of ⟨100⟩ dark lines in semiconductor lasers.

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Order-Disorder Transition in Single-Crystal Silicon Induced by Pulsed uv Laser Irradiation

Raphael Tsu, Rodney T. Hodgson, Teh Yu Tan, and John E. Baglin
IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598

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Single-crystal silicon samples have been made disordered by irradiation with pulses from a frequency-quadrupled neodynium-doped–yttrium-aluminum-garnet laser with 10^{-8}-s pulse length. We have studied the resulting amorphous layer by transmission electron microscopy and He-ion backscattering. Irradiation with longer-wavelength pulses restored the disordered layer to its original crystalline state. This order-disorder laser-radiation–induced transition is repeatable.

A wide variety of amorphous films may be prepared by vacuum evaporation on cooled substrates. Some substances such as boron and germanium have been made disordered by the technique of splat quenching.[1] Except for using ion implantation, to the best of our knowledge, there has not been any report on the formation of a disordered layer on single-crystal silicon by thermal means. In principle, however, if a layer of the substance is melted and rapidly cooled, with the cooling time shorter than the time taken for epitaxial regrowth, silicon should become disordered.

In order to achieve such rapid cooling, a frequency-quadrupled, 10-ns pulsed Nd:YAlG (neodynium-doped, yttrium aluminum garnet) laser was used to heat a very thin layer at the crystal surface. At a photon energy of 4.7 eV, the absorption length is only 100 Å, and cooling rates in excess of 10^{11} °C/s may be achieved. This rate is at least an order of magnitude higher than any rate of epitaxial regrowth.

Figure 1 shows the calculated[2] temperature versus time, assuming that light is absorbed at

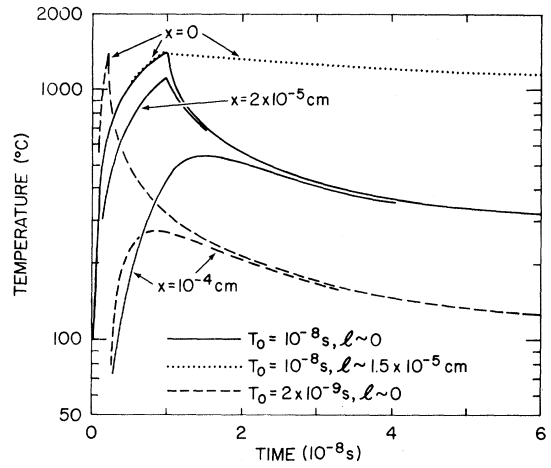

FIG. 1. Calculated temperature vs time for a silicon surface irradiated by a 10-ns, 4×10^6-W/cm^2, fourth-harmonic Nd:YAlG laser beam. T_0 is the pulse length, l is the absorption length, and the depth from the surface is denoted by x. The curves for $T_0=2\times10^{-9}$ s have been normalized to those for $T_0=10^{-8}$ s for easy comparison.

the surface and instantaneously converted into heat. The results for two pulse lengths, $T_0=10^{-8}$ and 2×10^{-9} s, are indicated by the solid and dashed curves, respectively. In order to simplify calculation, we have used an average thermal conductivity $K\sim0.85$ W/cm K instead of a temperature dependent $K(T)$. The incident power density used for the solid curve is 4×10^6 W/cm^2 corresponding to the typical values in our experiments. Reflection is taken to be 0.3. For comparison, the dotted curve in Fig. 1 is calculated for $T_0=10^{-8}$ s and an absorption length, l, of 1.5×10^{-5} cm appropriate to light at 2.35 eV, i.e., the frequency-doubled laser. Note that the maximum cooling rate at the surface, $X=0$, for $l\sim0$ is at least 50 times greater than that for $l\sim1.5\times10^{-5}$ cm. This explains why we cannot induce disorder in silicon with the frequency-doubled laser, since the absorption length exceeds 1.5×10^{-5} cm at 2.35 eV. In order to illustrate that the shorter the pulse, the more rapid is the rate of cooling near the surface, we have also plotted curves for the case of a pulse length of 2×10^{-9} s. Note that the cooling rate at a depth of 10^{-4} cm is not any faster for a 2×10^{-9}-s pulse than for a 10^{-8}-s pulse. Therefore, extremely high absorption is more important than an extremely short pulse for inducing disorder.

In preparing the samples for transmission-electron-microscopy and He-ion–backscattering studies, we have used a focused uv pulsed-laser beam with a diameter about half a millimeter. By overlapping scans, an area of 0.5 cm$\times$0.5 cm was obtained. The irradiated region has a typical appearance of an amorphous film, being somewhat milky. Because our laser has a 20% power fluctuation at the fourth harmonic, it was difficult to produce a large uniform area because of self-annealing back to the crystalline state at a wrong power level. We have repeated the process of producing an amorphous layer on the ⟨100⟩ silicon surface using the fourth harmonic, and recrystallizing back to single crystal with the second harmonic. There is no observable surface damage, even after repeating this process several times.

Transmission-electron-microscopy studies were carried out on ⟨100⟩ specimens prepared by jet thinning from their backsides to a thickness of less than 500 Å and examined with a Philips 301 electron microscope. Figure 2(a) shows the electron diffraction patterns which contain the halos typical of amorphous material. The positions of the first and second rings correspond closely to those of the amorphous Si prepared by ion bombardment. Note that the 220 reflections of the crystalline Si, top and bottom dots, lie noticeably off the second ring, which shows that our irradiated surface is not a microcrystalline layer.

An examination of the bright-field TEM micrograph showed that the amorphous silicon so produced exhibits thickness variations as shown in Fig. 2(b). The thickness variation is confirmed by the dark-field observation of the same area using the diffraction intensity in the first halo as shown in Fig. 2(c). Note that the dark area in Fig. 2(b) is now seen as the bright area in Fig. 2(c). Topographical images indicate the presence of thickness variation.[3] A slight variation in thickness of the laser-induced amorphous silicon may be due to local variation in impurities and defects.

In one speciman, we have observed isolated amorphous regions of 20- to 40-Å particle size, embedded in the matrix of single-crystal silicon. The diffraction pattern produced by such amorphous regions consisted of an essentially correct first halo, but the intensities normally associated with the second halo appeared to be more evenly distributed in reciprocal space between the first and second rings. We have seen the same phenomenon on an amorphous layer on single-crystal silicon thermally annealed at fairly low tempera-

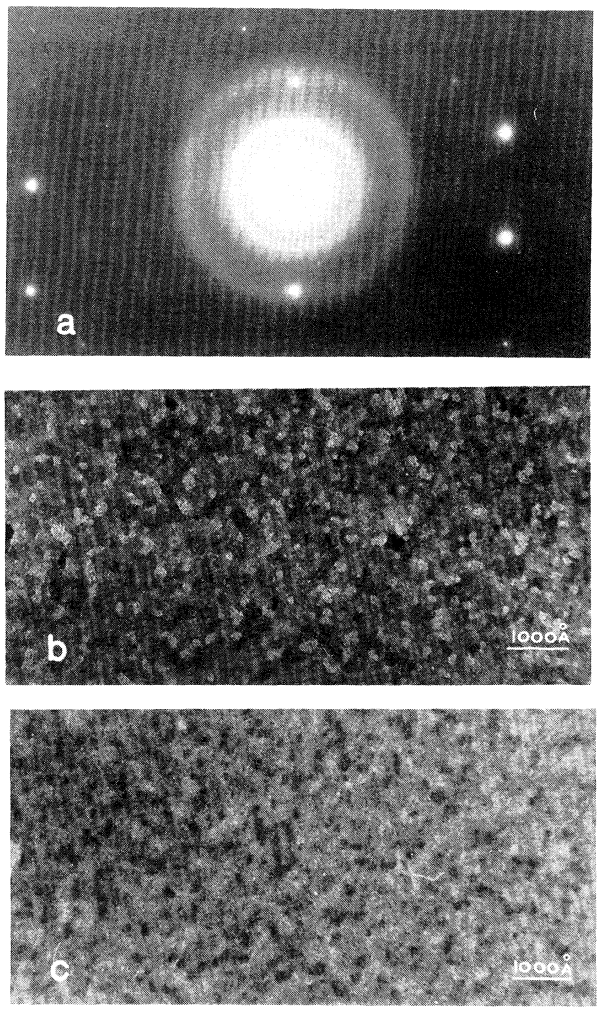

FIG. 2. (a) Electron diffraction pattern of irradiated region showing the typical halos of amporphous silicon; (b) bright-field image of amorphous silicon showing a thickness variation; (c) corresponding dark-field image of (b) taken with the diffraction intensity of the first (amorphous) halo. The diffraction pattern was obtained with an orientation few degrees from the ⟨100⟩ in order to maximize the intensity of the halos.

tures, i.e., 450°C for 30 min. More detailed investigation on this is being carried out.

To further confirm our characterization of the irradiated samples, we have performed axially channeled He-ion backscattering at 2.3 MeV. In Fig. 3 the scattering yield from the uv-laser-irradiated region shown as the solid curve is much higher than the curves for virgin Si⟨100⟩ and for the same sample after annealing with the second harmonic of the Nd:YAlG laser. The estimated mean thickness of the amorphous layer is 100 Å, which agrees quite well with the absorption data

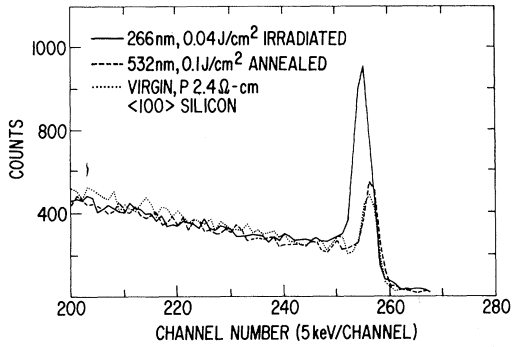

FIG. 3. Axially channeled He-ion backscattering of the amorphous, annealed, and virgin regions of a silicon sample having a ⟨100⟩ orientation.

for Si at 4.7 eV.[4] (Because the disordered layer is too thin to cause dechanneling of the ion beam, the scattering yield corresponding to channel numbers below 250 is as low as that of the crystalline silicon.)

Preliminary results indicated that it is much easier to induce disorder on Si⟨111⟩ than on Si⟨100⟩. Intuitively, Si⟨111⟩ requires the alternate formation of triple and single bonds resulting in slower rate of regrowth. Recently, Lau, Mayer, and Tseng[5] have observed a factor of 25 higher growth rate for the ⟨100⟩ surface than for the ⟨111⟩ surface.

Since the experimental results of this paper were obtained, silicon has been made amorphous using picosecond pulses at the first, second, and fourth harmonics of a Nd:YAlG laser.[6] Presumably, nonlinear processes limit the absorption depth sufficiently so that the longer-wavelength light is almost as effective as the ultraviolet. Combinations of pulse length and wavelength could be used to investigate the detailed nature of this disorder formation and crystal growth in general.

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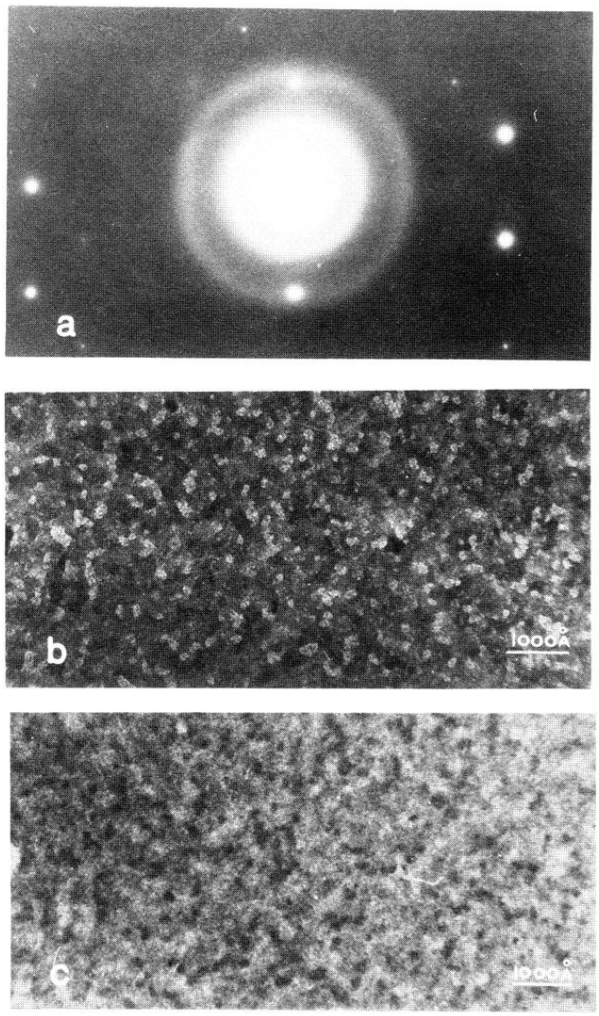

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