Transmission Electron Microscopy and Mobility of Interfaces, Hysteresis and Internal Friction

R. Gotthardt, J. Stoiber, Institut de Génie Atomique, Ecole Polytechnique Fédérale de Lausanne, CH-1015 Lausanne

Introduction

Martensitic transformations in non-ferrous alloys are generally accompanied by hysteresis effects. Hysteresis occurs when, during the martensitic transformation, energy is dissipated. It indicates that energy which is needed to advance the forward transformation cannot be used for the reversed one. The reason for dissipating energy can be many-fold. As examples can be mentioned: nucleation processes, interaction of migrating interfaces with the microstructure of the matrix (point-defects, precipitates etc.) and diffusion of point defects to interfaces after transformation. A thermodynamic description of how all these parameters influence the shape of the hysteresis curve is given by Delaey [1] while Hornborgen [2] discusses the effect of the transformation temperatures. Clearly, hysteresis effects allow the transformation behaviour to be modified through the control of microstructure by suitable thermo-mechanical treatments. This is particularly important in cases where additional elements are added to alloys to get finer grains for better processing [3].

Considerable effort has been made to understand the different observed hysteresis effects. Often, however, the given models can only explain the single system being studied and until now, no unique extensive description exists which is generally accepted. So the aim of this paper cannot be to review all the data on hysteresis effects. An overview of the studies on this topic is shown in recent conference proceedings [4],[5]. In this paper further information is given which comes out of transmission electron microscopy observations of in-situ transformation and from new results of internal friction measurements. They could contribute to reveal and understand mechanisms controlling hysteresis effects.

Experimental Techniques

Transmission Electron Microscopy (TEM)

TEM-observations have the great advantage that micro-mechanisms supposed to be responsable for macroscopic properties can be studied directly. On the other hand, due to the often very high magnification, the choice of the part of the sample to be studied is very important in order to avoid the observation of artifacts. In addition, the samples have to be thin (~0.1 to 0.3 µm) in order to be transparent to electrons. This fact can change the microstructure of the observed part of the specimen and must be taken into account in the interpretation of results.

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The conventional TEM-technique (CTEM) is often used to determine crystallographic structure of the phases involved and to reveal crystallographic orientations and relationships of interfaces. In addition it allows the determination of Burgers vectors of dislocations and displacement vectors of stacking faults. (For more details see TEM handbooks [6]).

High resolution electron microscopy (HREM) allows even distances of interatomic scales to be revealed (columns of atoms) which has extended our knowledge of interface structures considerably. For such studies, extremely thin samples are needed, so the near surfaces (image forces) influence even more the microstructures. Thus, internal stresses, responsable for many properties observed macroscopically, are decreased or even completely relaxed.

Both techniques are based on static observations. With the help of goniometer specimen stages precise crystallographic relationships can be determined, but it is very difficult to relate these observations to the transformation process itself.

With special specimen stages it is possible to change the temperature of the sample, to apply a stress or even both simultaneously. Generally such specimen stages allow only a tilting about one axis, which is not enough for exact crystallographic studies. The major advantage is, however, that the transformation can be observed directly, which is of particular importance for the study of the thermoelastic martensitic transformation. By this method, the migration of an interface between a martensitic plate and the matrix, or, between different martensite plates can be followed during temperature, stress or combined temperature/stress variations. There are two modes for stress variations. Firstly, stress can be applied by a pneumatic system which guarantees a constant load without relaxation [7]. Secondly, the specimen may be deformed effectively to a certain amount with a given strain value which is then followed by relaxation. The appropriate specimen holder has to be chosen for a particular problem.

Internal Friction Measurements

By this method microscopic mechanisms can be dectected which consume energy during a periodic movement. For instance, if a specimen is exited to oscillation and one observes the decay of the free oscillation, the amplitudes decrease slowly in the case of low Internal Friction (I.F. or Q^{-1}) and will be attenuated rapidly when the I.F. is high. So, materials with high I.F. have high damping capacity. When I.F. is measured as a function of frequency at constant temperature, then a maximum is observed at the eigenfrequency characteristic of the respective mechanism. This measurement mode is not always applicable, because the oscillation frequency of the measurement system can often not be varied over a big enough frequency domain. Thus, many installations are working by varying the temperature at constant frequency. Thermal activated processes are then observed by maxima at characteristic temperatures, which depend obviously on the frequency used. The temperatures related to a martensitic transformation should therefore not be changed when varying the measuring frequency, whereas the amount of Q^{-1} can do so. More details about I.F. related to these transformations are given elsewhere [8],[9],[10]. Because of the fact, that I.F. measurements reveal mechanisms consuming energy, this method is particularly interesting for investigations of hysteresis effects related to the martensitic transformation. As mentioned before, details of the shape of the hysteresis are related to the interaction of the interface with the microstructure of the matrix. Even if there is no general agreement about the structure of the interface, it can be regarded as an ensemble of linear defects (misfit or coherence dislocations [11]) which can interact with point defects, impurities, precipitates, grown-in or transformation induced dislocations, other interfaces, etc. Each of these mechanisms influences the internal friction in a characteristic way. In addition I.F. depends on the density of mobile dislocations inside a martensitic plate and is indirectly sensitive to the distribution of internal stresses.

In order to get the maximum of information from the same sample before, during and after the transformation, a special installation for the measurement of I.F. has been constructed. It is an inverted torsion pendulum, operating at about 1 Hz. In addition to the I.F. it allows the simultaneous measurements of the frequency, the electrical resistivity and the rotational component of the shape change. The frequency gives useful information on the shear modulus. The electrical resistance shows the amount of transformed material. For quantitative interpretation it has to be considered that the sample can change its length and its section [12], which change the resistance, without changing the specific resistivity. The measured shape change can only be used qualitatively but it gives additional information on the characteristic transition temperatures. For this investigation the pendulum was modified in such a way, that a tensile stress could be applied during the oscillation of the sample, which allows the study of transformations under stress, important for most applications.

TEM Observations

The presented results concern mainly observations of in-situ transformation induced by temperature and/or stress variations with special emphasis on migrating defects which are able to consume energy. Crystallographic relationships are not given here, but are similar to those observed under static conditions by other authors [13],[14].

Interfaces austenite-martensite

Two types of interface migration have to be considered.

a) the movement of the tip of a plate
b) the thickening process.

The appearance and growth of a martensitic plate has been observed and analysed quantitatively in a Au-Cu-Zn alloy (Ms = 180 K). The plate appears suddenly and has immediately a certain thickness, showing a very high velocity parallel to the habit plane. In this situation the plate remains a certain time before jumping to a greater thickness. When the tip can be followed this process is seen by a stop-and-go movement. After a certain time, the thickening process becomes smooth (Fig. 1). The same behaviour is observed in Cu-Zn-Al martensitic alloys. An advancing tip of a martensitic plate induces an elastic stress field in the surrounding matrix, which can be seen by TEM in thin foil. Due to stress relaxation, the background intensity changes around the tip [15]. Such tips retract immediately when stress or temperature rates are inversed [16]. Thermal cycling seems to increase the number of favorably oriented martensitic plates in contrast to stress cycling [17].

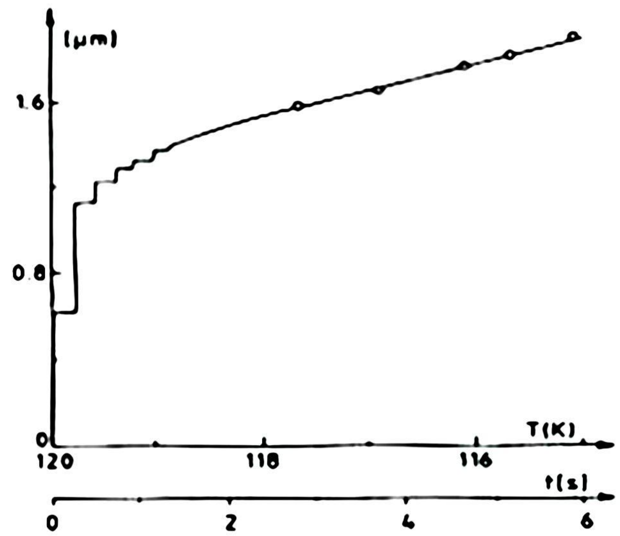


Fig. 1: Growth of a martensite plate (thickening) in a Au-Cu-Zn alloy; taken from TEM-video recording of an in-situ transformation.

Dislocation tangles, whether they are introduced during solidification or by thermal cycling had no stopping effect on a moving interface.

Careful observations have been made in order to detect the starting point of the transformation. The dislocation tangles mentioned earlier are seen not to be the origin of the transformation, at least not in thin parts even if the thermal cycling is carried out in an already thinned sample.

On the contrary, nucleation has been observed in parts where stress is acting, like at borders of holes or other stress concentrations seen by bend contours. This stress can, when favorably oriented, nucleate the new phase. This happened often, being not directly at the border but somewhere inside the sample.

Mostly, the martensitic plates are growing from thick parts into the electron transparent ones.

Interfaces martensite-martensite

As can be seen later from I.F. results, martensite-martensite interfaces seem to play an important role for the appearance of a hysteresis effect. So these interfaces have been studied too.

In-situ transformation has been carried out in a Jeol 200 CX microscope operating at 200 kV in a Cu-Zn-Al alloy (nominal composition 19.26 at.% Zn and 14.37 at.% Al). This composition was chosen in order to have a low M_s (≈160K) so the transformation can take place without being perturbated by thermal diffusion of vacancies. The martensite was induced thermally as well as by deformation. In both cases the interface migrations were accompanied by a change in the stacking fault arrangement. This can be detected by a change of the contrast of existing stacking faults and by moving limiting partial dislocations. Such an activity has already been observed in preliminary experiments [15].
In order to explain the observations, a part of the specimen is presented schematically in Fig. 2. "I" indicates the position of the interface. Above and below two variants are shown together with their stacking faults. Stacking faults able to move are labelled "P". When such a P dislocation arrives at the interface, its position is changed locally. Fig. 3 shows a 6 sec. section of the video film recorded during heating of a martensitic part of the specimen. The film was stopped at the shown moments, the images recorded by a videoprinter and then photographed in the shown arrangement. An intensive stacking fault migration is observed without a long distance interface migration. Advancing stacking faults can be followed by their characteristic superimposed contrasts, simplified graphics are added to find those contrasts.

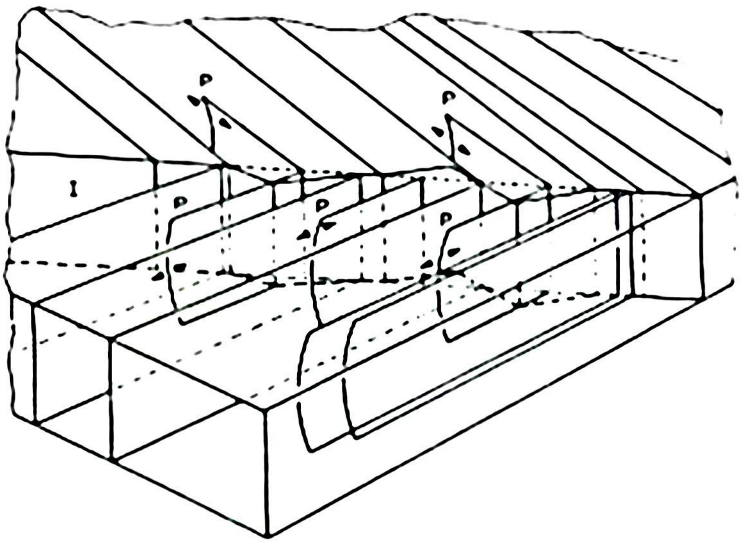
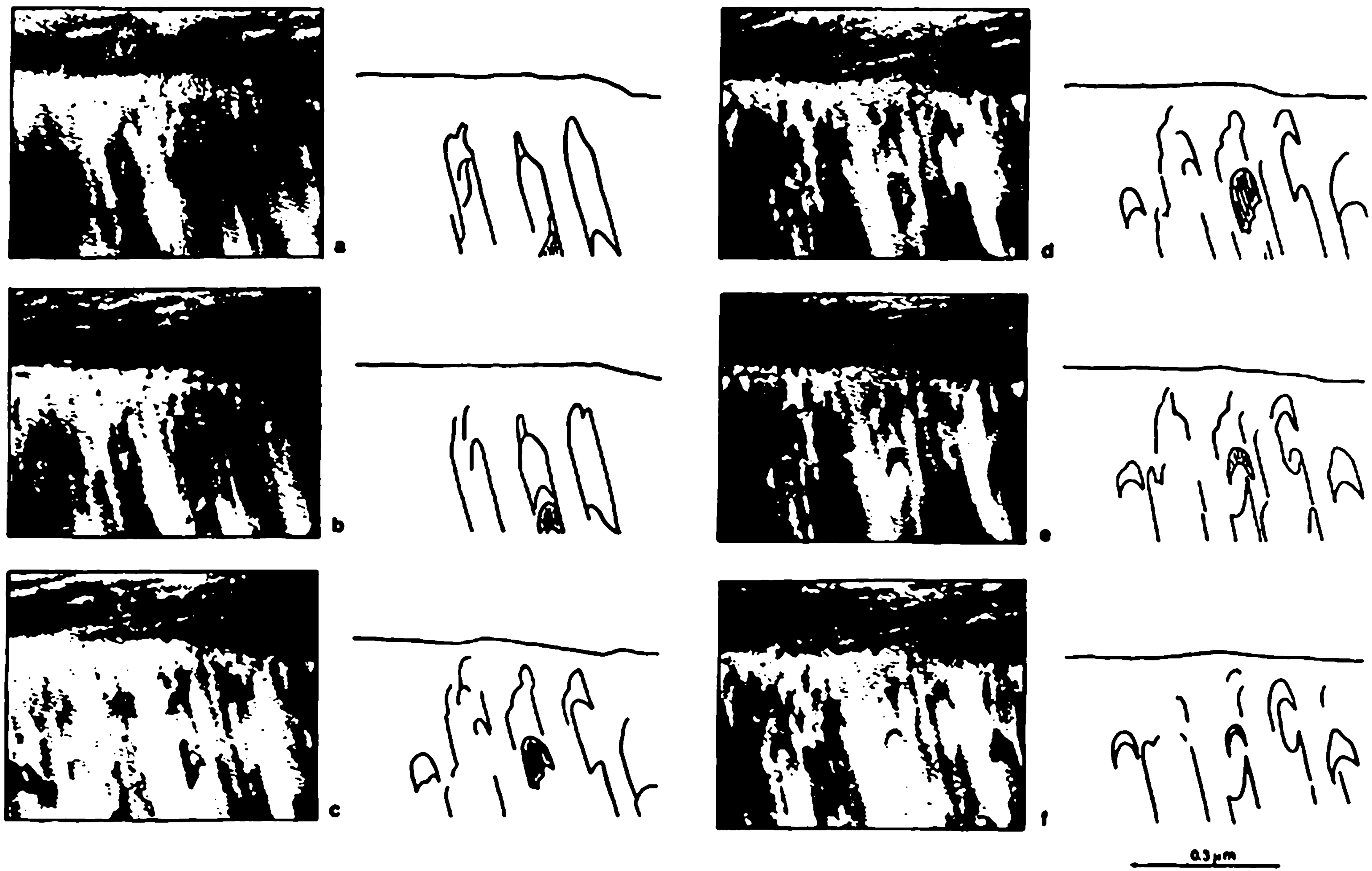


Fig.2: Schematic representation of a part of a TEM sample. I: plane of interface, P: moving partial dislocation limiting growing or shrinking stacking faults



<u>Fig.3:</u> TEM micrographs taken from a video recording during heating of a martensite plate of a Cu-Zn-Al alloy near As temperature. Two variants are shown, separated by a horizontal interface. Growing or shrinking stacking faults can be seen by the change of contrast.

Internal Friction

All results are measured in alloys of nominal composition of Cu-26% Zn - 14% Al with a Ms of about 290 K measured by calorimetry.

Fig. 4 shows the evolution of Q^{-1} as a function of temperature. The most important result is that the I.F. at the beginning of the transformation does not depend on the measurement amplitude ε, where in the middle of the transformation domain Q^{-1} is much smaller for higher ε than for small ones. Inside the martensite phase Q^{-1} increases with increasing ε, as usual. The transformation temperatures seen by the resistance measurements are not changed by varying the amplitude.

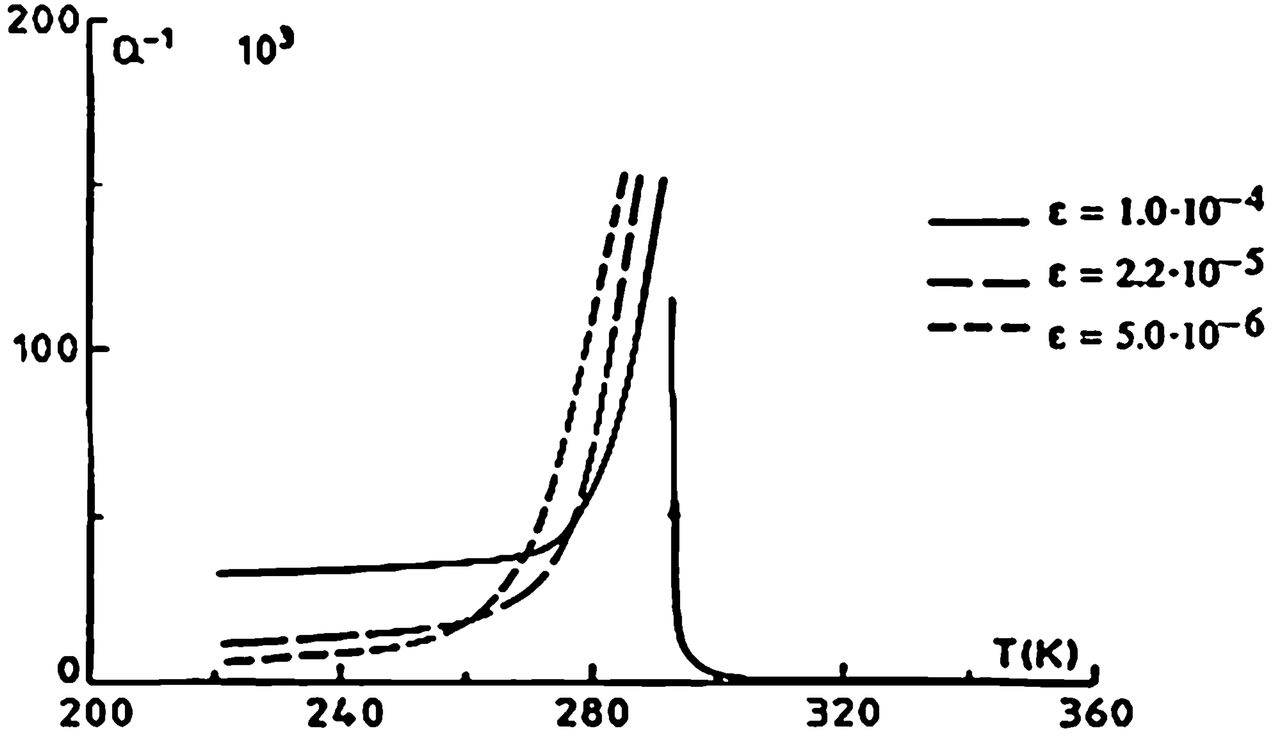


Fig.4: Internal friction (Q^{-1}) evolution as a function of temperature, measured with different amplitudes in a Cu-Zn-Al-alloy (Ms = 290 K)

If the temperature change is stopped every 5°, the Q^{-1} values drop drastically to lower values. Fig. 5 shows clearly for the case of cooling that the biggest contribution to Q^{-1} stems from transitory effects, the term depending on the temperature change rate $\dot{T} = dT/dt$.

The Q^{-1} evolution is shown for an amplitude $\varepsilon = 2.2 \times 10^{-5}$. Changing ε, one finds that the $\dot{T}$-contribution during transformation decreases with increasing ε values. This is shown in more detail elsewhere [17].

In order to study hysteresis effects at small scale, temperature cycles of 10 K width have been performed at different temperature

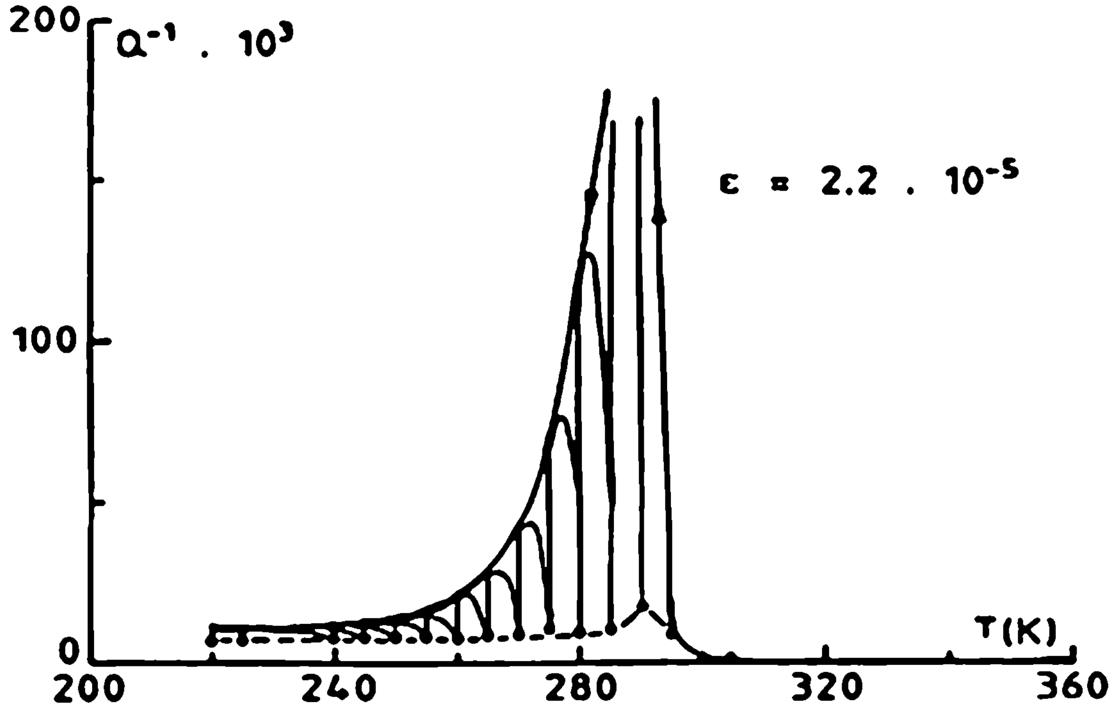
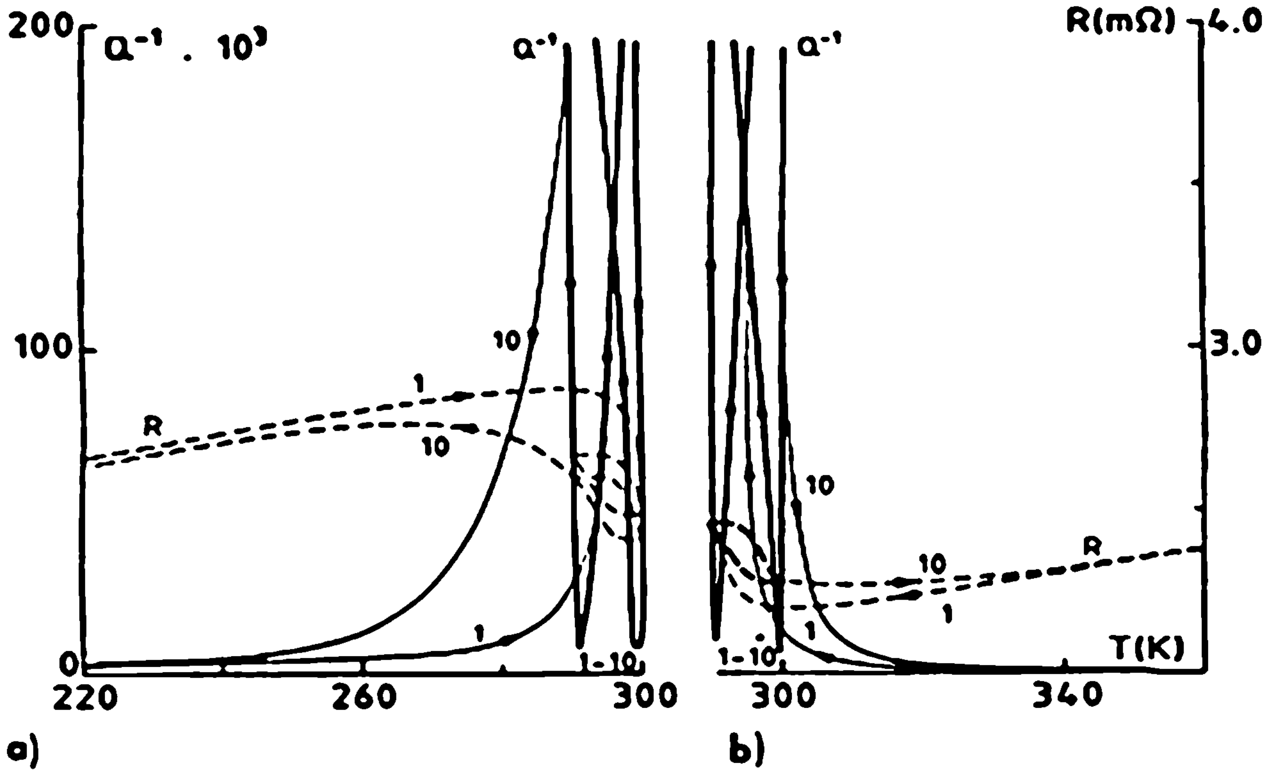


Fig.5: Q^{-1} evolution as a function of temperature during cooling. The decreasing temperature has been held constant every 5 K.

intervals. The results are shown in Fig. 6. Fig. 6a and 6b present the results of cycling in the same interval but starting from martensite or from austenite respectively, whereas in Fig. 6c cycling is performed around As. For the discussion of differences in resistance-temperature cycles measured in the same temperature region (290 K - 300 K) the corresponding resistance curves which are at different resistance levels are superimposed in Fig. 7.



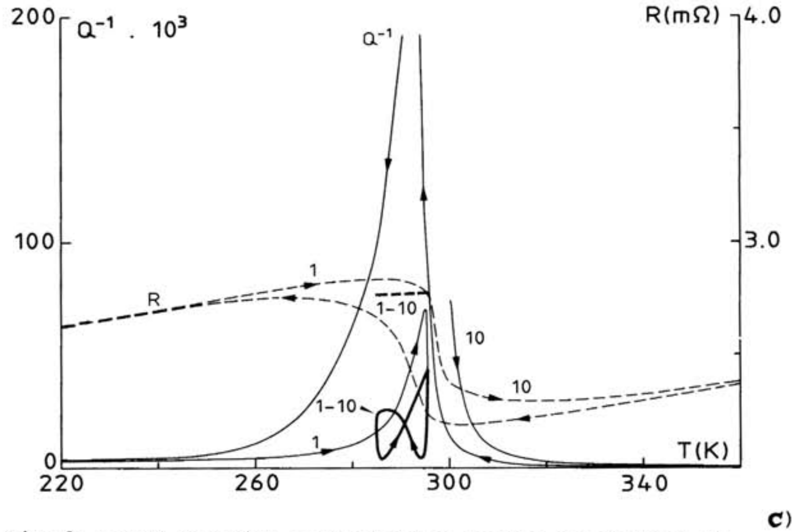


Fig.6: Small partial temperature cycles performed at different temperature intervals, cycles between 290 K and 300 K (a) starting from martensite (b) same interval starting from austenite (c) cycling around As starting from martensite.

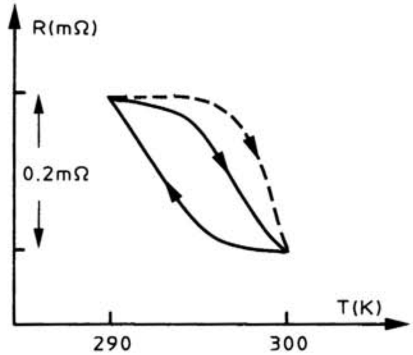


Fig.7: Electrical resistance as a function of temperature during partial cycling, taken from Fig. 6 a and b.

Discussion

The discussion will be concentrated on the interface migration. In the domain of the entire hysteresis, which is in general the transformation region, the high I.F. values are normally believed to come from migrating martensite-austenite interfaces [18].

Partial temperature cycles made at different parts of the hysteresis together with TEM observations of in-situ transformation give interesting information on migrating defects.

The small cycles around As-temperature (Fig. 6c) show no evolution of the resistance curves, they remain straight and do not decrease with the number of cycles. The Q^{-1} values, however, change in such a way that a characteristic "butterfly" shaped curve results. Due to the fact that resistance curves indicate changes in the transformed volume, this result reveals other than transformation mechanisms giving rise to the quite high I.F. variations. At each end of the cycle the sign of $\dot{T}$ is changed in the pendulum and $\dot{T}$ passes with a small phase shift through zero at the sample resulting in smallest Q^{-1} values, as also seen in the case of the entire Q^{-1} curve presented in Fig. 5.

The cycles made at higher temperatures, Fig. 6a and Fig. 6b should reveal effects of transformation because the resistance values change significantly. In both cases the cycling has been performed in the same temperature interval, so thermally activited processes, when present, should contribute in the same way. The total amount of martensite in the case of Fig. 6a is bigger than in the case shown in Fig. 6b. The changes of the Q^{-1} values are very strong, they are as big as they are observed during complete cycles. Nevertheless, as the resistance curves indicate, only when starting from austenite (Fig. 6b) material is transformed during the whole interval, during cooling as well as during heating.

On the contrary, when starting from the martensitic state, material is transformed only in the second half of the heating period (see also Fig. 7, dotted line). So again there should be another mechanism which induces high I.F.

The TEM observation of a martensitic plate during heating reveals a very active stacking fault migration (Fig. 3). Stacking faults arriving at the interface produce local changes in the internal stress field which alters the position of the interface, often accompanied by the emission of stacking faults in the adjacent neighbour variant. During the whole sequence shown in Fig. 3, the interface has not moved remarkably. Such an activity creates a high number of migrating defects and a highly locally mobile martensite-martensite interface, giving rise to high I.F. without changing the transformed volume. So these mechanisms are probably responsible for the Q^{-1} evaluation observed in Fig. 6c and in the first half of the heating interval of Fig. 6a.

The observations of migrating stacking faults during in-situ transformation are in contradiction to displacement vector determination under static observation conditions made by CTEM studies [19] or HREM studies [20],[21]. They conclude that no shear type stacking faults are present in Cu-Zn-Al (18R type) martensite, although moving stacking faults are observed frequently when interfaces move [15].

This discrepancy can be explained by the different conditions during the respective observations. In the static cases, especially in HREM studies, internal stresses which appear necessarily during the transformation are relaxed. The shear type stacking faults which are evidently possible during in-situ transformation when temperature and existing internal stresses change simultaneously, convert before observation in the static cases. In other words, the internal stresses acting during the in-situ transformation allow the observed shear type stacking faults which transform immediately to non shear type ones when stress is relieved.

Glide processes parallel to basal planes are also proposed by Rios-Jara [22] to explain the preferential presence of certain types of stacking fault displacement vectors.

Considering moving stacking faults, it is possible to discuss the I.F. and resistance measurements, made in the same temperature interval (Fig. 6 a and b).

Coming from austenite, the resistance decreases immediately when reheating the specimen (Fig. 7, continuous line). This means, that there are still enough plates ending with tips at the end which are surrounded by elastic stress fields. These tips retract when changing the sign of $\dot{T}$, thereby decreasing the transformed volume. In the case of transforming from martensite practically no single plates with tips at their ends are present. So when heating, first stacking fault rearrangement occurs as shown in Fig. 3 without changing the transformed volume (Fig. 7, dotted line). Then retransformation take place creating some free ending tips. These tips start growing immediately when cooling again, explaining the superimposed cooling curves in both cases (Fig. 7).

The amplitude ε dependence of the I.F. evolution during transformation (Fig. 4) is not yet understood, but can be related to the number of migrating stacking faults. The higher I.F. usually measured inside the martensitic phase with higher ε values, can be explained by reorientation of martensite-martensite interfaces.

Conclusions

- Internal friction measurements and TEM in-situ transformation observations have shown that, in order to explain hysteresis effects, not only martensite-austenite interfaces, but also martensite-martensite interfaces have to be considered. Their mutual influence depends on the position inside the hysteresis (beginning, middle, end) and the phase distribution for which temperature and/or stress variations are considered. Martensite-martensite interfaces become locally highly mobile due to changes in stacking fault arrangements and thereby create high internal friction.

- The observed stacking fault movement is probably only possible due to the simultaneous action of temperature change and stress.

Without applied stress, still stacking fault movement is observed which makes evident the important role of internal stresses during martensitic transformation.

- Using relatively pure alloys, the migration of martensite-austenite interfaces is quite smooth after the early stages of growth. Dislocation tangles seem not to be visible obstacles to the migration of interfaces.

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