Ion beam-induced anisotropic plastic deformation of silicon microstructures

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Amorphous silicon micropillars show anisotropic plastic shape changes upon irradiation with 30 MeV Cu ions. The transverse plastic strain rate is $(2.5\pm0.2)\times10^{-17}$ cm^2/ion at 77 K, which is about one order of magnitude less than that of silica glass. In contrast, crystalline silicon pillars, irradiated under the same conditions, do not exhibit anisotropic deformation. A viscoelastic and free volume model is used to qualitatively describe the data. By irradiating partially amorphous structures a variety of silicon microshapes can be fabricated.
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Amorphous materials such as metallic and silica glasses, subjected to high-energy ion irradiation, show irreversible anisotropic plastic flow at temperatures far below the glass transition temperature;[1–3] these materials expand in the direction perpendicular to the ion beam and shrink in the direction parallel to the ion beam, while their volume remains constant. The deformation strain increases with ion fluence at a constant rate that increases with ion energy and decreases with increasing substrate temperature.[3] Recently, we found that this beam-induced anisotropic deformation leads to a dramatic shape change of free-standing colloidal particles.[4] The deformation was observed at ion energies as low as 300 keV.[5]

Ion beam deformation of silicon has not been studied in detail, with the exception of hydrogenated amorphous Si (*a*-Si:H) foils that did show deformation under 360 MeV ion irradiation.[6] As ion implantation of Si is a technologically important process, it is important to study deformation of this material. The deformation effect may then be exploited to tailor the shape of Si microstructures. In addition, by comparing ion irradiation effects in the crystalline and amorphous phases of Si, insights into the mechanism behind the deformation process may be acquired.

In this letter we investigate the irradiation of 1.5-μm-tall Si micropillars with 30 MeV Cu ions. We find that preamorphized pillars show anisotropic plastic deformation under irradiation, while crystalline pillars do not. We determine the deformation rate constant, compare our data with a viscoelastic and free volume model for deformation, and demonstrate the formation of microstructures in amorphous/crystalline composite structures.

Crystalline Si(100) pillars of 1.5 μm height were made by anisotropic reactive ion etching of a silicon-on-insulator substrate (1.5 μm Si/3.0 μm SiO_2/Si) in a SF_6/O_2 plasma using a method described in Ref. 7. By careful tuning of the etching parameters near-vertical sidewalls were achieved. Figure 1(a) shows a side-view scanning electron microscope (SEM) image (2° tilt), taken using a 20 keV electron beam, of Si pillars with a width of 1.8 μm. The pyramidal footprint in the contact region with the silica substrate layer is characteristic for the etching process and is caused by the reduced etch rate of Si in the (111) directions.[7]

Some pillars were turned amorphous using 3 MeV Xe irradiation at fluences up to 1.0×10^{15} cm^{-2}, while cooling

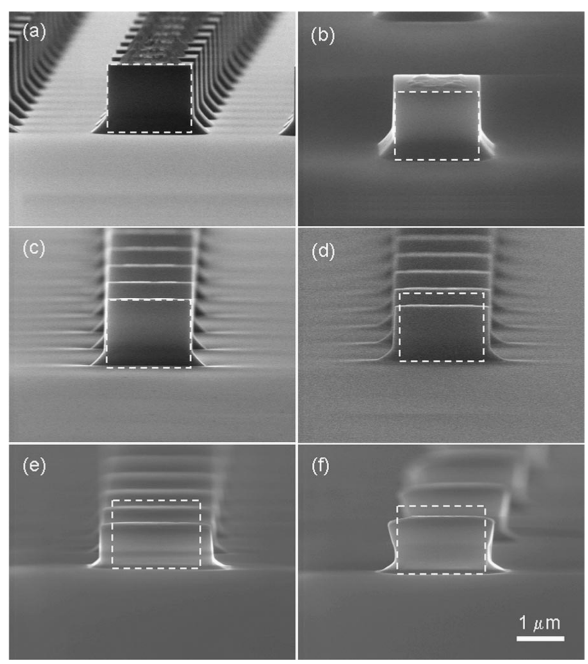

FIG. 1. Scanning electron microscopy images of (a) unirradiated *c*-Si(100) pillars, (b) Si pillars amorphized with 3 MeV Xe, 1.0×10^{15} cm^{-2}, (c) *c*-Si pillars irradiated with 30 MeV Cu, 1.8×10^{15} cm^{-2}, (d) *a*-Si pillars irradiated with 30 MeV Cu, 1.8×10^{15} cm^{-2}, (e) *a*-Si pillars irradiated with 30 MeV Cu, 8.2×10^{15} cm^{-2}, and (f) partially amorphized Si pillars, subsequently irradiated with 30 MeV Cu, 6.1×10^{15} cm^{-2}. SEM images were taken with a side-view tilt angle of $2°\pm1°$ [(a),(c),(d)–(f)] and $10°\pm1°$ (b). Magnification is the same for all micrographs.

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the substrate holder with liquid nitrogen. These irradiations were performed with the ion beam tilted 7° away from the normal to avoid ion channeling. The thickness of the amorphous layer was determined by channeling Rutherford backscattering spectrometry on a planar Si(100) sample irradiated under the same conditions, and was at least 1.5 μm. Electron backscatter diffraction patterns of the pillars during SEM confirmed that the pillars were amorphous.

Figure 1(b) shows a side-view SEM image (10° tilt) of amorphized silicon pillars. The dashed box in Fig. 1(b) outlines the outer boundaries of a pillar and is identical to that for the crystalline pillars in Fig. 1(a). Thus, no measurable deformation is observed after amorphization using 3 MeV Xe. Since ion beam-amorphized Si (a-Si) has a density that is 1.8% lower than that of crystalline Si (c-Si),[8] the pillars must have homogeneously expanded by about 0.6%. This expansion is too small to be observed in SEM.

Substrates containing both c-Si and a-Si pillars were clamped to a liquid-nitrogen cooled copper block and then irradiated with 30 MeV Cu ions to fluences in the range $(1.8–8.2)\times 10^{15}$ cm^{-2} using tandem accelerators in Rossendorf and Utrecht. The irradiations were performed at normal incidence or with the ion beam tilted 4° away from the normal to avoid channeling in the c-Si pillars.

Figure 1(c) shows a SEM image of c-Si pillars after irradiation to a fluence of 1.8×10^{15} cm^{-2} at an ion beam energy flux of 0.7 W/cm^2. No anisotropic deformation of the c-Si pillars is observed. Amorphous Si pillars [from Fig. 1(b)] irradiated under the same conditions are shown in the SEM image of Fig. 1(d). It can clearly be seen that the a-Si pillars have expanded perpendicular to the ion beam and contracted parallel to the ion beam direction, retaining the rectangular cross sectional shape. The relative lateral expansion is $12.2\pm 2.0\%$. From measurements of the transverse and longitudinal dimensions it follows that (within $\pm 3\%$) the a-Si pillar's volume has remained constant after deformation.

By comparing the data in Figs. 1(c) and 1(d) we thus conclude that there is a clear difference in ion beam-induced effects in c-Si and a-Si. Ion irradiation-induced anisotropic deformation is a universal phenomenon in amorphous materials.[9] The driving force for this effect is the electronic stopping S_e, the energy loss of the ions into electronic excitations and ionization of target atoms.[10] It has been demonstrated that the deformation effects in silica and metallic glasses can be well described by a viscoelastic model derived by Trinkaus and Ryazanov.[11] In this model it is assumed that thermal expansion of a cylindrically shaped region around the ion track leads to the generation of shear stresses that subsequently relax due to local atomic rearrangements in (overlapping) shear sites (regions with a local free volume).[9] The net effect of this shear stress relaxation is a local in-plane expansion perpendicular to the ion track, which freezes in upon cooling down of the thermal spike. Within this model, anisotropic deformation is not expected to occur in crystalline materials, due to the lack of shear sites.[2,9] In those materials, epitaxial recrystallization at the track's solid–liquid interface would restore the initial state, without anisotropic deformation. Our data on c-Si and a-Si provide clear experimental support for this model.

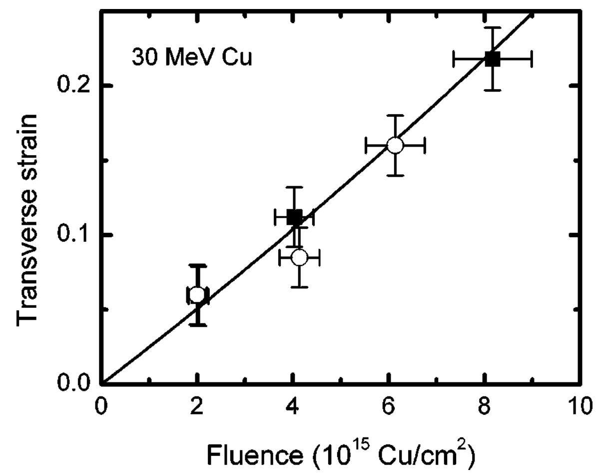

FIG. 2. Transverse strain of ion beam-amorphized Si pillars during 30 MeV Cu irradiation at a beam energy flux of 1.5 W/cm^2 as a function of Cu ion fluence (ϕ). The sample holder was cooled with liquid nitrogen. The solid squares refer to the deformation of Si pillars amorphized with 3 MeV Xe [e.g., Fig. 1(e)], the open circles to deformation of Si pillars amorphized with 1 MeV Xe [e.g., Fig. 1(f)]. The solid curve is a fit to the data (solid squares) using the relation $\varepsilon_T=\exp(A\phi)-1$ and results in $A=(2.5\pm 0.2)\times 10^{-17}$ cm^2/ion.

Figure 2 shows the fluence dependence of the transverse strain of a-Si pillars after irradiation with 30 MeV Cu ions at fluences in the range $(2.0–8.2)\times 10^{15}$ cm^{-2} at a beam energy flux of 1.5 W/cm^2 (solid squares). As can be seen, the transverse plastic strain increases gradually with ion fluence. A SEM image of the sample irradiated to a fluence of 8.2×10^{15} cm^{-2} is shown in Fig. 1(e); it shows a transverse plastic strain as high as $21.8\pm 2.1\%$. Note that the rectangular shape of the pillar with its pyramidal footprint remains conserved upon continued irradiation, except for the expansion factor. This is attributed to the radiation-induced lowering of the viscosity of the underlying SiO_2 substrate (a known effect[12]) that enables Newtonian plastic flow around the interface of the expanding a-Si and the SiO_2 substrate. Assuming a constant deformation rate constant A, the transverse plastic strain should increase exponentially with ion fluence ϕ: $\varepsilon_T=\exp(A\phi)-1$.[3] The solid line in Fig. 2 is a fit of the transverse strain data (solid squares) to this relation. From the fit we find that $A=(2.5\pm 0.2)\times 10^{-17}$ cm^2/ion.

To further illustrate how we can obtain local control over the shape of silicon microstructures by ion irradiation we have irradiated Si pillars that were partially amorphized. The amorphization was done by irradiating 1.5-μm-tall c-Si pillars with 1 MeV Xe ions to a fluence of 2.1×10^{15} cm^{-2} at 77 K. This resulted in the amorphization of a 710-nm-thick top layer. SEM micrographs did not show deformation of the Si pillars due to the 1 MeV Xe irradiation. Next, these pillars were irradiated with 30 MeV Cu ions to fluences of 2.0×10^{15}, 4.1×10^{15} and 6.1×10^{15} cm^{-2} at a beam energy flux of 1.6 W/cm^2, while cooling the substrate holder with liquid nitrogen. Figure 1(f) shows a SEM image of a partially amorphized pillar irradiated to a fluence of 6.1×10^{15} cm^{-2}. As can be clearly seen, the amorphous top part of the pillar has expanded whereas the bottom crystalline part has remained undeformed. Note that the expansion at the lower part of the amorphous region is constrained by the nondeforming c-Si underneath. The transverse strain measured at the top of the pillar is included in Fig. 2 for the three fluences used (open circles). These data fit well with the trend observed for the completely amorphous pillars.

At this stage we have clearly demonstrated that 30 MeV Cu irradiation of *a*-Si leads to anisotropic deformation with a well-defined rate constant. We can now compare the deformation of ion beam-amorphized Si with the well-known deformation of silica glass (SiO_2). Benyagoub *et al.* have studied the deformation of vitreous silica under high-energy ion irradiation.[3] From interpolation of their data a strain rate constant of 2.5×10^{-16} cm^2/ion is derived for silica at an electronic stopping equivalent to that in our experiment ($S_e = 6.0$ keV/nm). This is ten times larger than the strain rate found for *a*-Si in this work (Fig. 2). In addition, the deformation of colloidal silica particles upon MeV ion irradiation was found to be at least ten times larger than that of *a*-Si found here.[4] We can also compare the deformation of ion beam-amorphized Si with that of hydrogenated *a*-Si under 360 MeV Xe ion irradiation as studied by Klaumünzer *et al.*[6] From that work a deformation strain rate of at least 5×10^{-16} cm^2/ion can be estimated at $S_e = 6.0$ keV/nm, again much higher than the value found here for pure *a*-Si. Finally, Chicoine *et al.* have studied surface deformations during off-normal 24 MeV Se irradiation of several amorphous materials and found that lateral mass transport in pure *a*-Si was about 30 times smaller than in fused silica.[13]

In the viscoelastic model the deformation rate depends on the parameters that determine the effective diameter and the thermal expansion of the heated region around the ion track (i.e., S_e, specific heat, mass density, thermal expansion coefficient).[11] Using parameters for *a*-Si and silica, it is found that the calculated deformation rate of these two materials would be quite similar, contrary to what is observed. One possible explanation of the difference is that *a*-Si may not contain a high enough amount of free volume to mediate full shear stress relaxation. Indeed, the excess free volume in *a*-Si (relative to *c*-Si) is only 1.8%, whereas this difference for amorphous SiO_2 and quartz amounts to $\sim$17%. Similarly, the large free volume in *a*-Si:H (25 at. % H) may explain the much larger deformation rate of this material compared to pure *a*-Si. A counter argument, however, to the amount of free volume being a limiting factor determining the deformation is the fact that metallic glasses, that are known to have small free volume,[2] do show large anisotropic deformation strain rates, similar to SiO_2.

One phenomenon that distinguishes silicon from most other materials is that its molten phase has a larger density (by 8% at the melting point) than the solid. This would imply that the amorphous region around the ion track would initially expand as the temperature increases, but then contract as the ion track region becomes molten. This effect could cause a reduced deformation rate for *a*-Si compared to other amorphous materials such as SiO_2.

Finally, we note that not only thermally induced viscous flow but also ion irradiation-induced defects may cause shear stress relaxation in the ion tracks (and thus deformation). Thus, the density of such defects, or the radiation-induced viscosity, is also a parameter that might determine the deformation rate. Values for the radiation-induced viscosity during ion irradiation have been measured for both *a*-Si[14] and SiO_2,[12] and, under comparable conditions (2 MeV Xe), the viscosity of *a*-Si was found to be 5–10 times higher than that of SiO_2, implying that *a*-Si flows less than SiO_2.

A quantitative model of the deformation of *a*-Si must therefore necessarily include (at least) the effects of free volume, density change upon ion track melting and radiation-induced flow, and remains thus quite a challenge to establish.

In conclusion, ion beam-amorphized Si pillars show anisotropic plastic deformation under 30 MeV Cu ion irradiation at a transverse strain rate of $(2.5\pm0.2)\times10^{-17}$ cm^2/ion. In contrast, crystalline Si pillars irradiated under the same conditions do not show anisotropic deformation due to the lack of shear sites with local free volume. The relatively low value of the strain rate for pure *a*-Si compared to SiO_2 could be related to the smaller amount of free volume in *a*-Si, contraction of silicon upon melting, or its relatively high radiation-induced viscosity. By irradiation of partially amorphous structures, a variety of microshapes can be fabricated.

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