precisely in the range under consideration, might at least qualitatively explain the observed trend. This is probably too small an effect to account for the magnitude of the measured variation. Secondly, in the lubrication scheme, a modulus enhancement can possibly be caused by the annihilation of the pre-existing lubricating agents. This can occur because of the arrival of faster moving defects, the self-interstitials, at dislocation sites. The important implication would then be that the lubrication agents are of the vacancy type. Since this already takes place below stage I_D, no long-range diffusion is involved. Only the self-interstitials created in the strain field of dislocations or diffusing along the dislocation lines are able to play such a role. This is a likely explanation. At higher temperatures, when the self-interstitials start diffusing into the bulk, their flux towards dislocation sinks is considerably larger, thus resulting in a more marked modulus increase. An alternate, plausible explanation of the earlier, progressive modulus increase lies in the rearrangement of the lubricators along the dislocation lines. This scheme will receive detailed attention in a forthcoming paper about the effect of cold work on modulus [1].

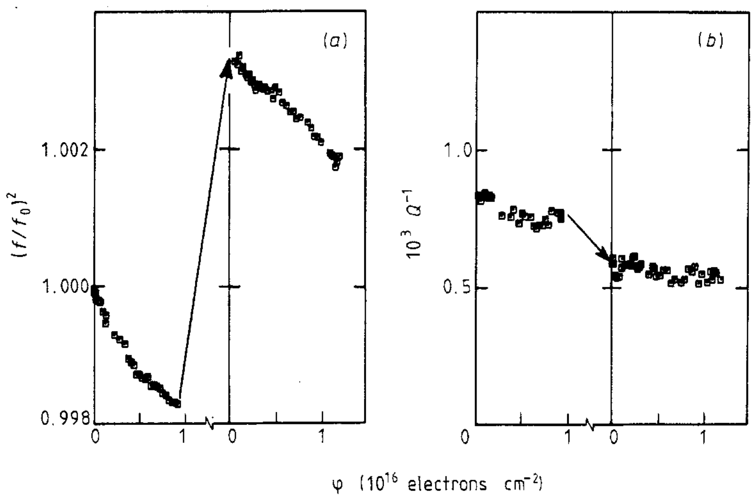

Figure 6. (*a*) Modulus changes and (*b*) damping recorded during two distinct irradiation experiments at 5.7 K, with an intermediate anneal at 80 K, obtained by a linear heating to this temperature, at a rate of 2 K min^{-1}. In both cases, the modulus decreased monotonously. The corresponding internal friction did not vary significantly during either irradiation. However the annealing treatment resulted in a lower damping at 5.7 K, in conformity with the expectation for dislocation pinning.

Finally, it can be conjectured that the modulus increase manifested reflects a saturation effect of the lubrication process, as soon as annealing is started, followed by an inversion. A specific experiment sheds light on this eventuality. A sample was first irradiated at 5.7 K, in the well annealed condition. Then it was given an anneal at 80 K, where pinning definitely takes place, as revealed by a modulus increase beyond the pre-irradiation value. Then the temperature was brought back to 4.7 K and irradiation was started a second time. The in-flux variation of modulus was found again to be a decrease, as evidenced in figure 6. Surprisingly enough, the rate of decrease of modulus was found to be about the same for the two cases examined, the one of virgin dislocations and the other of dislocations having collected the self-interstitials produced by the irradiation.

These observations were further complemented and confirmed by a series of irradiation experiments carried out alternately at 5.7 K and 60 K, that is at temperatures when the self-interstitials are immobile or, inversely, undergo long-range migration. The corresponding modulus and damping behaviours are displayed in figure 7. The most striking feature is that, while the modulus gets steadily less during the course of the two 5.7 K irradiations, it progressively increases during the 60 K irradiations. Thus the modulus variations are opposite in sign at these two irradiation (and measurement) temperatures. Accessorily, it is verified that the 5.7 K modulus is also enhanced, even more than the 60 K modulus, by the high temperature irradiation.

Taken together these results show the inverse role played by vacancies and self-interstitials with respect to dislocation motion. In short, as is widely recognised, the self-interstitials essentially impede the motion of dislocations. In contrast, the vacancies make it smoother, which is quite a discovery.

This lubricating action of vacancies is still more selectively evidenced by studying the evolution of the modulus consequent upon anneals extended to the temperature region where the radiation-created vacancies that have survived to the stage I recovery become mobile. This is known to occur around 250 K or above, in aluminium (stage III of

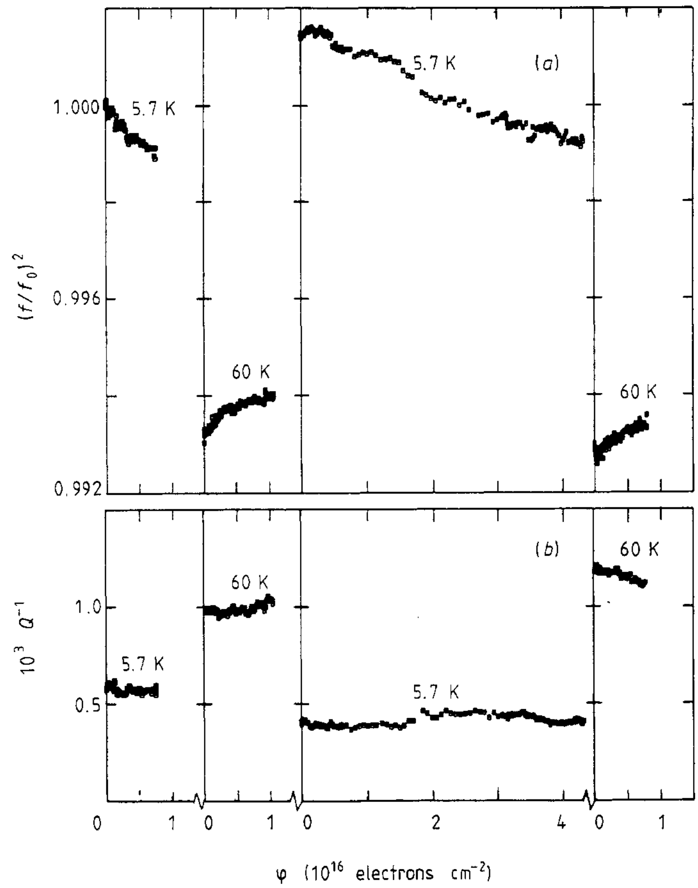

Figure 7. (*a*) Modulus measurements performed during alternate irradiations at 5.7 K and 60 K, at the same dose rate of 1×10^{12} electrons $cm^{-2}\,s^{-1}$. The evolution sign is reproducibly inverted when temperature is changed. (*b*) Concurrently, internal friction was measured. No significant evolution was observed at either irradiation temperature.

irradiated aluminium), depending on the dislocation density and vacancy population involved. Figure 8 clearly shows that the incoming flux of vacancies at dislocations results in a significant modulus decrease, in accordance with the expectation for a lubrication mechanism.

It is interesting also to consider the effect of quenching on dislocation mobility. Unfortunately, the available information is scarce. What is more, the data were collected after quenches from too high temperatures, that is in conditions when the vacancy supersaturation is too large to avoid the formation of multiple vacancies during the fast cooling and subsequent anneals [14, 15]. Young's modulus of aluminium was then observed to be decreased by the presence of the frozen-in vacancies. This was correctly assigned to an elastic bulk effect. Indeed this effect is weak, typically a few % per at.% vacancies, and cannot be traced to dislocation lubrication. Clearly more appropriate experimental conditions are required to observe the lubrication that can be predicted on basis of the present experimental findings.

The present analysis of the results enables us to answer at least in part the puzzling questions concerning the reaction partners required in order that lubrication take place and the detailed mechanistic process that makes the dislocation motion easier, even

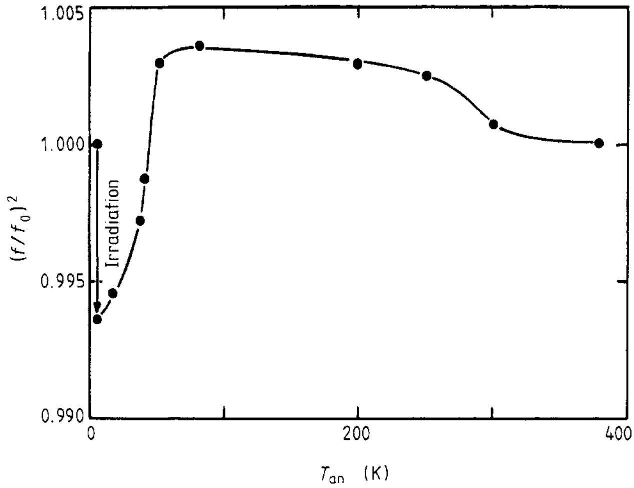

Figure 8. Modulus variations observed as a result of post-irradiation anneals at successively increased temperatures up to 380 K. Note the softening stage around 270 K, when the vacancies undergo long-range diffusion. All measurements were at 4.7 K.

at the lowest temperatures explored. More generally, these questions pertain to the 'tribology' of dislocations in FCC metals.

On the basis of a model suggested recently [3], a description of the underlying mechanism can tentatively be made in terms of a local modification of the stacking-fault ribbon width, which would make the non-thermally activated formation of double kinks, down to the lowest temperatures, possible. This scheme will be discussed in more detail, together with pertinent cold-work results, in a forthcoming paper [1]. Not only vacancies but also selected impurities in very low concentration can give rise to the process suggested, which explains the surprising absence of increase of the elastic limit, which is obvious in FCC metals, when the temperature is decreased below the one at which the thermally activated formation of double kinks proceeds.

6. Conclusion

The effect on the elastic modulus of minute concentrations of elementary Frenkel pairs selectively introduced by irradiation with fast electrons at 5.7 K—that is, without modification of the dislocation density and array—enabled us to perceive hitherto undetected elastic effect. It was clearly distinguished from the dia-elastic or elastic bulk effects or the dislocation pinning effects usually found in irradiated metals. Detailed analysis of this original effect, which is indeed very marked in the appropriate conditions present, showed that it arises from a specific interaction of intrinsic point defects with dislocations, giving the result of a softening of dislocation motion. It was further inferred that the agents of this lubrication are the vacancies, while the self-interstitials predominantly govern all the other effects mentioned above.

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References

[1] Lauzier J, Hillairet J, Benoit W and Gremaud G 1989 to be published
[2] Gremaud G, Bujard M and Benoit W 1987 *J. Appl. Phys.* **61** 1795
[3] Benoit W, Bujard M and Gremaud G 1987 *Phys. Status Solidi* a **104** 427
[4] Revel G 1965 *PhD Thesis*, Centre d'Etudes de Chimie Métallurgique, Vitry sur Seine
[5] Minier C, Duclos D and Lauzier J 1983 *J. Phys. F: Met. Phys.* **13** 13
[6] Minier C, Lauzier J, Disdier F and Lagnier R 1983 *J. Physique Coll.* **44** C9–377
[7] Wollenberger H J 1970 *Vacancies and Interstitials in Metals* ed. A Seeger, D Schumacher, W Schilling and J Diehl (Amsterdam: North Holland) p 217
[8] Oen O S 1973 *Cross Sections for Atomic Displacements in Solids by Fast Electrons, ORNL Report 4897UC34* (Springfield, VA: National Technical Information Service, US Department of Commerce)
[9] Robrock K H and Schilling W 1976 *J. Phys. F: Met. Phys.* **6** 3
[10] Spiric V, Rehn L E, Robrock K H and Schilling W 1977 *Phys. Rev.* B **15** 672
[11] Rehn L E, Holder J, Granato A V, Coltman R R, Young F W Jr 1974 *Phys. Rev.* B **10** 349
[12] Bujard M, Gremaud G, Benoit W 1987 *J. Appl. Phys.* **62** 3173
[13] Maury F, Lucasson P, Lucasson A, Vajda P, Dimitrov C, Dimitrov O, Le Hericy J and Mathieu G 1982 *Point Defects and Defect Interactions in Metals* ed. J Takamura, M Doyama and M Kiritani (Tokyo: University of Tokyo Press) p 383
[14] Keefer D, Robinson J C and Sosin A 1968 *Acta. Metall.* **16** 927
[15] Folweiler R C and Brotzen F R 1959 *Acta Metall.* **7** 716