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The Study of Shock Launching in Silicon by Pulsed X-Ray Diffraction

J. S. WARK

Laboratory for Laser Energetics
University of Rochester, NY 14623

R. R. WHITLOCK

Condensed Matter Physics Branch
Condensed Matter and Radiation Sciences Division

A. HAUER

Los Alamos National Laboratory
Los Alamos, NM 87545

J. E. SWAIN AND P. J. SOLONE

Lawrence Livermore National Laboratory
Livermore, CA 94550

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Multikilobar shocks were launched into a single crystal of (111) silicon overcoated with 1000 Å aluminum and a 25 μm transparent plastic layer by irradiation with a 1 nsec pulse of 1.06 μm laser light at 0.8-8 J cm^{-2}. Peak lattice compressions (densities) were directly measured and shocked lattice stresses inferred as a function of irradiance and time by Bragg diffracting a short ($<$100 psec) burst of probing x-rays through the shock launching region. Compressions of up to $\sim$4% were measured, corresponding to stresses of order 70 Kbar, at irradiances of $\sim 4 \times 10^9$ W cm^{-2}.

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Robert R. Whitlock

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*Laboratory for Laser Energetics, University of Rochester, NY 14623
+Los Alamos National Laboratory, Los Alamos, NM 87545
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THE STUDY OF SHOCK LAUNCHING IN SILICON BY PULSED X-RAY DIFFRACTION

The laser generation of shocks encompasses a wide range of physical processes and effects,[1-3] since virtually every property of condensed matter can be altered with pressure. For the more conventional shock drivers, the shock launching region, i.e. the initial layer of compressed material, is obscured by the driving mechanism. Whilst laser driven shocks do not present this limitation, so far *direct* measurements of the lattice spacing of the impact surface have been intractable. Such measurements are of import because many physical phenomena not in agreement with classical dislocation theory, such as elastic-plastic interactions, originate at this surface.

In this letter we report results of an experiment in which, for the first time, we have probed the initial few (4-5) microns of the shock launching region in a crystal lattice with pulsed x-rays, and directly observed the spatial and temporal evolution of lattice compression and subsequent relaxation in one dimension by Bragg x-ray diffractometry. We observe the existence of a range of compressions (lattice spacings) in the launch phase, and have also examined the experimentally determined peak compression as a function of laser irradiance.

The use of x-rays to measure the compression of shocked materials was first performed by Johnson and co-workers in the early 1970's.[4] They shocked LiF to several hundred kilobars using conventional explosive techniques,[5,6] and diffracted a 40-50 nsec pulse of x-rays[7] off the shocked material. However, to our knowledge, the work presented here is the first of a similar nature to be presented since then, with a five hundred-fold improvement in temporal resolution, and it is the first time that either the spatial or temporal development of the compression within the launch region has been studied directly.

The experiment was performed on the JANUS research laser system at the Lawrence Livermore National Laboratory, California. The experimental setup is shown in schematic form in Fig. 1. The shocked targets consisted of 250 μm thick (111) silicon wafers 5 cm in diameter, the surface of which had been coated first with 1000 Å aluminum and then with 25 μm plastic (CH). The motivation for such a target design is explained below. Half of the target was irradiated in a vacuum by a 1 ± 0.05 nsec pulse of 1.06 μm laser light at an irradiance varying from 0.8 to 8 J cm^{-2} with a beam diameter on target of 3.9 cm. A beam block prevented irradiation of the other half of the target; diffraction from this unshocked region gave us a reference point from which to measure the changes in Bragg angle. After the shock had been launched in the silicon crystal, a second laser beam containing $\sim 10J$ of 0.53 μm light in 100 psec, which was synchronous with known and variable delay with respect to the shock launching beam, was focused to a ~ 40 μm diameter spot on a second, calcium-containing target. The ionized helium-like calcium x-ray lines thus produced[8] were Bragg diffracted off the silicon (111) planes and recorded on Kodak SB5 x-ray film. The reduction in x-ray intensity due to passage through the 25 μm plastic overcoat is estimated to be $\sim 35\%$, with a further $\sim 4\%$ reduction due to passage through the aluminum. A single laser shot provided recordable x-ray levels; a series of shots was taken to obtain data at different irradiances and delay times.

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Previous experiments at irradiances similar to those used here have shown that uncoated silicon is simply heated by the laser light rather than shocked,[9] due to the several millimeter absorption length of 1.06 μm light in silicon at room temperature. The aluminum coating was therefore applied to act as an abrupt absorber to the incident radiation. This effectively prevented the penetration of the laser light into the silicon, and instead produced an aluminum plasma which in turn generates a pressure pulse for driving the shock into the material. Overcoating the aluminum with plastic transparent to 1.06 μm light causes the expanding aluminum plasma to be inertially confined between silicon and plastic, which in turn increases the strength of the shock launched into the silicon. Such overcoating techniques have previously been used to enhance shock pressures in the tens of kilobar range.[10]

A set of typical diffraction patterns is shown in Fig. 2. The three lines in the unshocked region are the resonance line (3.177 Å), the intercombination line (3.193 Å), and the unresolved j, k dielectronic satellites (3.21 Å) of helium-like calcium.[8] In the shocked region of the crystal, the lattice spacing d is reduced, increasing the diffraction angle θ in accordance with a differentiation of Bragg's law:

$$\Delta(2d)/2d = -\cot\theta \, \Delta\theta.$$

This effect can clearly be seen in Fig. 2. Thus a simple measurement of the maximum angular shift gives us a direct measurement of the peak compression of the (111) planes, and knowledge of the distribution of angular shifts yields information on the distribution of (111) lattice spacings within the probed region.

The probe depth of the x-rays in such highly strained crystals is determined by the photoelectric absorption coefficient rather than the extinction length. This is because x-rays incident at a particular angle only diffract off that particular region of the crystal at which the lattice spacing is such that the Bragg condition is satisfied; the rest of the crystal up until that point (with slightly different lattice spacings) simply acts as an x-ray filter. A Beer's law attenuation factor of $1/e$ for the helium-like calcium lines in silicon corresponds to an x-ray path length of 8.5 μm, or a depth of 4.3 μm below the surface at the Bragg angle. Taking further account of the signal dynamic range we estimate a probe depth of 4-5 μm below the surface.

Diffraction measurements such as those shown in Fig. 2 were made for a variety of levels of irradiance and delay times. We define the laser beam delay as the interval between the arrival, at chamber center, of the peak of the long (shock-producing) laser pulse followed by the peak of the short (x-ray producing) laser pulse. The x-ray probe delay was 0.09 nsec greater than the delay between laser beams, due to the setup geometry and time of flight considerations. It can be seen from Fig. 2 that at 0.0 nsec delay we still observe diffraction at the original Bragg angle as well as diffraction over a range of angles, corresponding to a range of lattice spacings up to a peak compression of 3.35%. The obvious physical explanation for this is that, at this early time, the shock has not yet penetrated past the maximum probe depth, and we are probing the shock front as well as the as yet unshocked region. The range of lattice spacings represents a spatial strain gradient. For all data collected after this delay time, when the shock has proceeded further into the material, we no longer observe diffraction from the unperturbed crystal.

In Fig. 3 we show the peak change in interatomic spacing of the (111) planes as a function of time for those shots where the average energy density of the shock-producing laser pulse was 4 J cm^{-2} on target (i.e. an average irradiance of 4×10^9 W cm^{-2}). The accuracy of the laser timing may include a constant systematic error estimated to be no more than ±200 psec; random timing fluctuations were evidently of lesser magnitude. The error bar on the compression measurement primarily results from a consideration of observed effects of laser non-uniformities on the maximum Bragg diffraction angle. (These effects can be seen in Fig. 2a as a variation in angular shift in the diffracted radiation.) Other effects, such as intrinsic x-ray line width, crystal rocking curve breadth, and x-ray source broadening, are of considerably lesser importance. A maximum change of the lattice spacing of 3.85% was observed at this energy density, and the data are consistent with a pressure pulse FWHM of 1 nsec. It can be seen that the rise time of the shock compression is similar to that of the laser pulse. However, after peak compression has been reached, the compression falls off with a far longer decay time than that of the laser. Indeed, even 5 nsec after the peak of the shock-producing pulse, the crystal is still compressed by ~1%. At no time did we observe the front-surface density to fall below that of solid. Such inhibition of the rarefaction wave has previously been observed by other diagnostic methods[11] (using similar layered targets), and is due to the tamping effect of the plastic.

The Hugoniot elastic limit (HEL) has been reported to occur in (111) silicon at a compression of 2.6%,[12,13] corresponding to a pressure of about 54 Kbar. Following the stress-volume curve of Gust and Royce[12] to the observed 3.85% compression we estimate a peak stress of 67 Kbar at an energy density of 4 J cm^{-2}. However, in comparing these results to Hugoniot measurements, it should be borne in mind that we are probing the shock launching phase, rather than merely observing the arrival of an already steepened shock front. Furthermore, elastic response may extend above the HEL on a transient basis.[14,15]

Figure 4 shows the experimentally measured peak compressions as a function of irradiance for various probe delay times. The rapid falloff in compression at low irradiances is due to the increased importance of thermal conduction into the silicon at lower irradiances,[11] as well as an increase in the fraction of laser energy expended in the latent heat of vaporization of the aluminum.

The technique of pulsed x-ray diffraction from laser-shocked materials opens up several new avenues for the study of the transient response of crystal lattices. We have shown here that it is possible to measure one vector component of the strain on the front surface of a shocked crystal. The temporal resolution afforded by this technique, coupled with its ability to probe crystal structure, may allow us to time-resolve pressure-induced phase transitions, as well as study possible transient elastic-plastic effects in crystals compressed beyond the HEL.[14,15] Furthermore, a detailed study of the x-ray reflectivity as a function of angle may yield compression-depth information using methods similar to those developed for studying laser-annealed crystals.[16,17]

In conclusion we have directly observed the temporal history of lattice compression within the front surface of a laser-shocked crystal by short pulse x-ray diffraction. We have observed diffraction from material both in front of and behind the developing shock front at early stages during the laser drive pulse. We have followed the lattice compression to values above the HEL, and further observed the onset of rarefaction as the pressure pulse decayed. Crystallinity was preserved throughout this process.

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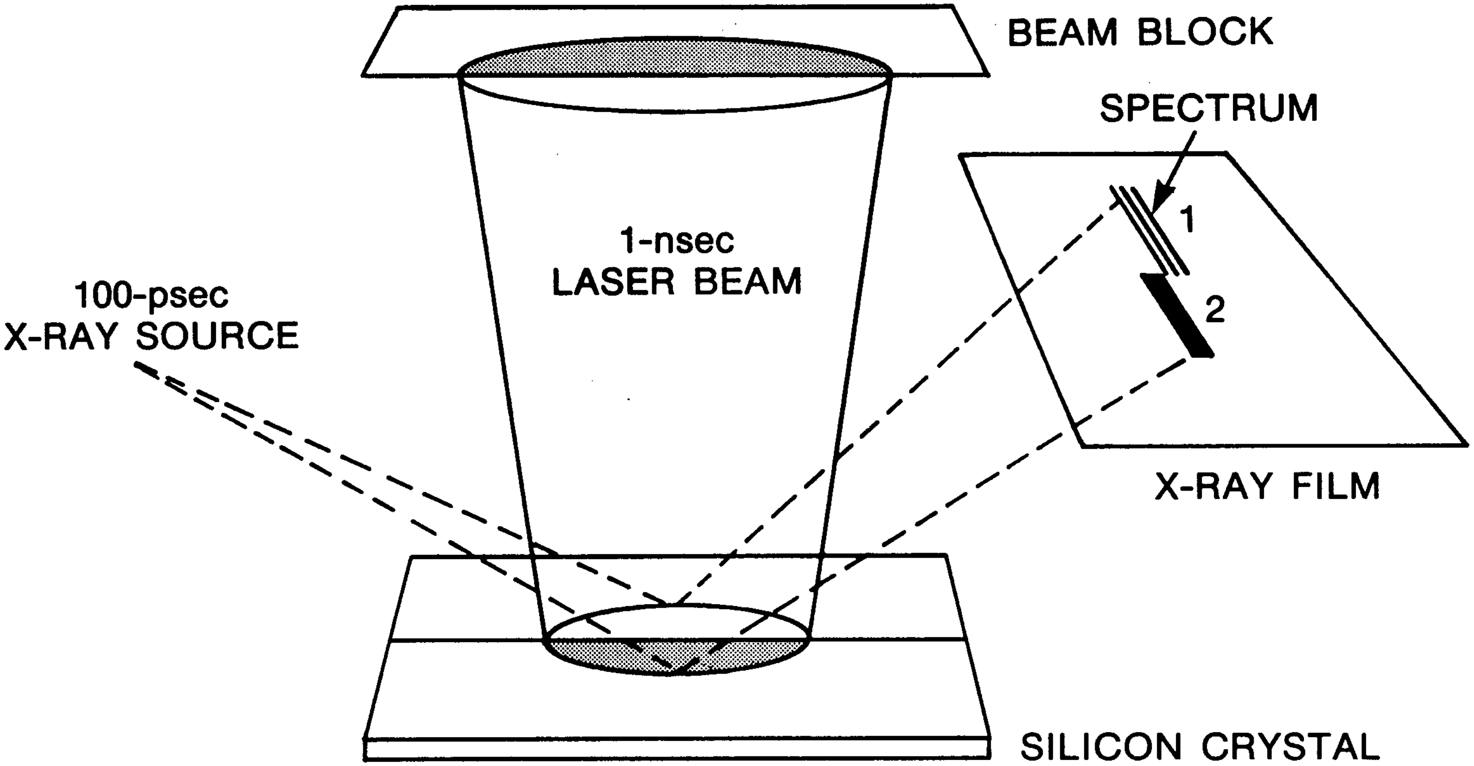

Fig. 1 — The experimental setup showing schematically the x-ray diffraction from the unshocked (1) and shocked (2) crystal

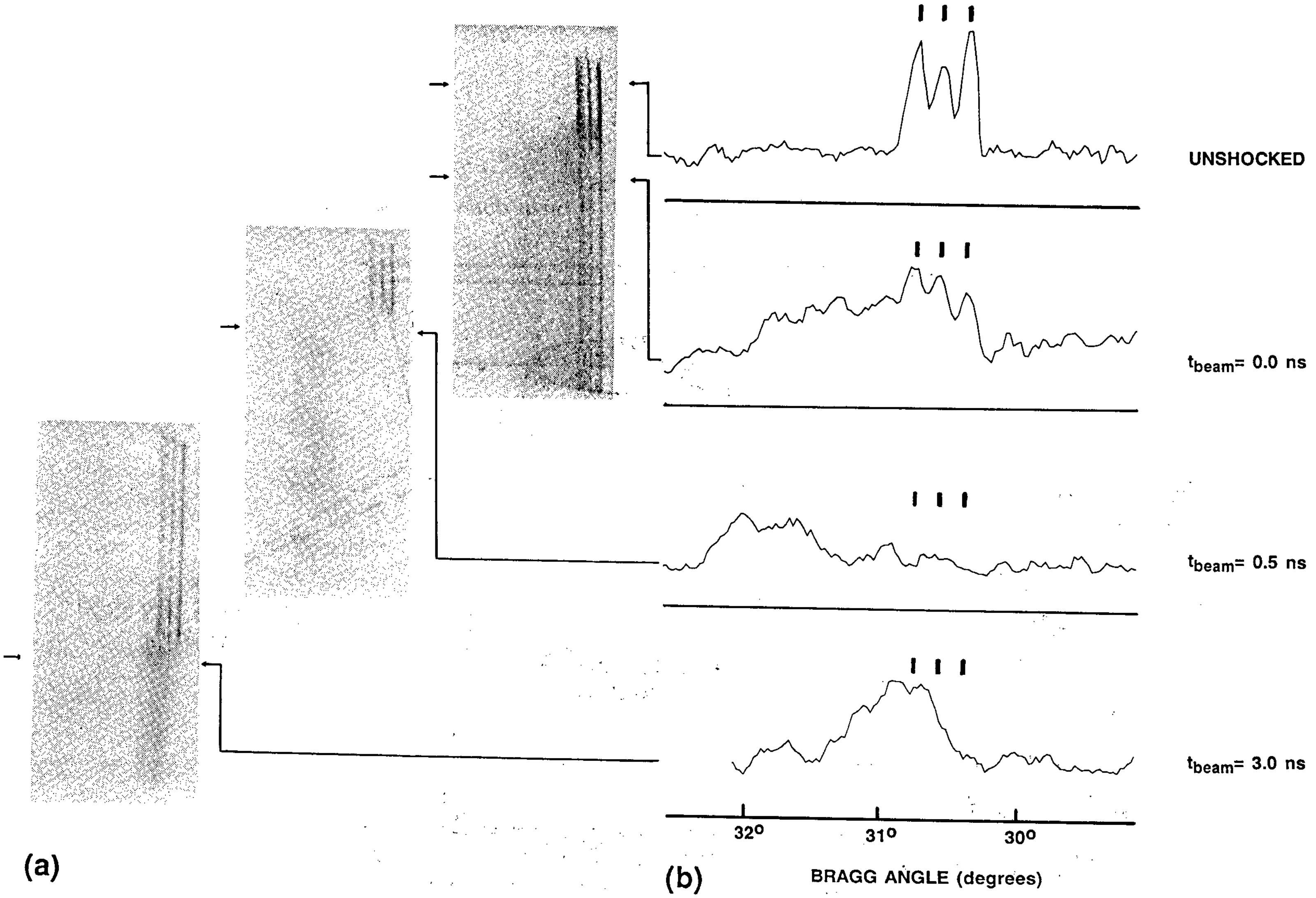

Fig. 2 — Diffraction results. (a) X-ray line spectra diffracted from silicon shocked at an incident energy density of 4 ± 0.3 $J\ cm^{-2}$ (an irradiance of $4 \times 10^9\ W\ cm^{-2}$) are shown adjacent to reference lines simultaneously diffracted by the unstrained silicon lattice. Each photograph represents a separate shot and beam delay. (b) Densitometer scans, taken at indicated locations through the spectra in (a), are shown for a typical unshocked spectrum, and shocked spectra at 0.0 nsec, 0.5 nsec, and 3.0 nsec beam delays.

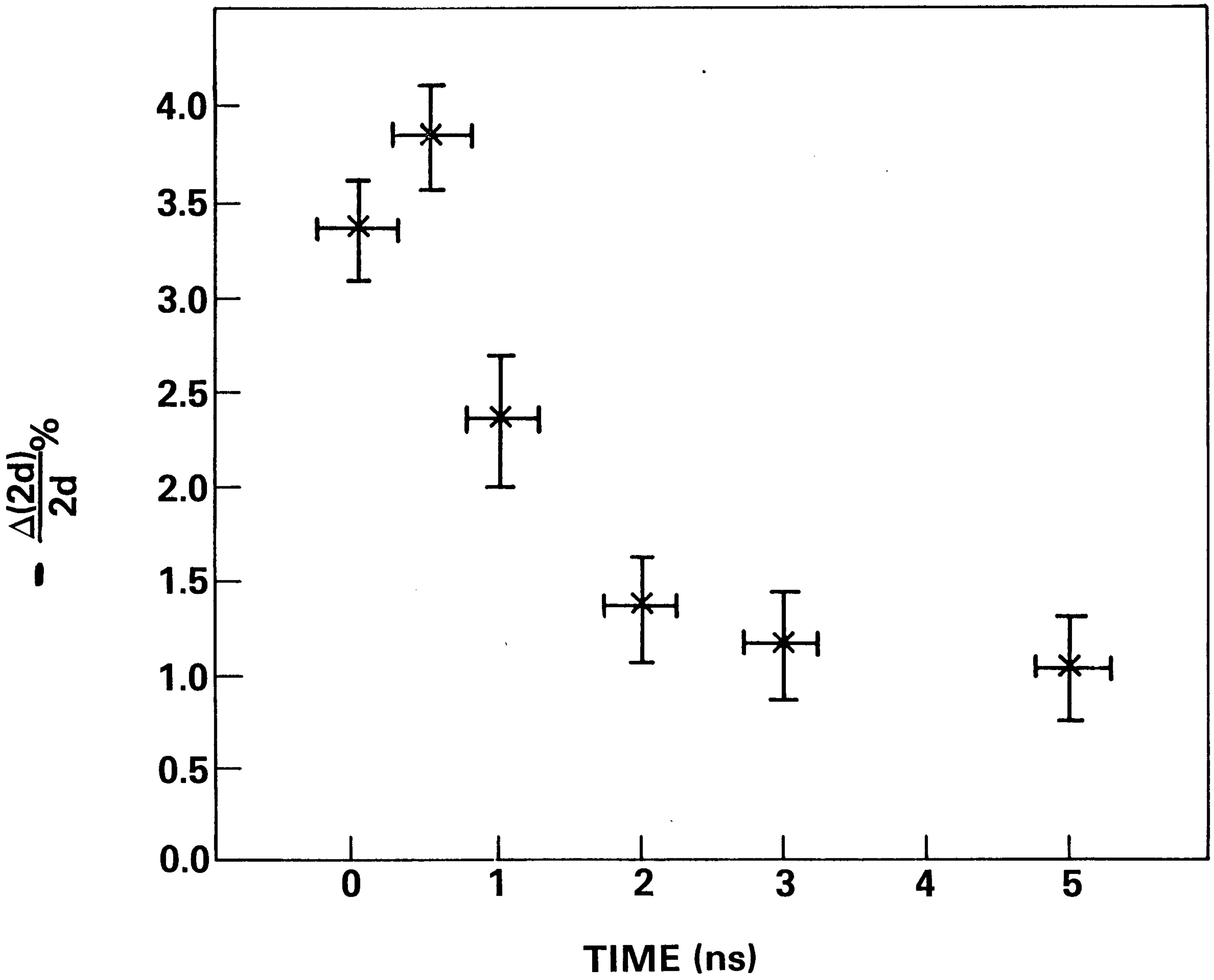

Fig. 3 — The measured peak change in interatomic spacing as a function of time for those laser shots with an energy density of 4 ± 0.3 $J\ \mathrm{cm}^{-2}$

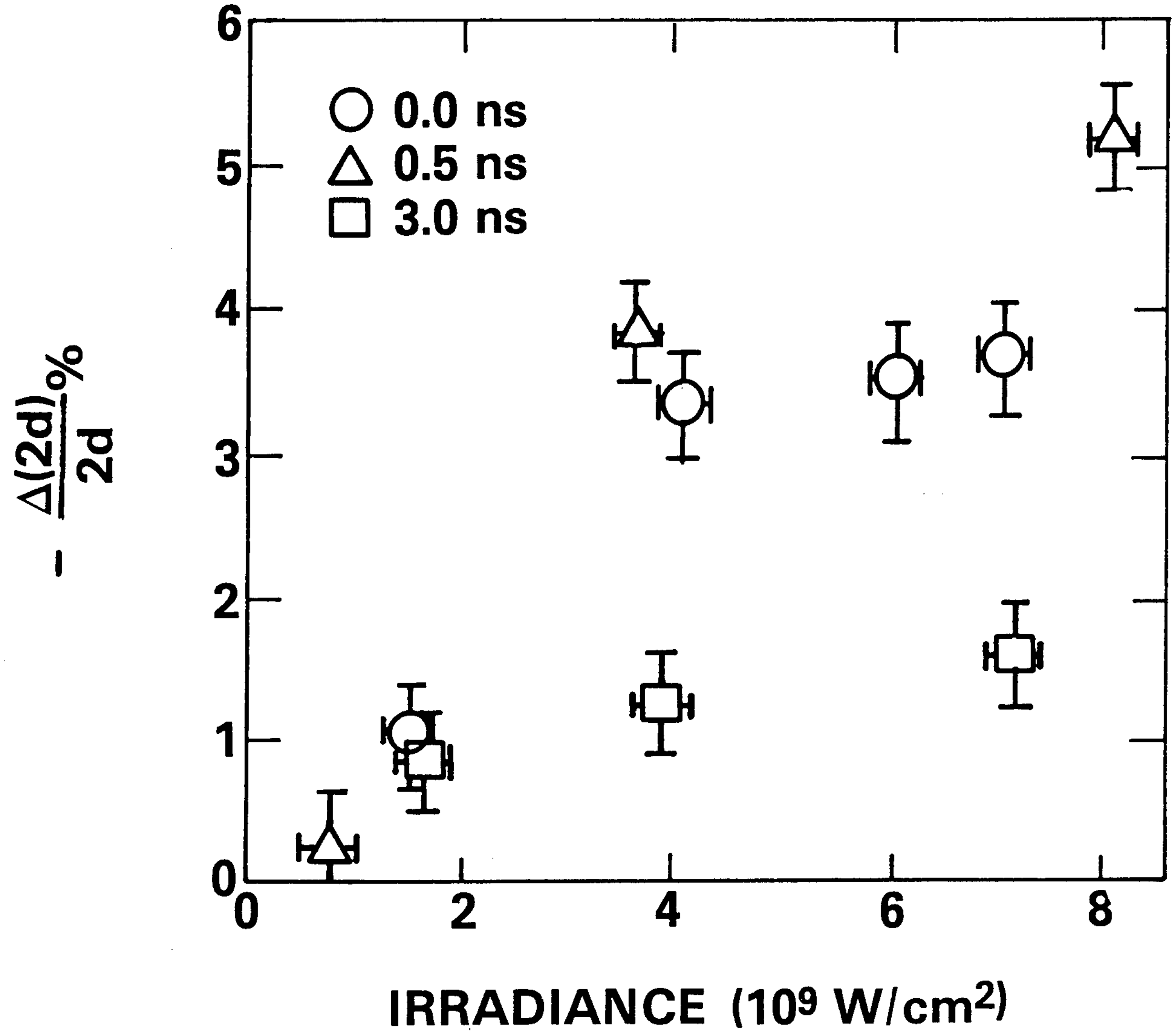

Fig. 4 — The peak change in interatomic spacing as a function of irradiance for delay times indicated. The HEL is at 2.6% compression.