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Time-Resolved Synchrotron X-ray Diffraction on Pulse Laser Heated Iron in Diamond Anvil Cell

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Abstract. We present time-resolved synchrotron x-ray diffraction to probe the ϵ - δ phase transition of iron during pulse-laser heating in a diamond anvil cell. The system utilizes a monochromatic synchrotron x-ray beam, a two-dimensional pixel array x-ray detector and a dual beam, double side laser-heating system. Multiple frames of the diffraction images are obtained in real-time every 22 ms over 500 ms of the entire pulse heating period. The results show the structural evolution of iron phases at 17 GPa, resulting in thermal expansion coefficient $1/V(\Delta V/\Delta T)_p = 7.1 \times 10^{-6} /K$ for ϵ -Fe and $2.4 \times 10^{-5} /K$ for γ -Fe, as well as the evidence for metastability of γ -Fe at low temperatures below the ϵ - γ phase boundary.

1. Introduction

Understanding the dynamic response of solid under extreme conditions of pressure, temperature and strain rate is a fundamental scientific quest and a basic research need in materials science. Specifically, obtaining the atomistic/molecular level description of structural and chemical changes of solid under rapid heating and/or compression over a large temporal, spatial and energy range is critical to understanding material stability or metastability, transition mechanism, crystal order or disorder, and novel physical and chemical properties of solids.

The high brightness of third-generation synchrotron x-rays has revolutionized the field of high-pressure material science and geoscience in recent years. High-pressure synchrotron research activities to date have, however, concentrated largely on probing static properties of materials that are stable near minimum energy configurations and governed by thermodynamic constraints. Only a few studies have acquired dynamic properties of materials such as metastable structures, chemical mechanisms, transition dynamics, and mechanical deformation, much of which require the time-resolved structural information of solid that undergoes rapid phase or chemical changes. In this regard, developing time-resolved x-ray diffraction capabilities is significant for high-pressure sciences and synchrotron technology developments. It is also timely with the proposed development of dynamic compression sector (DCS) at the APS and other similar efforts at the NSLS-II and LCLS.

The machine characteristics of third-generation synchrotron are ideally suitable for probing dynamic single-event phenomena such as shock-induced phase changes and highly exothermic metal

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combustions. As the first step, we have recently demonstrated a novel time-resolved x-ray diffraction capability probing structural and chemical evolutions of metallic solids that undergo rapid phase and chemical changes [1]. Briefly, these experiments were performed using monochromatic synchrotron x-rays at 16IDD beamline of the HPCAT/APS. The system utilized monochromatic synchrotron x-rays and a two-dimensional (2D) pixel array x-ray detector (PILATUS) in combination of a fast-rotating diffraction beam chopper, providing a time (in azimuth) and angle (in distance)-resolved x-ray diffraction image, continuously recorded at a time resolution of $\sim 30 \mu\text{s}$ over a time period of 3 ms, as shown in Fig. 1. Multiple frames of the TARXD images were obtained with time resolutions between 30 and 300 μs over three to several hundreds of ms, depending on the dynamics of experiments in interest. This method is applicable to a wide range of dynamic experiments to study both single event phenomena of solid under thermal, electric or mechanical impact conditions and non-single event changes using dynamic-DAC and high frequency pulse lasers.

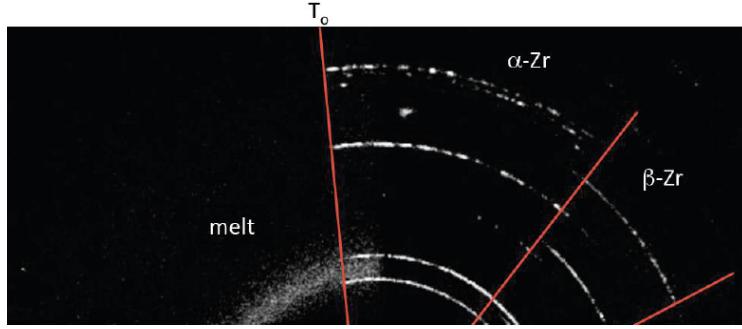


Figure 1. Time- and angle-resolved x-ray diffraction of pulse-heated Zr, showing the early time structural phase transition and melting, followed by the late time highly exothermic combustion of the melt (not shown). This 2D diffraction pattern was recorded for $\sim 30 \mu\text{s}$ x-ray exposure over ~ 3 ms. Time runs clockwise from T_0 - concentric to the Debye-Scherrer's diffraction rings.

The ultimate goal of our efforts is to perform similar time-resolved structural studies at high pressures and temperatures using diamond anvil cells (DAC), dynamic-DAC, and pulse laser-heated DAC. In this paper, we describe a preliminary result showing the time-resolved diffraction change associated with the $\epsilon\text{-}\gamma$ phase transition in pulse laser-heated iron in DAC.

2. Experimental Method

All diffraction experiments were carried out using monochromatic synchrotron x-rays at 14.413 keV (or 0.8602 Å in wavelength) from an undulator at the 16IDD of the Advanced Photon Source (APS). We used a focused x-ray beam of ~ 50 (Horizontal) $\times 30$ (Vertical) μm at the sample.

Figure 2 shows the actual experimental setup for time-resolved x-ray diffraction using laser-heated DAC in the hutch. The system consists of (i) a 2D pixel array x-ray detector (PILATUS), (ii) a dual beam, double side laser-heating system, and (iii) a set of electronics for time synchronization and controls of the experiments, which includes a power supply, a function generator, a delay generator, and a digital scope analyser. The power output of the laser is adjusted to ramp up and down by sending a saw-shape analogue input signal to the laser power supply, as shown in Fig. 3 (the blue curve). The temperature of the sample is measured from thermal emission of laser-heated area during this ramp up/down period [2], while the real-time thermal emission signal is also recorded on a fast photodiode through an optical fibre (Fig. 3, green). Note that the photodiode signal is offset from the laser power about 10-20 ms both at the start and the end. This is due to low thermal emission intensity proportional to the fourth power of temperature, which makes difficult to measure the intensity at low temperatures

at the beginning and ending of pulse heating. For the same reason the measured temperature is actually close to the “maximum” temperature rather than “average” temperature. In this particular experiment, the maximum temperature of ~ 2400 (± 100) K is achieved using about 15% of our laser power. A total of twenty diffraction images were collected during the entire period of one heating and cooling cycle for 500 ms, by acquiring 22 ms x-ray exposure per frame with 3 ms readout (or blockout) time between the frames, as illustrated in Fig. 3, purple. To avoid multiple heating of sample, we gate the laser only for time long enough for one ramp heating cycle (Fig. 3, red).

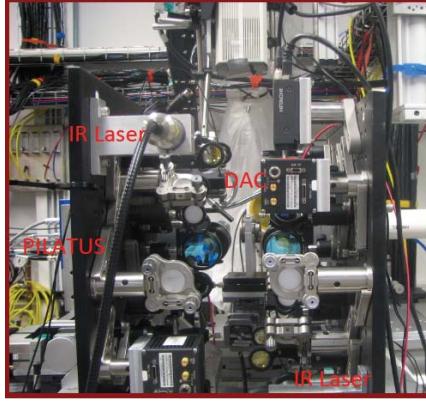


Figure 2. The experimental setup employed for time-resolved x-ray diffraction on double-side, pulse laser-heated iron in DAC at the 16IDD/APS.

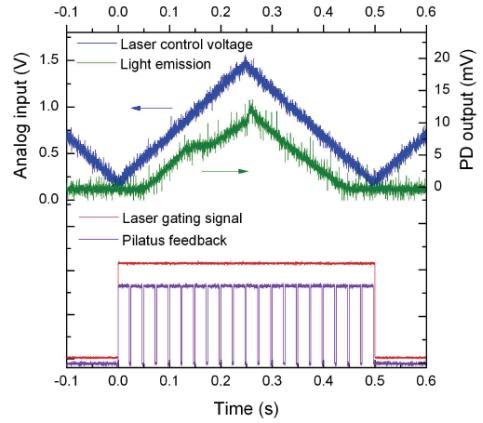


Figure 3. An input signal for pulse laser power, a photodiode output of thermal emission, and a PILATUS feedback of x-ray records, illustrating the time-synchronization of the experiments.

3. Results

In this paper, we describe an experiment of iron in Al_2O_3 pressure medium performed at 17 GPa and the temperature ramped up to ~ 2400 K - near the melting temperature of iron [3-5]. Note that the photodiode record in Fig. 3 exhibits a knee at ~ 120 ms after the heating starts, signifying the occurrence of the $\epsilon\text{-}\gamma$ phase transition during heating. Interestingly, such knee is absent during cooling. These results are consistent with the time-resolved diffraction data shown in Figs. 4-6.

Figure 4 shows a selected set of time-resolved diffraction images of pulse laser heated iron at 17 GPa, showing the reflections of ϵ (hcp)-Fe (white dots), γ (fcc)-Fe (green dots), and Al_2O_3 (orange dots) used as a pressure medium. The quality of the diffraction images is rather poor, largely due to a short x-ray exposure time for each image (22 ms) and, more importantly, high level of the structureless background x-ray intensity from diamonds and strong diffraction lines from Al_2O_3 . Nevertheless, it does depict the evidence of the $\epsilon\text{-}\gamma$ phase transition that occurs at $\sim 100\text{-}120$ ms after the initial pulse heating, as mentioned in Fig. 3. Note that the diffraction lines of Al_2O_3 appear relatively strong with respect to those of iron. This is likely due to the low energy x-ray (15 keV) used in this experiment, which also limits the time-resolution of the present experiment. In fact, we found that it was possible to obtain a 1 ms time-resolution using nitrogen pressure medium.

By integrating the time-resolved diffraction images in Fig. 4, we obtain the time- and angle-resolved diffraction patterns as shown in Fig. 5. Again, the signal-to-noise ratio of the spectra is not good, but sufficient to discern three diffraction lines from ϵ -Fe (010, 002 and 101, marked in blue), two lines from γ -Fe (111 and 002 in green), and four lines from Al_2O_3 (014, 110, 113, and 024, marked in orange). Note that the 002 line of ϵ -Fe shows relatively strong intensity in comparison with the

100, probably representing a preferred orientation of the sample. The γ -phase appears at \sim 100 ms during heating and then remains over the entire cooling period, while the ϵ -phase disappears during heating and reappears during cooling. This indicates the metastability of γ -Fe at low temperatures well below the γ/ϵ phase boundary at this pressure, confirming the photodiode record in Fig. 3. A similar metastable behaviour of γ -Fe was often observed in the previous laser-heating experiments [3-5]. The diffraction data also shows subtle changes in the peak positions of diffraction lines with time, apparently a signature for thermal expansion (or contraction). The thermal expansion of ϵ and γ -phases can be seen more clearly in Fig. 6.

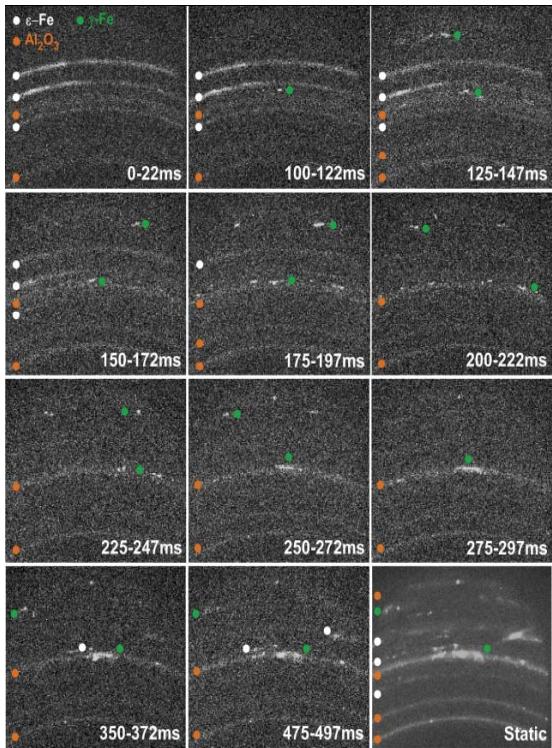


Figure 4. A selected set of time-resolved diffraction images of pulse laser heated iron at 17 GPa, showing the ϵ (hcp, white dots)- γ (fcc, green dots) phase transition together with the features from Al_2O_3 (orange dots).

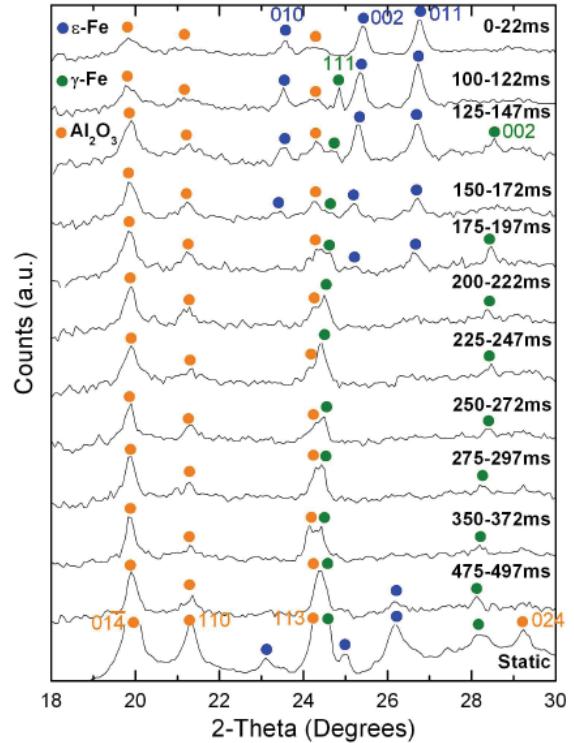


Figure 5. Time- and angle-resolved x-ray diffraction patterns of iron at 17 GPa, showing the ϵ - γ phase transition during pulse laser heating. The blue, green, orange dots signify the diffraction lines of ϵ -Fe, γ -Fe and Al_2O_3 , respectively.

Figure 6 plots the time (or temperature)-dependent volume changes of the ϵ and γ - phases, obtained from the data in Fig. 5. Again, the plot depicts the coexistence of two ϵ and γ phases between 120 ms and 200 ms. The ϵ - γ phase transition accompanies about \sim 3% volume expansion occurring abruptly at the transition, 120 ms. Based on the measured “maximum” temperature 2400 K and the ϵ - γ transition temperature of 1000 K at 17 GPa, we estimate the thermal expansion coefficient $1/V(\Delta V/\Delta T)_p = 7.1 \times 10^{-6} / \text{K}$ for ϵ -Fe and $2.4 \times 10^{-5} / \text{K}$ for γ -Fe at 17 GPa. These values are compared with those measured at ambient pressure, $6.5 \times 10^{-5} / \text{K}$ for γ -Fe and $3.3 \times 10^{-5} / \text{K}$ for α (bcc)-Fe [6]. Based on these data, we find the decrease of thermal expansion coefficient for γ -Fe with increasing pressure in accordance with $\{\delta(\ln \alpha)/\delta(\ln V)\}_T = 6.1$, which agrees well with the previously reported value of 6.5 (± 0.5) determined by a wire-heating technique in DAC [7].

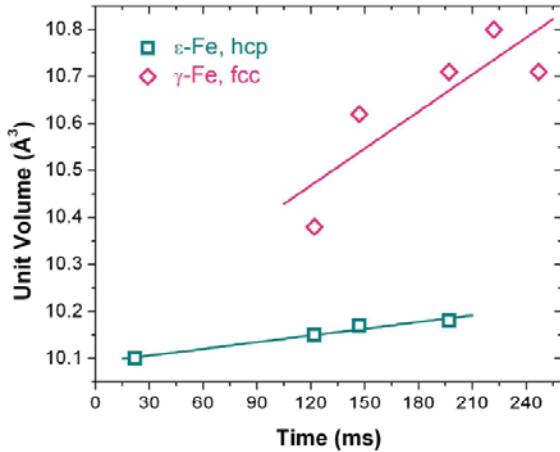


Figure 6. Time (or temperature)-dependent volume changes of the ϵ (hcp) and γ (fcc) phases, obtained from the data in Fig. 5. The phase transition occurs at around 120 ms – consistent with the knee observed in the PD record in Fig. 3.

4. Conclusion

We have shown the feasibility of time-resolved x-ray diffraction applied to pulse laser-heated samples in DAC. The results performed on Fe at 17 GPa show real-time evidences for the ϵ - γ phase transition in time-resolved thermal emission (or temperature) and diffraction records. Based on the diffraction data, we obtain the thermal expansion coefficient of $1/V(\Delta V/\Delta T)_p = 7.1 \times 10^{-6} / \text{K}$ for ϵ -Fe and $2.4 \times 10^{-5} / \text{K}$ for γ -Fe at 17 GPa.

The present method employs a high-resolution 2D PILATUS detector, providing a convenient way of obtaining time-resolved powder x-ray diffraction images in a ms resolution, or in a μs time resolution when it is used in combination with a fast recording chopper as shown in Fig. 1 [1], with high sensitivity nearly comparable to an intensified CCD but substantially lower noise. The time resolution of the present experiment can be enhanced to 10-100 μs permitting a signal averaging over a reasonable experimental period of ~ 5 min. The spatial-resolution can also be increased by use of a larger detector such as PILATUS 300K or 2M of which active area can be 10-100 times the present one. The main limitation, on the other hand, is relatively low x-ray energy used in the present experiments. This is because the quantum efficiency of PILATUS drops rapidly at high x-ray energies, from 99% at 12 keV to 55 % at 15 keV or $\sim 20\%$ at 20 keV, which limits the use of thick diamond anvils and pressure medium.

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