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MODELS FOR FROZEN-FREE-SPLIT PHENOMENA IN e⁻-IRRADIATED Al AND Al-Fe

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Abstract.—Samples of pure Al and Al containing 400 atppm Fe have been irradiated with electrons at 4.2K and annealed at subsequently higher temperatures. During the annealing program in pure Al, the growth and decay of two pairs of mechanical relaxation peaks was observed, and in Al-Fe of one pair. The members of each pair grow and decay in a parallel fashion. They are explained as being due to the stress induced occupation of two different sets of elastic energy levels via two different relaxation modes by the reorientation of the same defect. For the pairs in pure Al and for the pair observed in Al-Fe atomic models are given based on computer simulation studies.

Introduction.—Many examples are known where irradiation induced self-interstitial atoms give rise to mechanical relaxation peaks /1,2/. In the case of pure Al, these peaks were attributed to single and multiple self-interstitials (S) /3,4/, and in Al-Fe to so-called SI-complexes, which consist of one or several self-interstitials bound at an impurity atom (I) /5/. Since the peak heights are proportional to the concentrations of the defects responsible for the peaks, it is possible to follow the change of concentrations of individual defects during the course of an isochronal tempering treatment. In some cases it was observed, that two peaks of the relaxation spectrum showed the same annealing behaviour in contrast to the different behaviour of the others. This parallel annealing must be considered as a very strong indication, that each peak doublet is caused by one defect via two separate relaxation modes rather than by two different defects. This phenomenon has been named "frozen-free-split"-phenomenon in the literature /6/. The aim of the present article is to describe atomistic models, by which the occurrence of double relaxation modes observed in pure Al and Al-Fe can be explained.

Experimental.—Details of the experiments are described elsewhere /3,4,5/. The irradiations were performed at the low temperature irradiation facility of KFA Jülich at 4.2K with 3 MeV electrons. The elastic after effect measurements on Al were carried out in a torsional pendulum using a laser interferometer technique, the internal friction measurements on Al and Al-Fe in the same apparatus, using an electromagnetic pick up system. The concentration of radiation induced defects were monitored by simultaneously irradiated resistivity samples.
Results.- In fig. 1 and 2, the results are shown. The respective upper figures display the relaxation spectra of electron irradiated Al and Al-Fe, for a fixed relaxation time. The bars indicating the peaks are positioned at those temperatures, where the processes have a relaxation time of 100s (Al) and 0.04s (Al-Fe), respectively. The heights of the bars correspond to the maximum peak heights, $\Delta_0$, obtained during the tempering treatment.

![Fig. 1](image1)

![Fig. 2](image2)

The respective upper figures represent a schematic view of the mechanical relaxation spectrum of electron irradiated Al at $\tau = 100$ s and Al-Fe at $\tau = 0.04$ s, where $\tau$ is the relaxation time. The relaxation strength, $\Delta_0$, is the largest value obtained during a 10 min isochronal annealing treatment. The respective lower curves show the build-up and recovery of the different relaxation peaks observed during the annealing treatment.

Below the normalized peak heights, $\Delta/\Delta_0$, are plotted as a function of annealing temperature. In pure Al, process $M_1$ has been identified as the migration and simultaneous reorientation process of the single self-interstitial in form of 100-dumbbells /3/. As these migrate, they may encounter other self-interstitials or the Fe-atoms in solution, with which they form either multiple self-interstitials in the case of pure Al, or SI-complexes in the case of Al-Fe. The fact that processes $R_0$, $R_2$ and partially $R_4$ in Al as well as processes 1 and 4 in Al-Fe grow parallel with the decay of $M_1$ has led to the conclusion, that these processes are caused by reaction products of the migrating self-interstitials: Multiple self-interstitials in the case of pure Al and SI-complexes in the case of Al-Fe. The remarkable feature is not only that all these processes appear in a parallel fashion, but that they follow pairwise the same behaviour during the subsequent tempering treatment:
The \((R_2,M_2)\)-doublet decays in a narrow temperature range around 47K, the \((R_0,R_1)\)-doublet remains constant up to about 60K and also decays in a narrow temperature range to zero. Finally, the \((1,4)\)-doublet in Al-Fe decays in a parallel fashion by about 15% between 80K and 200K, and then in a step to zero at 220K.

The annealing curves of the other processes \(R_1\), \(R_3\), \(R_5\) and 2, 3, 5, 6 follow entirely different trends than those of the paired peaks.

**Discussion.** The parallel behaviour of the two members of each of the pairs is a rather strong indication, that they result from the reorientation of the same defect via two different relaxation modes rather than from two different defects, which undergo fortuitously the same reactions during the tempering treatment. This view is supported by recent computer simulation studies /7-10/ of the stability and mobility of different self-interstitial-configurations which will be discussed in the following.

One of the outstanding properties of the \(<100>\) dumbbell self-interstitial is its capability to perform low frequency resonant libration modes. These have been found for single \(<100>\) dumbbells /8/, for dumbbells which are members in a multiple interstitial /7/ and also for mixed dumbbells /8/. These libration modes are very sensitive to perturbations, and can be made locally unstably by smallest changes in the potential used in the computer simulation. These local instabilities may cause a tilt of the dumbbell away from the exact \(<001>\) direction for up to 10°. Such tilts have been found for diinterstitials /7/ and for mixed dumbbells /9/.

The diinterstitial consisting of two parallel tilted dumbbells is shown in fig. 3. Fig. 3a shows the two equivalent orientations, which these dumbbells may occupy. Transitions between these two may cause an IF peak with an activation

![Diagram](image.png)

Fig. 3: (a) "Tilt"-jump and (b) 90°-jumps of the diinterstitial atom consisting of two parallel tilted dumbbells.
energy of the order of 0.01 eV according to the computer simulation. This diinterstitial may undergo a second reorientation as shown in fig. 3b, which is expected to give rise to a second relaxation effect with an activation energy of the order of 0.1 eV. This latter jump causes simultaneously with the reorientation effect a long range diffusion of the diinterstitial. This is exactly what has been observed for the peak pair \( (R_2,M_2) \), where in \( R_2 \) only a local reorientation is involved, whereas in \( M_2 \) in addition to the reorientation a long range diffusion occurs \(^{14}\).

The only serious discrepancy in this assignment lies in the fact, that the experimentally observed activation energy of \( R_2 \), which is 0.09 eV is much higher than expected for a slightly tilted dumbbell.

Larger multiple interstitial atoms may also contain tilted dumbbells. Tilt-jumps and 90°-jumps as before of dumbbells in one of these multiple interstitial atoms may be invoked to account for the occurrence of the second peak doublet \( (R_0,R_4) \) observed in Al.

The <001>-mixed dumbbell is shown in fig. 4a, where an impurity atom and a host atom are members of the dumbbell. There are 6 equivalent positions of the mixed dumbbell around the octahedral centre, where the impurity-end points always to this centre. Reorientation jumps may occur by jumps of the impurity-atom from one site to another like in a cage.

Fig. 4a: The <001> mixed dumbbell with the "cage" in which the impurity may rattle around.

Fig. 4b: Tilted mixed dumbbell. The fourfold degeneracy of the corners of the octahedral cage is lifted. Two jumps at different rates \( v_1 \) and \( v_2 \) occur within the squares and from one square to the other, respectively.

Fig. 4c: Split trigonal cage. It develops from the previous one (Fig. 4b) by increasing the size of the squares, whereby the size of the triangles decreases.

If the dumbbell is tilted the fourfold degeneracy of the corners of the octahedral cage is lifted and a new cage with 24 equivalent configurations is formed as shown in fig. 4b. Two different jumps can occur in this cage: jumps within each of
the squares at a rate $v_1$, and jumps from one square to another at a rate $v_2$.

Although this "tilted" dumbbell model explains in principle the occurrence of a peak doublet because of the two different jumps, it turns out that the peak pair $(1,4)$ in Al-Fe cannot be explained by this model because of the following peculiar property of this pair: Both peaks are characterized by $\delta C \neq 0$ and $\delta C' \approx 0$, i.e. both peaks are induced by a pure C-shear deformation /5/. In order to account for the non vanishing relaxation strength of the high temperature peak (4) it is necessary to assume that the full thermodynamic equilibrium distribution of the Fe-atoms among the 24 positions can only be achieved by the $v_2$-jumps, and already not by the $v_1$ jumps at low temperatures. It turns out now that this requirement is just not fulfilled by the tilted dumbbell: The system can already relax into the final equilibrium via $v_1$-jumps solely /7/, and therefore a second peak under a C-deformation cannot be explained despite of the two different jump modes.

Several other cage structures have been examined. One possible candidate, which would explain the present observations is shown in fig. 4c. This cage can be envisaged as developing from the previous one by increasing the squares, whereby the size of triangles decreases. As the separation in the triangle becomes small, the low temperature peak would occur via jumps in the triangle, and the high temperature peak via jumps in the squares. This cage could in fact account for all the properties of the peak doublet $(1,4)$ in Al-Fe.

There is however a severe objection against this model coming from an entirely different side: The localized diffusion of the Fe-atoms in a cage has also been observed recently by Mössbauer measurements /11/. The results of these measurements however are not in agreement with the cage shown in fig. 4c, i.e. the cage which explains the IF-peaks well does not so for the Mössbauer measurements /12/. All cages compatible with the cubic symmetry of the fcc lattice containing up to 24 interstitial sites for the solute atom have been examined. There are two other cages which could equally well account for the present IF-results on Al-Fe like the previous one, but again both models fail to explain the Mössbauer results.

Conclusión.— Peak doublets which manifest themselves by their parallel annealing behaviour have been observed in irradiated Al and Al-Fe. Atomistic models involving tilted dumbbells and localized diffusion of the Fe-atoms in interstitial cages, respectively, may in principle explain these observations. In detail, however, it is not possible to present distinct models at the present time. For Al-Fe in particular a model accounting for the Mössbauer results simultaneously with the present IF-results is still lacking.
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