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Pump and probe measurements of shock-compressed states

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Abstract

A pump and probe technique is used for time-resolved measurements of the microstructure of condensed matter under laser shock compression. Two types of experiment (picosecond x-ray diffraction and nanosecond Raman spectroscopy) are performed. The picosecond time-resolved x-ray diffraction results for laser-shocked Si(111) give the time evolution of the strain profiles in 60 ps intervals. Nanosecond time-resolved Raman spectroscopy for laser-shocked poly-tetrafluoroethylene shows transient bond scission of the polymer chain.

1. Introduction

Time-resolved and microscopic measurements of matter at molecular levels are required in order to understand excitation and relaxation processes of matter induced by applying an impulse of high pressure or shock waves. X-ray diffraction and vibrational spectroscopy are well suited for examining changes in crystal lattices and molecules, respectively. However, the signals are usually very weak and hard to detect in a single-shot experiment such as a conventional shock experiment using a powder gun. Using an intense pulsed laser and a pump and probe technique, shock can be generated repeatedly and weak signals can be detected by data accumulation. In the pump and probe experiments, a laser beam is divided into two beams and one is used as a pump beam for shock generation and the other is used as a probe beam. Two probe techniques (picosecond x-ray diffraction and nanosecond Raman spectroscopy) were applied to study shock-compressed states.

2. Picosecond x-ray diffraction

The time-resolved x-ray diffraction study of Si(111) under laser-induced shock compression was performed using the laser pump and x-ray probe technique with a time step of 60 ps. The

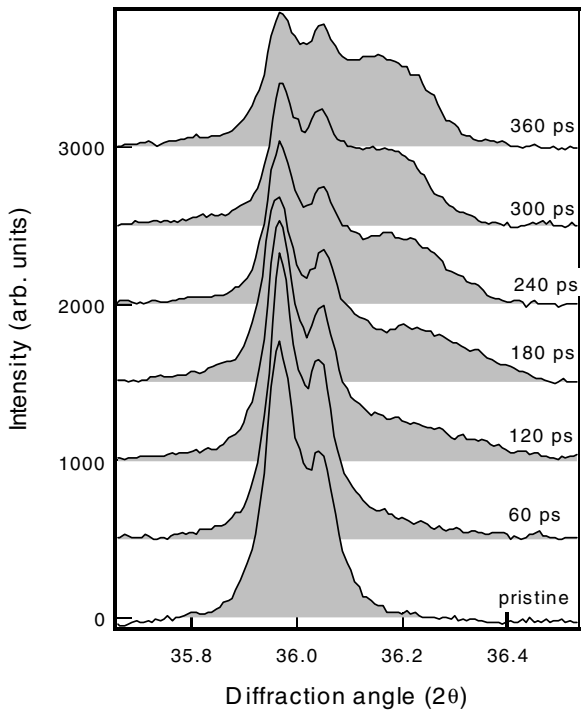

Figure 1. Picosecond time-resolved x-ray diffraction of Si(111) under laser irradiation with 300 ps beams at 4 GW cm^{-2}.

laser used was a T^3 (Table Top Terawatt) laser system, which generates a peak power of 4 TW at maximum at a wavelength of around 780 nm. The 300 ps laser beam was separated by a beam splitter into two beams. One beam was pulse compressed to 50 fs and used for generating x-ray pulses.

The other beam was introduced to an optical delay line and used to irradiate the Si(111) for shock generation. The delay time between the 300 ps beam (pump beam) and the x-ray pulses (probe beam) was controlled by changing the length of the light path. The pulsed x-rays (6 ps) were generated by laser plasma [1], which was formed by focusing femtosecond laser (50 fs) beams onto an Fe target at 10^{17} W cm^{-2}. The shock of the Si(111) was generated by focusing 300 ps laser beams at approximately 4 G cm^{-2}. The Bragg diffraction configuration was used. Diffracted x-rays were detected by an x-ray linear sensor or an x-ray CCD. Time-resolved x-ray diffraction was performed every 60 ps after the 300 ps laser irradiation up to 1380 ps. Figure 1 shows a typical example of picosecond time-resolved x-ray diffraction by laser irradiated Si(111) within 360 ps after the pumping. The rocking curves were obtained by accumulation of 300 laser shots. The virgin sample give both Kα1 (1.9360 Å) and Kα2 (1.9399 Å) diffraction lines. When the pump laser beam was irradiating, the intensities of these lines decreased and the signals at higher angles appeared [2]. The shift and the intensity of the higher angle diffraction gradually decrease after the end of the pump beam irradiation. The shift of diffracted signals towards higher angle corresponds to a decrease in crystal lattice spacing and means that the silicon lattice is compressed upon irradiation with a pump laser beam.

The strain ($\Delta 2d/2d$) is estimated from the equation $\Delta 2d/2d = -\theta \cot\theta$, where d is the lattice spacing and the θ is the Bragg angle. The maximum strain was obtained as

−1.05% (compression) of the lattice spacing and the maximum pressure induced in the present experiment was estimated to be 2.18 GPa. Detailed strain profiles inside the sample can be obtained by analysing the rocking curves with dynamical diffraction theory. Figure 2 shows the strain profiles obtained, which reproduced the observed rocking curves well. In the calculation, the depth (10 μm) was divided into 100 layers. The Takagi–Taupin equation was solved using the one-dimensional layer approximation and the method of Wie *et al* [3]. The evolution of strain profiles inside the Si was obtained without any disturbance such as a material gauge. The decrease of the shift and the intensity of the higher angle diffraction were explained as indicating that the compressed states propagate (shock wave propagation) inside the silicon deeper than the x-ray penetration depth and the compression near the surface is released.

3. Nanosecond Raman spectroscopy

Nanosecond time-resolved Raman spectroscopy has been performed to study polymer films, poly-tetrafluoroethylene (PTFE, $(C_2F_2)_n$), under laser driven shock compression by using a laser pump and laser probe technique. The laser system used was a Q-switched Nd:YAG laser with a maximum output of 3 J/pulse at a wavelength of 1064 nm. The second-harmonic light (532 nm) was generated by using a KD*P crystal. The pulse widths of the fundamental and second-harmonic light are 10 and 8 ns, respectively [4]. In pump and probe measurements, the IR beam is used as a pump beam, which generates a shock wave, and the visible beam is used as a probe beam, which excites the Raman scattering. The delay time between the pump and probe beams can be controlled within 25 ns, since the length of the optical delay line is about 8 m. The target was a glass-confinement target fabricated with a back-up glass substrate, an aluminium foil, a PTFE film sheet, and a cover glass substrate. The peak pressure in the PTFE was estimated to be 2.3 GPa, using an empirical equation for ablation pressure [5] at the laser power density of 4 GW cm^{-2}.

The Raman spectrum was obtained by accumulation of 1000 laser shots. The target was moved for each laser shot in order to expose a pristine surface. The observed lines at 291, 381, and 729 cm^{-1} are assigned to twisting, bending, and symmetric stretching modes of CF_2, respectively. The line at 1379 cm^{-1} is assigned to a C–C stretching mode. Overtone and combination modes are observed at 1215 and 1295 cm^{-1}. Broad lines at 550 and 1100 cm^{-1} are due to the glass substrate.

Figure 3 shows a typical example of the nanosecond time-resolved Raman spectra detected at delay times of 9.3, 14.7, 17.6, and 20.6 ns after the pump beam irradiation. After the shock generation, a new peak appeared at around 1900 cm^{-1} and its intensity increases with increase of the delay time. However, this new peak is not observed in the Raman spectrum of the recovered sample, which is identical to that of the pristine PTFE. The new peak, therefore, is due to the transient species generated under the shock compression. The increase of the new peak can be explained in connection with the propagation of shock waves. The most likely candidate for explaining the new Raman peak at 1900 cm^{-1} is a C = C stretching mode. Since the thermal degradation of the PTFE is of depolymerization type, a C_2F_4 monomer may be formed by bond scission of PTFE under shock compression.

Since Raman spectroscopy probes the whole volume of the PTFE, the observed Raman spectrum is made up by the superposition of the scattering from both volumes under shock compression and in front of the shock wave. As a result of propagation of the shock wave inside the film along with the delay time, the volume under the shock compression increases and the intensity of the new peak increases.

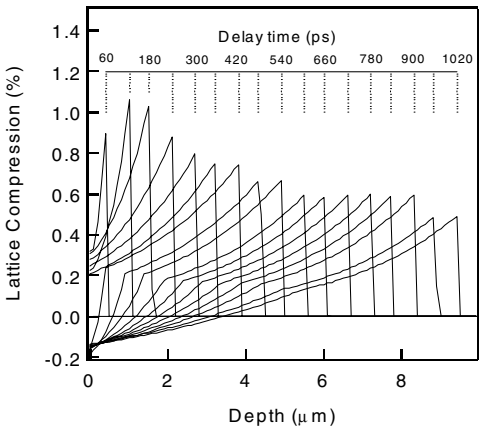

Figure 2. Time evolution of strain profiles inside Si(111) determined from the time-resolved x-ray diffraction with analysis using dynamical theory.

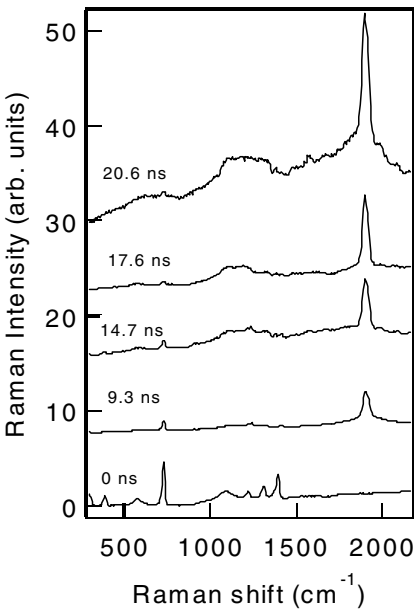

Figure 3. Nanosecond time-resolved Raman spectra of laser-shocked PTFE at 2.3 GPa.

4. Conclusions

A pump and probe technique was applied to measure changes in microstructure of condensed matter under laser shock compression. This technique made it possible to detect weak signals such as Raman scattering and x-ray diffraction by data accumulation. Two types of experiment (picosecond x-ray diffraction and nanosecond Raman spectroscopy) are described. The picosecond time-resolved x-ray diffraction results for laser-shocked Si(111) give the time evolution of the strain profiles at 60 ps intervals. Nanosecond time-resolved Raman spectroscopy for laser-shocked PTFE shows transient bond scission of the polymer chain. The techniques developed open a new research field in the dynamics of mesoscopic structure of condensed matter under shock compression.

Acknowledgments

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