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Mechanical dynamical spectroscopy in low-flux neutron irradiated pure copper

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Abstract. The effects of low-flux neutron irradiation performed at room temperature on mechanical dynamical spectroscopy in pure copper (99.99%) are shown. A new damping peak at approximately 730 K appears in the irradiated polycrystalline samples, which is absent from the unirradiated and single-crystals one. This peak could be controlled by a cooperative mechanism between grain boundary sliding, the dragging of point defects which follow the dislocation line and the stress assisted break away of dislocations from the pinning points which were generated by the irradiation process.

1. Introduction

Mechanical dynamical spectroscopy (MDS), also called mechanical spectroscopy, is a technique very sensitive to the sample microstructural state. The characteristic temperature dependence of the damping or internal friction, F, arising from the thermally activated motion of defects, gives information on the parameters characterizing this motion. Besides this, the dependence of the elastic modulus on temperature can be determined simultaneously with the damping [1]. Furthermore, it is well known that amplitude dependent damping measurements are a powerful tool for studying the interaction between dislocations and point defects [2, 3].

The interaction between dislocations and point defects can be usually interpreted by means of two different physical models. (i) *Dislocation pinning*. The point defects produce a shortening of the average free length of dislocation segments [3]. As consequence of this model the damping must decrease for a larger number of defects. (ii) *The dragging process*. The pinning points follow the motion of dislocations. In this model the damping increases for a larger number of defects [4–7].

Besides this a detailed review of other radiation effects and their reactions during recovery has been reported in [8–13].

In this work MDS measurements, as a function of temperature and strain, were performed in high-purity copper (99.99%) irradiated at room temperature with low neutron flux, about 10^7 fast neutrons cm^{-2} s^{-1}.

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The damping spectrum in irradiated polycrystalline samples shows a new damping peak at approximately 730 K, absent from the unirradiated ones. This new peak does not disappear completely after the first heating, during the MDS test, until 1000 K. A possible physical mechanism which controls the new damping peak generated by the low-flux neutron irradiation in polycrystalline copper is proposed.

2. Experimental procedure

2.1. Irradiation

The irradiation was carried out in a nuclear reactor, Siemens SUR 100, which was operated at 0.7 W, at room temperature.

The samples were positioned in the horizontal channel, which passes through the reactor core, in two conditions. These are inside poly-methyl-methacrylate and without any cover. In fact, the samples which were irradiated under the poly-methyl-methacrylate cover received predominantly thermal neutrons, while the ones which were irradiated without any cover received the whole reactor spectrum. The thermal- and fast-neutron fluxes were about 5.2×10^7 neutrons cm^{-2} s^{-1}. Their energies were approximately 0.0025 eV and 10 MeV respectively. The irradiation was performed during different periods of time, which are shown in table 1.

Table 1. Sample status.

Sample denomination	Annealing at 1073 K, under high vacuum (h)	Cold work (area reduction) at room temperature (%)	Irradiation at room temperature
Single crystal [111]			
1	1	none	none
1′	1	none	under cover, 5 h
Polycrystal			
2	1	none	none
2′	1	none	under cover, 5 h
2″	1	none	without cover, 5 h
2‴	1	none	under cover, 10 h
3	5	none	none
3′	5	none	under cover, 5 h
3″	5	none	without cover, 5 h
4	15	none	none
4′	15	none	under cover, 5 h
4″	15	none	without cover, 5 h
5	1	1	none
5′	1	1	under cover, 5 h
6	1	2	none
6′	1	2	under cover, 5 h

2.2. Samples

The samples were made with high-purity electrolytical copper (99.99%), in sheets for the polycrystalline ones and in cylindrical form for a single crystal with [111] orientation. The final size of each kind was 3.5 mm wide, 0.3 mm thick and 30 mm long and 1 mm radius and 20 mm long, respectively. They were thermally treated at 1073 K under high vacuum during different periods of time, prior to the cold work and neutron irradiation as shown in table 1. The cold work of the samples also was performed prior to the neutron irradiation. The status of checked samples is summarized in table 1.

After neutron irradiation the samples were stored at room temperature until sufficient radioactive decay had occurred to permit the usual handling. Besides this, all the samples, prior to the MDS test, were cleaned by etching in HNO_3.

2.3. MDS measurements

The MDS measurements were performed in a torsion pendulum of inverted type, with variable moment of inertia, which operates at frequencies between 0.1 and 40 Hz under high vacuum [14, 15]. The natural oscillation frequency at room temperature, for all the checked samples, was chosen about 0.7 Hz. The maximum shear strain on the sample, ε_0, was less than 5×10^{-5}. The heatings were performed up to 1000 K with a rate of increase of 0.5 K min^{-1}. The decrease of temperature was also performed at the same rate.

On increasing the heating rate toward higher values, in both the unirradiated and irradiated samples, the damping curves were rigidly shifted without changing their shapes. The damping values were measured in free decay with an error of less than 1%. These values for the irradiated polycrystalline samples in the temperature range between 570 and 810 K (the temperature range where the new peak appears) were amplitude dependent and they were converted to intrinsic damping, because this degree of dependence cannot be neglected [16]. However, outside this temperature range for the irradiated polycrystalline samples, and over the whole temperature range in the single crystals (samples 1 and 1′) and in the unirradiated samples, the damping was considered amplitude independent. In fact the corrections made to the measured damping to obtaining the intrinsic values turn out to be not greater than 5×10^{-5} [16–18]. The amplitude dependent damping effects were checked by measuring the freely decaying torsional amplitudes, A_n ($n = 1, 2, \ldots, N$), at constant temperature, T, by means of a data acquisition system [14]. As a first step, polynomials were fitted to the data $\ln(A_n)$ against n representing the decaying oscillations. The damping as a function of amplitude was determined by the first derivative of the polynomial $\ln(A_n)$ against n [19]. The results were depicted as F against strain, ε. Best fits were obtained with polynomials of degree three or less. The amplitude dependent damping degree was determined through the slope of the $F(\varepsilon)$ curves, $S = \mathrm{d}F/\mathrm{d}\varepsilon$. This may be replaced in a restricted range of ε by the mean value $s = \Delta F/\Delta\varepsilon$ [19]. In the amplitude independent damping range, typical s values were about zero. Meanwhile, in the amplitude dependent range typical s values were about 100.

The intrinsic damping at ε_0, $F_i(\varepsilon_0)$, was calculated by means of the following expression in the case of the single crystal which has the shape of a circular bar [20]:

$$F_i(\varepsilon_0) = F(\varepsilon_0) + (\varepsilon_0/4)\,\mathrm{d}F(\varepsilon_0)/\mathrm{d}\varepsilon_0. \qquad (1)$$

Meanwhile, in the polycrystals, which were sheets, the intrinsic damping was calculated by means of a method of deconvoluting the intrinsic damping from the measured damping [17, 18]. In fact, the intrinsic damping can be expressed as an expansion of Chebyshev polynomials, valid in the interval of deformation $[0, \varepsilon_0]$ as

$$F_i(\varepsilon_0) = \sum_{n=0}^{N} C_n T_n(\varepsilon') \qquad (2)$$

with $\varepsilon' = 2(\varepsilon/\varepsilon_0) - 1$, which reduces the expansion to the interval $[-1, 1]$. Moreover the measured damping can be written as

$$F(\varepsilon_0) = \sum_{n=0}^{N} C_n I_n(\varepsilon_0) \tag{3}$$

where

$$I_n(\varepsilon_0) = \left(\int_V T_n(\varepsilon')\varepsilon^2 \, \mathrm{d}V \right) \bigg/ \left(\int_V \varepsilon^2 \, \mathrm{d}V \right) \tag{4}$$

with $\varepsilon = (\varepsilon_{xx}^2 + \varepsilon_{xy}^2)^{1/2}$, ε_{xx} and ε_{xy} being the non-zero shear strain components of the strain tensor of the sheet (assuming the x axis across the sheet, the y axis along the width and the z axis the torsion axis of the sample) [18]. Following the work of [18], using the measured F values as a function of strain and calculating the corresponding coefficients $I_n(\varepsilon_0)$, equation (3) transforms into a linear equation system where the unknown C_n coefficients can be obtained. Inserting them in the expression (2) gives the intrinsic damping as a function of strain in the interval $[0, \varepsilon_0]$. The number of experimental points to be used, and consequently the order N of the expansion, was chosen so as to give a good representation of the measured damping.

3. Results

Figure 1 shows the intrinsic damping spectra measured during increase of temperature at $\varepsilon_0 = 4.5 \times 10^{-5}$, for the sets of samples 1 and 2 of table 1. The triangles, plotted with respect to the right-hand axis, correspond to the damping values measured in the unirradiated single crystal, sample 1. The dashed line which also has been plotted with respect to the right-hand axis corresponds to the damping values of the irradiated single crystal, sample $1'$. On the left-hand axis are indicated the damping values which correspond to the set of samples number 2, i.e. samples 2, $2'$, $2''$ and $2'''$. These have been plotted by means of full, short-dashed, long-dashed and short- and long-dashed lines respectively.

The dotted line in the figure represents the damping spectrum measured during heating for a sample of type x', i.e. irradiated under cover for 5 h, but measured at a lower shear strain, $\varepsilon_0 = 2 \times 10^{-5}$.

The damping spectrum which has been measured for the unirradiated polycrystalline samples is in agreement with the results reported in the literature for copper [21–24]. The low- and intermediate-temperature grain boundary damping peaks called LTP and ITP, which appear in the annealed copper at 550 and 720 K respectively (at 1 Hz), have been measured.

It should be pointed out that the low-flux neutron irradiation performed at room temperature produces a new damping peak in polycrystalline samples, at about 730 K, absent from the unirradiated ones and in the single crystals. The new damping peak is higher in the sample irradiated with the whole reactor spectrum than in the sample irradiated predominantly with thermal neutrons. The peak also increases with longer irradiation time. Furthermore a strong dependence of damping on the strain can be also observed from figure 1.

The intrinsic damping values which correspond to the sets of samples 3 and 4 have been plotted in figures 2 and 3 respectively. The x, x' and x'' kinds of sample, in each figure, have been plotted by means of full, short-dashed and long-dashed lines respectively. The dotted lines also represent the damping values for the x' kind of sample measured during heating at $\varepsilon_0 = 2 \times 10^{-5}$. As can be seen from the figure for the sets of samples 3 and 4 the LTP and ITP appear overlapped [21].

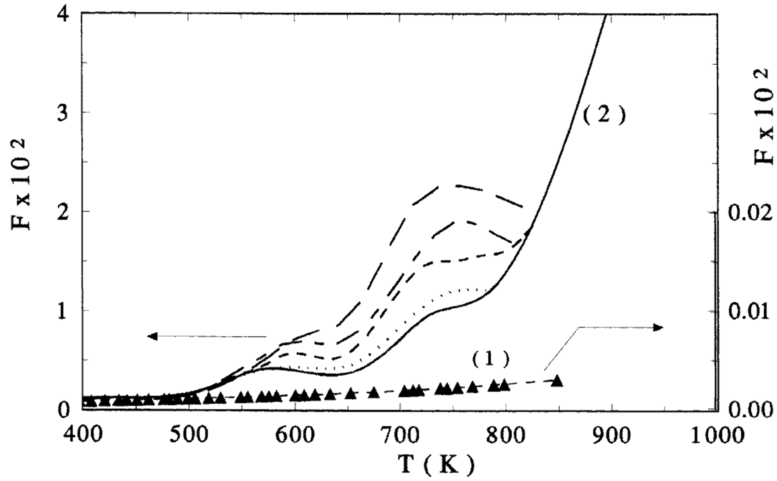

Figure 1. Right axis, MDS spectra measured at $\varepsilon_0 = 4.5 \times 10^{-5}$ for the single crystal [111], unirradiated (triangles) and irradiated under cover for 5 h (short-dashed line). Left axis, MDS spectra measured at $\varepsilon_0 = 4.5 \times 10^{-5}$ for the set number 2 of samples (full line, 2; short dashed, 2′; long dashed, 2″; long and short dashed, 2‴). The dotted line represents the MDS spectrum for the sample irradiated under cover for 5 h, but measured at $\varepsilon_0 = 2 \times 10^{-5}$.

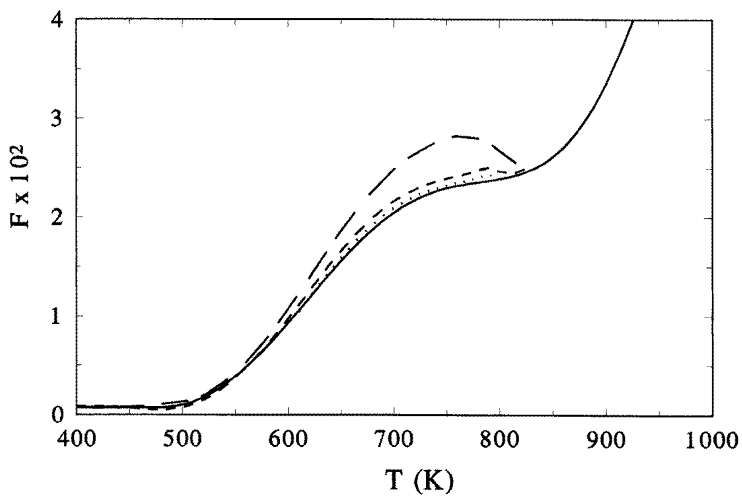

Figure 2. MDS spectra measured at $\varepsilon_0 = 4.5 \times 10^{-5}$ for the set number 3 of samples (full line, 3; short dashed, 3′; long dashed, 3″). The dotted line represents the MDS spectrum for the irradiated sample under cover, but measured at $\varepsilon_0 = 2 \times 10^{-5}$.

The increase in the damping values produced by the irradiation, related to the appearance of the new damping peak, can be also observed in figures 2 and 3. However, as can be seen from the figures, the appearance of the new peak in the total spectrum is very difficult to determine. Therefore, the increase in the damping spectrum could be also related to a change in shape of the ITP and not to a new damping peak.

The irradiation effects on the damping were smaller in the samples with a longer

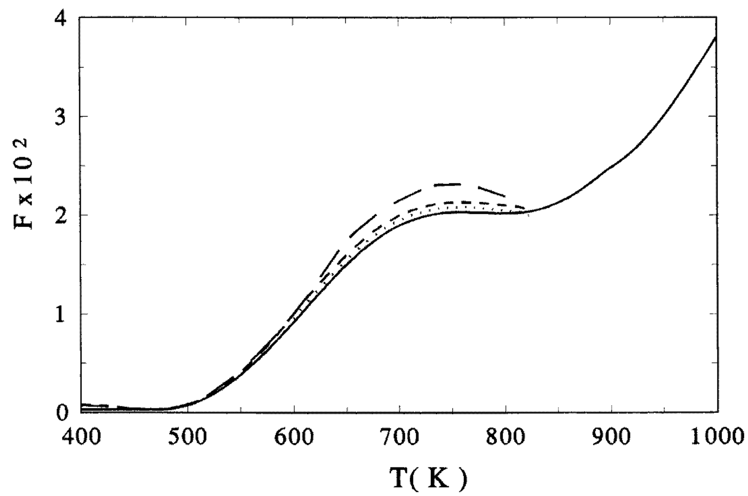

Figure 3. MDS spectra measured at $\varepsilon_0 = 4.5 \times 10^{-5}$ for the set number 4 of samples (full line, 4; short dashed, 4′; long dashed, 4″). The dotted line represents the MDS spectrum for the irradiated sample under cover, but measured at $\varepsilon_0 = 2 \times 10^{-5}$.

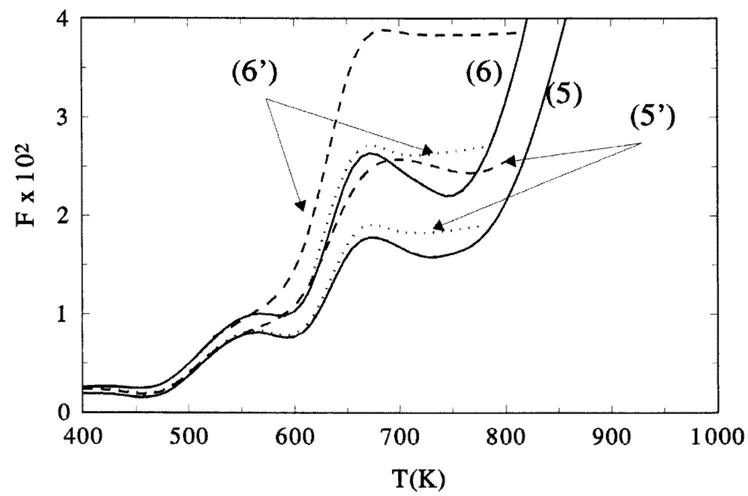

Figure 4. MDS spectra measured at $\varepsilon_0 = 4.5 \times 10^{-5}$ for the cold-worked samples 5 and 6, unirradiated (full line) and irradiated (short-dashed line). Dotted lines represent the MDS spectra for the irradiated samples, but measured at $\varepsilon_0 = 2 \times 10^{-5}$.

annealing time. A strong decreasing in the damping when it is measured at a lower strain also was found.

Figure 4 shows the intrinsic damping values for the samples 5 and 5′, and 6 and 6′. The samples 5 and 6 have been plotted by means of full lines, while 5′ and 6′ have been plotted by means of short-dashed lines. The dotted lines in the figure have the same significance as above. As can be seen from figure 4, in samples 5 and 6, the damping peak related to cold work, which appears approximately at 650 K, was measured [24]. Furthermore, it can be established that the effects of the irradiation on damping are longer in the deformed samples than in the undeformed ones.

It should be pointed out that the new peak which is generated by the low-flux neutron irradiation appears in polycrystalline samples and is absent from single-crystal ones. Furthermore, the peak height increases for the following four conditions: (i) irradiation with the whole reactor spectrum; (ii) longer irradiation times; (iii) higher degree of cold work and (iv) shorter annealing. Besides this, the damping peak values are made strongly amplitude dependent, as can be observed by the reduction in the peak height when the amplitude is decreased to 2×10^{-5}. Moreover, neither could the new damping peak be found in samples irradiated in either condition (with and without cover) when the damping was measured at a lower $\varepsilon_0 = 1 \times 10^{-5}$. As already pointed out, the amplitude dependent damping behaviour only appears within the temperature range where the new peak appears. Outside this temperature range, i.e. in the tails of lower and higher temperatures, the damping is amplitude independent.

On the other hand, the damping spectrum values and the amplitude dependent damping degree (s), within the temperature range of the new peak, decreased during cooling after having reached 1000 K in the MDS test. Their reduction as a percentage, at 730 K for the irradiated sets 2–4 of samples is given in table 2. Neither remnant effect in the F and s values could be measured during the run down in temperature for the samples measured at $\varepsilon_0 = 2 \times 10^{-5}$. The samples 5′ and 6′ are not included in the table due to the fact that the results are not representative of the recuperation degree related to the irradiation effects, because the unirradiated ones (samples 5 and 6) already show a hysteresis behaviour in the damping values related to recuperation of the structure.

Table 2. Reduction as a percentage of the damping value, F, and the amplitude dependent damping degree, s, between the run up and down in temperature, evaluated at 730 K.

Sample	Reduction in F	Reduction in s
2′	15	17
2″	30	31
2‴	14	15
3′	3	5
3″	8	11
4′	2	2
4″	6	7

In contrast, changes in the low- and high-temperature damping background between irradiated and unirradiated samples of table 1 during heating or cooling cannot be detected.

4. Discussion

The non-appearance of a damping peak in the irradiated single crystal (figure 1) suggests that this new peak in the irradiated polycrystalline samples could be connected with the grain boundary structure. Besides this, the dependence on the degree of cold work and the annealing state connects this phenomenon with the dislocation density. The increase of the peak height in the samples irradiated for longer times and in samples without a cover, i.e. the increase of the peak height for a larger number of defects, could be related to a dragging process, of peaking effect type [4–7]. The peaking effect has been reported for copper. In fact, early results for copper reported that the peaking effect appears as an amplitude independent damping phenomenon in room-temperature irradiated samples previously annealed at temperatures lower than 1023 K [10]. However, subsequent works have shown that amplitude dependent damping effects appear in the peaking effect for samples irradiated around 160 K previously annealed at 1173 K [6, 7].

The new damping peak also could have a contribution from the stress assisted break away of dislocations from pinning points [2, 3] which were generated by the irradiation. The strong dependence on the oscillating strain and the relatively high critical values with which the new peak disappears, $\varepsilon_0 = 1 \times 10^{-5}$, could relate this peak to the stress assisted break away of dislocations from the weak pinning points. In fact, it has been reported for copper that the lowest strain at which the peaking effect appears is about 10^{-6} [6, 7]. Therefore $\varepsilon_0 = 2 \times 10^{-5}$ could be the lowest strain which produces the minimum stress needed for the break away of dislocations. The stress assisted break away is the starting mechanism and subsequently the dragging process develops, both interacting with the grain boundary structure.

The dislocation motion prior to yielding in irradiated copper and its interaction with agglomerates has been verified for fluxes lower than 10^{13} fast neutrons cm^{-2} s^{-1} (fn cm^{-2} s^{-1}) [8]. In contrast, for bigger doses, about 10^{16} fn cm^{-2} s^{-1}, no dislocation motion was found for the stress application until the yield stress. Therefore the motion of dislocations at lower stresses in copper irradiated at 10^{7} fn cm^{-2} s^{-1} could be reasonable. Furthermore, the existence of agglomerates, which could interact with the dislocation motion, until temperatures of about 773 K has been verified employing x-ray diffraction techniques [9, 26].

On the other hand, the amplitude dependent effects which were found within the temperature range of the peak were absent from the lower and higher tails of the damping spectrum. Therefore this discounts the possibility that the non-linear effects are related to the damping background. Consequently the non-linear effects are related to the peak mechanism. Furthermore, the agreement between the low- (before peak) and high- (after peak) temperature damping background values for the unirradiated and irradiated samples permits us to relate the increase in the total damping spectrum to an increase of the new relaxation peak.

It should be pointed out that the new peak owing to the irradiation process could be really a new peak overlapped on the ITP or a change in the ITP produced by its interaction with the dislocation mechanism mentioned above. In fact, the problem of deconvoluting accurately the damping spectrum is not an easy task because the decomposition is not unique [25] and this problem will be discussed elsewhere.

Resuming, the physical mechanism which controls this new damping peak in irradiated polycrystalline copper could be a cooperative effect between the grain boundary structure, the dragging of pinning points which follow the dislocation line motion and the stress assisted break away of dislocations from pinning points which were generated by the irradiation processes.

In another light, the decrease in the peak height and the *s* degree in the temperature range of the new peak during cooling after having reached 1000 K in the MDS test could be related to the partial dissolution of agglomerates and also the successful structure recovery, which is in agreement with the above-proposed physical mechanism. However, attention should be paid to the fact that a remnant peak persists on decreasing temperature during the MDS test, after having reached 1000 K. In fact, the time involved for measuring the peak during heating and again during cooling is about 18 h, i.e. the sample is annealed for 18 h at temperatures higher than 730 K, which is near to the temperature for removing the radiation damage.

5. Conclusion

A new damping peak at approximately 730 K in low-flux neutron irradiated polycrystalline copper was found. This peak could be controlled by a cooperative mechanism between grain boundary structure, the dragging of point defects during dislocation motion and the stress assisted break away of dislocations from the pinning points which were generated by the neutron irradiation. Moreover the damping peak only appears for a maximum shear strain on the sample larger than 2×10^{-5}.

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