# Pattern formation in metallic glasses induced by helium-ion implanation. I. Experiments

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Ten different samples of metallic glasses were bombarded by 2- or 1-MeV helium ions. The sample temperature was kept below the crystallization temperature. Surface-deformation processes were dominated by flaking, i.e., from nearly the whole implanted area a layer flaked off with a uniform thickness corresponding to the ion range. The surface remaining behind the flaked layer showed a characteristic pattern. Such a periodic pattern did not appear on the annealed samples.

In the search for wall materials for future thermonuclear reactors, the peculiar properties of metallic glasses are now being studied extensively in many laboratories. A number of experiments[1–11] have been performed in an attempt to determine how long the surface of metallic glasses is able to resist intensive bombardment by light ions without deformation.

It is known from experiments[2,4,5,11] that the resistance of metallic glasses to H or He irradiation is considerably higher than that of their polycrystalline modifications. In view of this further studies on these materials seem to be promising.

In our first experiment on Metglas® 2826A bombarded by 2-MeV $He^+$ ions, it was experienced that almost the total bombarded area flaked off with uniform thickness corresponding to the implantation range and on the surface thus revealed there remained an interesting pattern, viz., a regular wavelike structure.[5] In the present paper we report on experiments aimed to clarify the conditions and the possible mechanism of this pattern formation.

To answer the question of how the appearance of the wavelike structure and its morphology depends on the experimental conditions, numerous irradiations were performed. For this purpose we used 2- and 1-MeV $^4He^+$ beams from our 5-MeV Van de Graaff accelerator. The samples were mounted on a target holder with good heat contact in a vacuum chamber of $7\times10^{-5}$ Pa. The implanation dose was determined from the integrated beam current and the area of the implanted spot. During bombardment the sample temperature was kept below the crystallization temperature by limiting the beam intensity. The temperature of the samples was estimated from the beam power, the heat conductivity of the samples, and the geometrical arrangement of the target holder. These values together with the other experimental parameters are listed in Table I.

For the present experiments, four different kinds of metallic glasses were used.

(1) Metglas® 2826A ($Fe_{32}Ni_{36}Cr_{14}P_{12}B_6$) manufactured by Allied Chemical Corporation (Morristown, New Jersey). It was produced by shooting the melted alloy onto a rolling cooled drum. The width and thickness of the ribbon produced in this way are 2 mm and 60 $\mu$m, respectively.

(2) $Fe_{80}B_{20}$ ribbon, produced by the Central Research Institute for Physics, Hungary using the same technology. Its width and thickness are 3 mm and 25 $\mu$m, respectively.

(3) $Ni_{76}P_{24}$ foil of thickness 14 $\mu$m produced also at the Central Research Institute for Physics, Hungary using different technology, viz., by means of electrolytic separation.

(4) $Ni_{76}P_{24}$ foil manufactured by the Csepel Metalworks, Hungary with the so-called electroless technology (chemical separation). The foil thickness is 150 $\mu$m.

Ten samples of these materials were prepared and implanted under different experimental conditions (see Table I). Samples *A*, *B*, *C*, *E*, *G*, *I*, and *J* were bombarded perpendicularly to their smooth, broad surfaces.

Sample *D* was prepared in the following way. A Metglas 2826A ribbon of 2 cm length was cut along its midline into two parts and one of them was folded in the middle to get a 1-cm-long "pair." This pair was fastened between two cooper blocks in such a way that the cut edges and the surfaces of both copper blocks were in the same plane. This surface of the sandwich was then polished mechanically and washed in an ultrasonic bath, after which it was bombarded with a beam spot of $3\times3$ mm$^2$.

Before implantation samples *C*, *G*, and *J* were annealed in a vacuum of $2\times10^{-3}$ Pa for 1 h at 400 °C or 600 °C (see Table I).

The bombardment of samples *F* and *H* was on their back surfaces (those surfaces which had been touched by the bearer during the separating process).

All the implanted samples were investigated by JEOL JSN-35 type scanning electronmicroscope. The implanted areas on all samples were dominated by flaking, i.e., from nearly all of the implanted spots a layer of thickness corresponding to the ion projected range flaked off (Fig. 1).

According to the literature concerning low-energy ion bombardment on metallic glasses the implanted surfaces were deformed by blistering. Our results show that in the case of MeV helium bombardment the dominant process is flaking, and blistering occurs only when samples of higher temperature are bombarded.

The critical doses that are needed for flaking on samples *I* and *J* (bombarded by 1-MeV $^4He^+$ ions) were determined and it was found that similarly to our earlier results[5] the resistance of the metallic glass sample to flaking is significantly higher than of the annealed one [$(1.5\pm0.1)\times10^{18}$ and $(0.8\pm0.1)\times10^{18}$ ions per cm$^2$, respectively].

The surface remaining behind the flaked layer of all

 

TABLE I. Samples, their implantation circumstances, and observations.

| Sample | Material | Energy (MeV) | Implanted surface | Dose ($10^{18}$ ions per cm$^2$) | Dose rate ($10^{13}$ ions per cm$^2$s) | Estimated temperature (°C) | Surface deformation |
|---|---|---|---|---|---|---|---|
| *A* | Metglas 2826A | 2 | smooth | 2.3 | 0.9 | 80 | flaking plus pattern |
| *B* | Metglas 2826A | 2 | smooth | 1.3 | 1.6 | 150 | flaking plus pattern |
| *C* | Metglas 2826A annealed at 400 °C | 2 | smooth | 2.3 | 0.9 | 80 | flaking plus pattern |
| *D* | Metglas 2826A | 2 | edge | 3.4 | 3.4 | 50 | flaking plus pattern |
| *E* | $Fe_{80}B_{20}$ | 2 | smooth | 2.4 | 1.8 | 400 | flaking plus blistering plus pattern |
| *F* | $Ni_{76}P_{24}$ Electro-litically separated | 2 | back | 1.3 | 1.61 | 250 | flaking plus blistering plus pattern |
| *G* | Metglas 2826A annealed at 600 °C | 2 | smooth | 2.8 | 1.9 | 50 | flaking only |
| *H* | $Ni_{76}P_{24}$ Electroless | 2 | back | 2.8 | 1.9 | 50 | flaking plus pattern |
| *I* | Metglas 2826A | 1 | smooth | 1.5 | 2.9 | 70 | flaking plus pattern |
| *J* | Metglas 2826A annealed at 600 °C | 1 | smooth | 1.5 | 2.9 | 70 | flaking only |

metallic glass samples contained regions on which a wavelike pattern was observable (Figs. 1–4). If before bombardment the sample was annealed at high temperature, i.e., if the metallic glass was fully crystallized (samples *G* and *J*), the above-mentioned structure did not appear. On the other hand, if annealing was just enough for the sample to relax but still to remain in the metallic glass state (sample *C*) the pattern formation henceforward was observable but the regularity was not so pronounced. Taking into account the accessible data concerning the morphology of surface deformations on other materials and our results on the 600 °C annealed samples one may conclude that such a structure does not appear on polycrystalline materials, so it must be related to the metallic glass state. Its appearance is independent of material composition (samples *A*, *E* and *H*), of manufacturing technology (samples *A*, *F*, and *H*), of implantation energy of helium ions (samples *A* and *J*), and of sample temperature during bombardment provided it is kept below the crystallization temperature (samples *A*, *B*, and *D*). The possibility that the pattern was originally there (i.e., developed during manufacture) is excluded, since our observations show that its appearance is independent of manufacturing technology and the pattern is in no way correlated with the direction favored in manufacturing. Moreover, we observed that the lateral positions and directions of the pattern are correlated with the border line of the bombarded spot.

In some cases the wavelike structure consisted of elevations of asymmetric triangular cross section (e.g., Fig. 2). One side of these elevations slopes up to a certain height with a small angle and its surface is rather smooth whereas the other descends steeply and not so smoothly, and is often extended in a zigzag line.

Generally the wave regularly extended for a relatively long distance, for example in the case of sample *A*, some hundreds of wave lines of 1 mm length followed each other in a parallel manner (Fig. 3). This surprising regularity is however, broken in some places by some kinds of imperfections. For instance, an outstanding diffractionlike picture can be seen around some local material inhomogeneity on Fig. 3. A typical imperfection of the wave structure appeared at those places where an additional line "comes" in, resembling an edge dislocation in a crystal (e.g., Fig. 4).

The surface on the central region of the implanted spot is rough in every case and going outwards, the waves appeared gradually. Usually at the region of its appearance the orientation is still ambiguous: Two directions are complete with each other and as a result an interference-

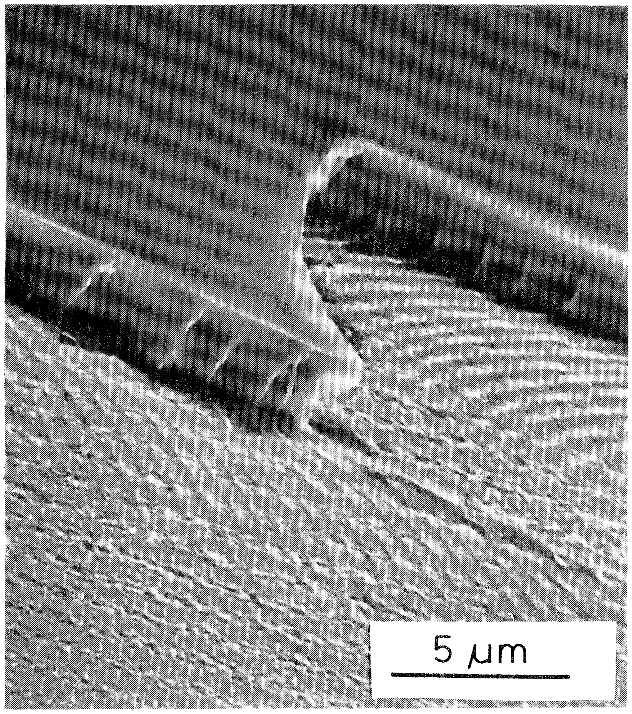


FIG. 1. Border of flaked area on sample *A*. Going upwards two regions covered by waves, the broken border of the flaked layer and the original surface can be seen.

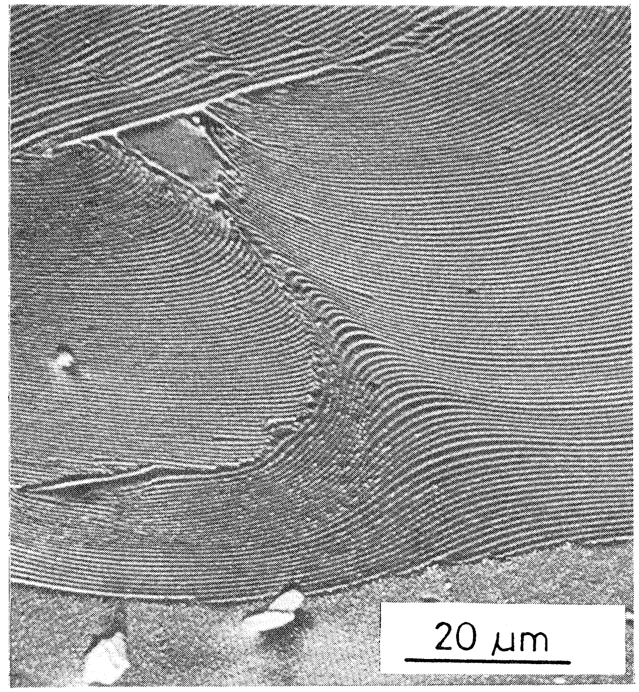


FIG. 3. Diffractionlike arrangement of waves on sample *A*.

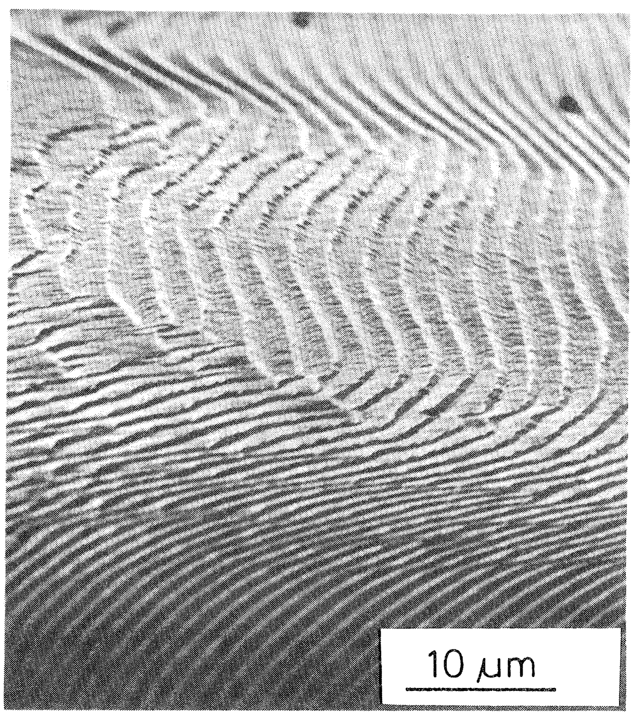


FIG. 2. Side view of wave structure on sample *A*.

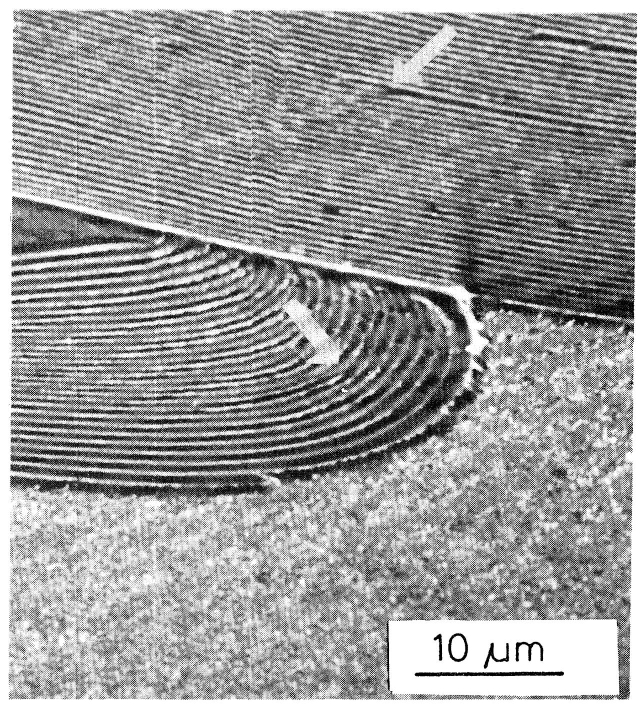


FIG. 4. Micrograph taken on sample *A*. Arrows show dislocationlike imperfections.

like picture is developed. Later on, one of these directions gradually disappears and a region of pure waves is formed. In the case of samples *A* and *D* this structure suddenly disappeared and was again followed by a rough surface. On sample *A* the region covered by waves developed three times.

Three further observations seem to be important: (1) the border of the last zone covered by waves is often given by the frame of the flaked layer and on the broken surface of the remaining part of the flaked layer the waves continue (Fig. 1); (2) the wavelength in all cases varies between 0.9 and 1.8 $\mu$m and its mean value does not change significantly by varying any one of the applied parameters, although on the same sample it can be rather different (e.g., Fig. 3); (3) on sample *D* where the irradiation was carried out on the edge of the ribbon the wave lines are directed not along the length of the strip but perpendicularly and the wave lines cross continuously the border line between the two stripes.

Summarizing the present experiment it is confirmed that compared with its crystalline modification metallic glass has a higher resistance against surface deformations caused by high-fluence bombardment of light ions.

In the case of MeV energy bombardment the dominant process is flaking and on the remaining surface left behind for each metallic glass a wavelike structure is observable. In contrast, the data accessible in the literature concerning low energy bombardment show that the dominant process was blistering[1–4,6–11] and wavelike structure was in no case observable.

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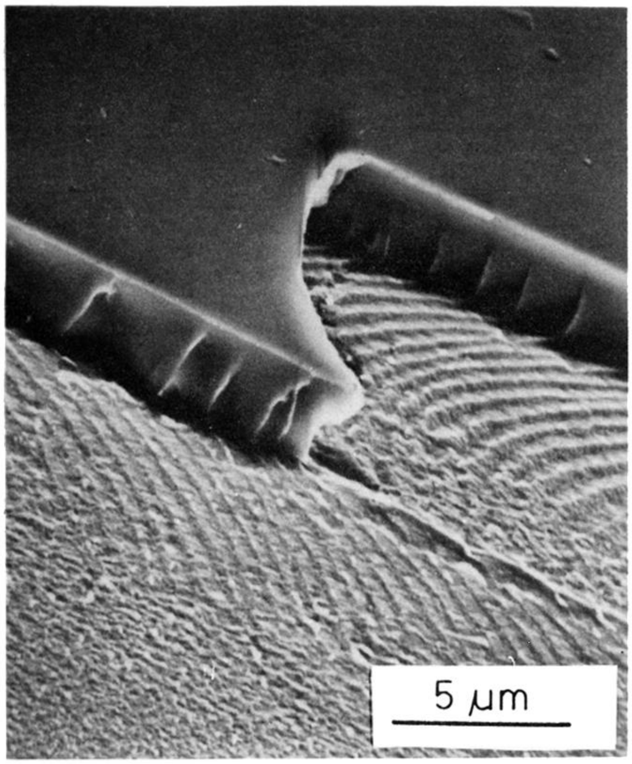


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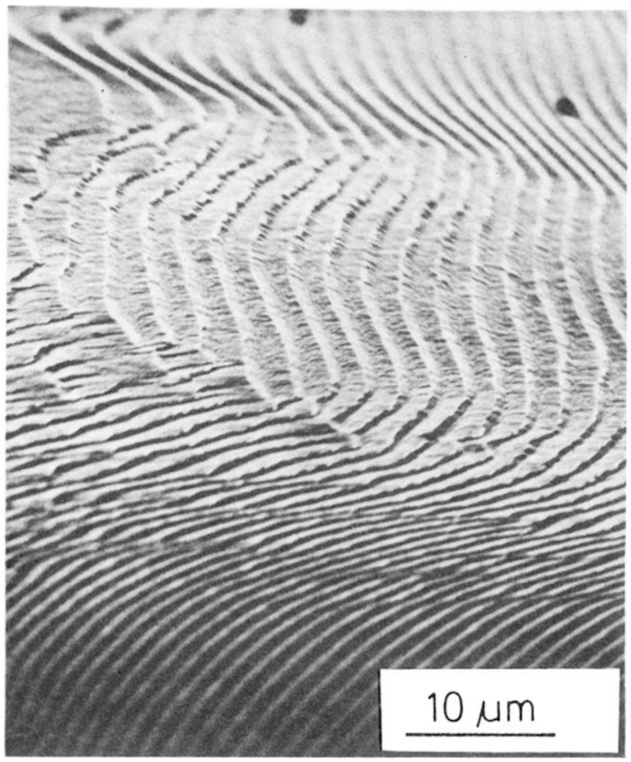


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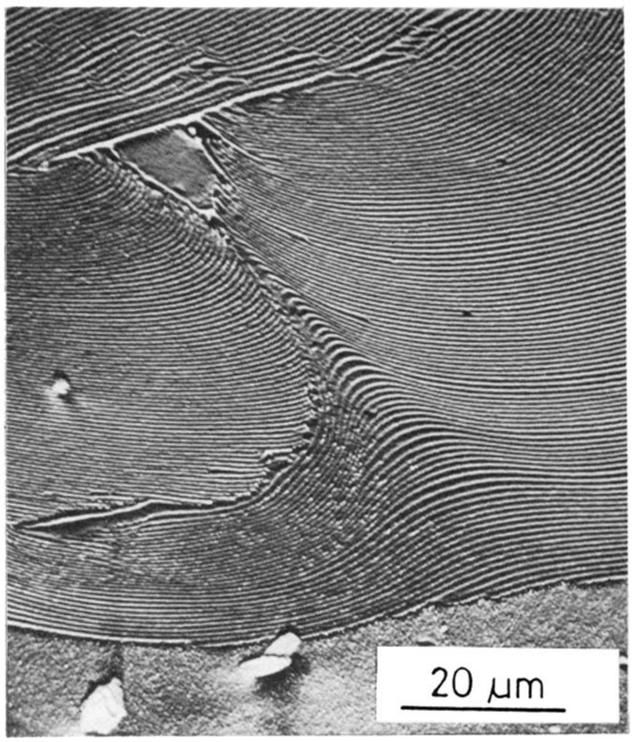


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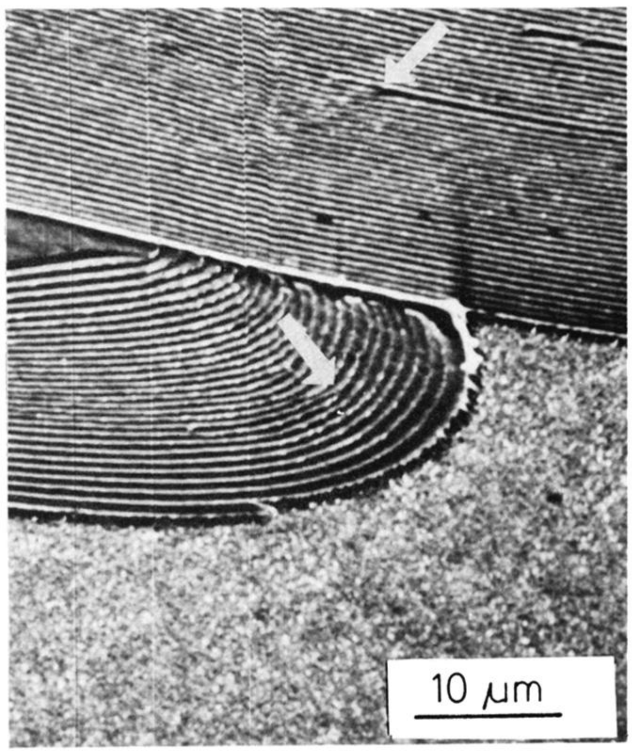


FIG. 4. Micrograph taken on sample *A*. Arrows show dislocationlike imperfections.