CONF-851217-24

[To be published in the Proceedings of the Materials Research Society, Symposium A, "Beam-Solid Interactions and Phase Transformations," held December 2-7, 1985, Boston, Massachusetts.

CONF-851217--24

DE86 005016

OVERHEATING IN SILICON DURING PULSED-LASER IRRADIATION?

B. C. Larson, J. Z. Tischler, and D. M. Mills

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

By acceptance of this article, the publisher or recipient acknowledges the U.S. Government's right to retain a nonexclusive, royalty-free license in and to any copyright covering the article.

SOLID STATE DIVISION
OAK RIDGE NATIONAL LABORATORY
Operated by
MARTIN MARIETTA ENERGY SYSTEMS, INC.
under
Contract No. DE-AC05-84OR21400
for the
U.S. DEPARTMENT OF ENERGY
OAK RIDGE, TENNESSEE 37831

December 1985

MASTER

OVERHEATING IN SILICON DURING PULSED-LASER IRRADIATION?*

B. C. Larson, J. Z. Tischler, and D. M. Mills[†]

Solid State Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831

[†]CHESS and Applied and Engineering Physics, Cornell Univ., Ithaca, NY 14853

ABSTRACT

Nanosecond resolution time-resolved x-ray diffraction measurements of thermal strain have been used to measure the interface temperatures in silicon during pulsed-laser irradiation. The pulsed-time-structure of the Cornell High Energy Synchrotron Source (CHESS) was used to measure the temperature of the liquid-solid interface of $\langle 111 \rangle$ silicon during melting with an interface velocity of 11 m/s, at a time of near zero velocity, and at a regrowth velocity of 6 m/s. The results of these measurements indicate 110 K difference between the temperature of the interface during melting and regrowth, and the measurement at zero velocity shows that most of the difference is associated with undercooling during the regrowth phase.

INTRODUCTION

The relationship between interfacial undercooling and crystal growth in silicon has been of interest both from the standpoint of fundamental crystal growth thermodynamics[1] and with regard to the amorphization of silicon for crystal growth rates of more than 15 m/s. It has been suggested that >200 K undercooling is required to drive 15 m/s crystal growth and that such undercooling favors a liquid-amorphous transition rather than a liquid-solid transition.[2] Likewise, the relationship between overheating and melting velocity is of interest.

We have previously reported[3] time-resolved x-ray diffraction measurements of the overheating and undercooling of silicon during pulsed laser irradiation; a greater than 100 K difference was measured between the interface temperature during 11 m/s melting and the interface temperature during 6 m/s regrowth along the $\langle 111 \rangle$ direction. Because of uncertainties in the absolute temperature scale brought about by uncertainties in the temperature dependence of the elastic constants and the thermal expansion coefficients, the proportion of overheating and undercooling could not be determined, however. We have now analyzed additional time-resolved x-ray scattering measurements made at a time of near-zero interface velocity, and in this paper we present results that separate the overheating and undercooling contributions.

EXPERIMENTAL

We have used nanosecond resolution time-resolved x-ray diffraction measurements of thermal expansion induced strain to determine the liquid-solid interface temperatures in Si during pulsed-laser irradiation. The time-resolved nature of the measurements and the analysis procedures have been discussed elsewhere[3,4] and only a brief description of the experiment will be presented here. Figure 1 shows a schematic picture of x-ray scattering from a crystal with a depth dependent thermal gradient.

*Research sponsored by the Division of Materials Sciences, U.S. Department of Energy under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

Fig. 1. Schematic representation of the scattering geometry depicting the thermally strained region of the laser irradiated sample and a schematic view of the x-ray scattering pattern resulting from the thermal strain distribution.

Through an analysis of Bragg scattering profiles, thermal strain profiles have been determined and lattice temperature distributions have, in turn, been extracted from the strain using the thermal expansion coefficients. Laser pulses of 248 nm wavelength and 25 ns time duration (as shown in Fig. 2) were synchronized with x-ray pulses from the Cornell High Energy Synchrotron Source (CHESS) to achieve nanosecond resolution x-ray diffraction measurements. The laser pulses used had energy densities of 1 J/cm^2 and the silicon samples were $\langle 111 \rangle$ oriented wafers of 0.5 mm thickness. The x-ray measurements were made in a step scanning mode in the region of the Bragg reflection using 18 laser shots at each angular setting of the crystal; the jitter in the laser firing was $\pm 2 \text{ ns}$.

Figure 2 shows heat flow simulations (using a computer code supplied by M. O. Thompson)[5] of the laser melting and regrowth process pertaining to the measurements in this experiment. The laser power, the interface velocity, and the melt-front penetration depth are shown as a function of time. As can be seen in the figure, measurements made at 15 and 45 ns correspond to interface velocities of 11 m/s melting and 6 m/s regrowth, respectively. Measurements made at these times have been reported previously; of particular importance for this work is the interface velocity indicated for measurements made at 27 ns. This time corresponds to that of maximum melt-front penetration and hence represents the time at which the interface velocity vanishes as the interface temperature passes through the equilibrium melting temperature.

RESULTS

Figures 3,4 show time-resolved x-ray measurements on $\langle 111 \rangle \text{ Si}$ for delay times of 16, 27, and 45 ns relative to the arrival time of the laser pulses. The error bars on the measured data represent the statistical uncertainty in the measured data and the solid lines correspond to least-squares fits of the data with x-ray scattering calculations for silicon containing a strained surface layer. In the fitting procedure the lattice strains as a function of depth were used as the adjustable parameters. The full lattice temperature distributions corresponding to Figs. 3,4 have been reported elsewhere and will not be presented here;[6] however, the

REPRODUCED FROM
BEST AVAILABLE COPY

Fig. 2. A composite view of the laser pulses and the timing used in this experiment together with calculations of the liquid-solid interface velocity and melt depth for 1 J/cm^2 , 248 nm laser pulses on silicon.

temperature distributions to depths of 1500 Å are shown in Fig. 5. The hatched areas indicate the estimated uncertainty in the determination of the lattice temperatures and it can be seen that the measured difference in the interface temperatures during melt-in (16 ns) and regrowth (45 ns) is $110 \pm 30 \text{ K}$. Of most significance, though, is the temperature profile for the 27 ns case. We see that the interface temperature at 27 ns (which corresponds to a time of near zero velocity) falls in the same range as that measured at 16 ns (11 m/s velocity). That is, the 110 K temperature difference seems to be associated mainly with undercooling during the 6 m/s regrowth while only a small fraction is associated with overheating during the 11 m/s melt-in.

The estimated precision in the interface temperatures is $\pm 25 \text{ K}$ for the 16 and 27 ns measurements and $\pm 10 \text{ K}$ for the 45 ns measurements. Within these uncertainties we can assign velocity coefficients for undercooling and overheating (defined by $\Delta T_U = \beta^U v$ and $\Delta T_O = \beta^O v$) for $\langle 111 \rangle$ silicon. Using these values we get limiting values of $\Delta T_O < 60 \text{ K}$ and $145 > \Delta T_U > 70 \text{ K}$, which in turn lead to $\beta^O < 6 \text{ K/m/s}$ and $11 < \beta^U < 24 \text{ K/m/s}$.

DISCUSSION

The result to be emphasized here is that overheating is observed to be small so that most of the measured differences in the interface temperatures during melt-in and regrowth must be attributed to undercooling during regrowth. Recognizing that β^O must be greater than zero, the midrange of the allowed β 's, consistent with the measurements made here, are $\beta^O \sim 3 \text{ K/m/s}$ for overheating and $\beta^U \sim 17 \text{ K/m/s}$ for undercooling. These values are to be compared with values of $\beta^U = 15 \pm 5 \text{ K/m/s}$ reported by Galvin et al.[7] from transient conductivity measurements on 3.5 ns ruby-laser irradiated $\langle 100 \rangle$ silicon-on-sapphire (SOS) and they can be compared

Fig. 3. Time-resolved Bragg profile measurements made at the (111) reflection on $\langle 111 \rangle$ oriented silicon during pulsed-laser irradiation. The solid lines are fits to the data measured at 16 ns and 45 ns.

Fig. 4. Time-resolved Bragg profile measurements made under the same conditions as those in Fig. 3, but at a time of 27 ns.

to $\beta^U = 17 \pm 3$ reported by Thompson et al.[8] from the analysis of transient conductivity measurements on 28 ns ruby-laser irradiated SOS. Thompson et al.[8] reported much lower values of $\beta^U = \beta^0 = 4 \pm 2 \text{ K/m/s}$ (for low interface velocities) obtained from the analysis of optical transmission measurements on picosecond range UV laser pulses on silicon. Although Thompson et al. expressed caution in interpreting the

Fig. 5. A composite view of the temperature profiles at the liquid-solid interface corresponding to the fits to the measured data for 16, 27, and 45 ns in Figs. 3,4.

picosecond results, the data presented showed significant non-linearities at high melting and regrowth velocities and indicated the velocity coefficient for overheating to be smaller than that for undercooling.

The fact that the indicated interface temperature in Fig. 5 for 27 ns is $1478 \pm 25^{\circ}\text{C}$ rather than 1412°C (as would be expected for the equilibrium melting temperature) is interpreted to be a result of inaccuracies in the temperature dependencies of the elastic constants and the thermal expansion coefficients. That is, because data on the temperature dependence of the elastic constants of silicon above 900°C is not available and because the temperature dependence of the thermal expansion coefficients are not believed to be accurate to better than $\sim 2\%$, the absolute temperature scale inferred from the lattice strain measurements has relatively large uncertainties. The relative temperatures are expected to be accurate, however, because systematic errors present in the absolute magnitudes are largely eliminated in the subtraction.

The large range of values reported in the literature for the velocity coefficients for overheating and undercooling indicates the need for additional work in this area. The transient conductivity results were made on $\langle 100 \rangle$ SOS while the results presented here were for $\langle 111 \rangle$ silicon; x-ray measurements are not available for the 27 ns time on $\langle 100 \rangle$ silicon, but the temperature differences measured between melt-in and regrowth are about half that found for the $\langle 111 \rangle$ direction. Additional measurements with higher resolution and lower uncertainties would be helpful with each of the techniques, and additional theoretical work is needed in this area as well.

REFERENCES

- [1] J. W. Cahn, S. R. Coriell, and W. L. Boettinger, p. 89 in Laser and Electron Beam Processing of Materials, ed. by C. W. White and P. S. Peercy, Academic Press, NY (1980).
- [2] A. G. Cullis, p. 16 in Energy Beam-Solid Interactions and Transient Thermal Processing 1984, ed. by D. K. Biegelson, G. A. Rozgonyi, and C. V. Shank (Materials Research Society, 1985).
- [3] B. C. Larson, J. Z. Tischler, and D. M. Mills, p. 187 (same as Ref. 2).
- [4] B. C. Larson, C. W. White, T. S. Noggle, J. F. Barhorst, and D. M. Mills, Appl. Phys. Lett. **42**, 282 (1983).
- [5] M. O. Thompson, private communication (see Ref. 8).
- [6] B. C. Larson, J. Z. Tischler, and D. M. Mills, J. Mat. Res. (in press).
- [7] G. J. Galvin, J. W. Mayer, and P. S. Peercy, Appl. Phys. Lett. **46**, 644 (1985); P. S. Peercy and M. O. Thompson, p. 54 in Energy Beam-Solid Interactions and Transient Thermal Processing 1984, ed. by D. K. Biegelson, G. A. Rozgonyi, and C. V. Shank (Materials Research Society, Pittsburgh, PA, 1985).
- [8] M. O. Thompson, P. H. Bucksbaum, and J. Bokor, p. 181 in Energy Beam-Solid Interactions and Transient Thermal Processing 1984, ed. by D. K. Biegelson, G. A. Rozgonyi, and C. V. Shank (Materials Research Society, Pittsburgh, PA, 1985).