Laser induced shock studies at RRCAT, Indore

P.A. Naik1, V. Arora, S. Bagchi, Y.B.S.R. Prasad, S. Barnwal, and P.D. Gupta
Laser Plasma Division, Raja Ramanna Centre for Advanced Technology, Indore 452 013, M.P., India
E-mail: panaik@rrcat.gov.in

Abstract. The knowledge of material response under shock compression is important in terms of proper understanding of the process and in view of the wide range of technological applications associated with it. At the Laser Plasma Division, RRCAT, Indore, among other things, we are also involved in studying the material response from two different perspectives. On one front, we have developed capability for acquiring Equation of State (EOS) of materials using laser generated shock waves with long pulse lasers (>200 ps). On the other front, we have started shock experiments on single crystals using time resolved x-ray diffraction technique, using ultra-short pulse lasers (<1 ps). In these initial experiments, characteristic sub-picosecond, high brightness Kα x-ray pulses generated by Ti: sapphire femtosecond laser produced plasmas of different target materials (Ti, Fe, and Cu) were used to probe the effect of compression generated by moderately intense (GW/cm2) sub-nanosecond laser (200 ps) pulse irradiation on the crystal surface. The dynamics of the shock propagation is manifested in terms of the temporal evolution of the rocking curve of shocked sample. The shock velocity deduced from these measurements is 12 km/s, consistent with the predicted velocities and probe depth. The observed maximum compression is 0.4 % which corresponds to a pressure of 0.8 GPa.

1. Introduction

The study of material response under extreme conditions is of great importance in several fields, in particular material science, astrophysics, geophysics, and research of target design in the context of inertial confinement fusion (ICF) studies [1,2,3]. The equation of state (EOS) data is necessary for hydrodynamic simulations dealing with nuclear fusion [4,5]. In the recent years, shock compression of condensed matter has emerged as a subject of considerable importance for a variety of research investigations such as phase transitions [6] (both structural and magnetic), strain propagation [7], etc. The simple technique of x-ray diffraction offers great potential in terms of direct observation of lattice distortion because of the inherently large penetration depth of x-rays in matter. Time resolved x-ray diffraction thus enables real time visualization of atomic arrangement in the sub ps time scale [8]. The x-ray bursts from sub-picosecond laser produced plasmas have been utilized for time-resolved study of processes like ultrafast excitation of the crystal lattice [9], non-thermal melting of semiconductors [10], acoustical phonon excitations [11]. In particular, the characteristic K-shell line

1 To whom any correspondence should be addressed.
radiation ($K_\alpha$) has generated much interest to be used as a narrow bandwidth and ultrafast probe to study fast processes in matter. Depending on the interaction parameters, the hot electrons are generated through collective mechanisms such as resonance absorption [12], vacuum heating, j x B heating etc. These electrons penetrate into the cold target material to generate continuum hard x-ray bremsstrahlung and characteristic $K_\alpha$ line radiation. The photon flux time, time duration, and monochromaticity of the $K_\alpha$ emission is primarily determined by the hot electrons parameters such as the incident number, energy and transport into the bulk solid [13].

Laser Plasma Division at RRCAT, Indore, is engaged in research related to shock physics in two areas. One area is EOS studies at megabar pressures using high power lasers and the other area is time resolved x-ray diffraction using ultra-short pulse lasers. The latter studies are at GPa (tens of kilobar) pressures. The article is organised as follows. Section 2 gives the experimental description and the results of the EOS studies. Section 3 gives the experimental description and the results of the time resolved x-ray diffraction studies. Our future plans in both these areas are presented in section 4.

2a. EOS experiments with high power lasers

EOS at pressures up to a few Mbar can be measured in static experiments using diamond anvil/gas gun. For higher pressures (>10 Mbar) the laser-induced shock wave technique is useful. Laser produced plasmas had been used to generate pressures up to 100 Mbar in different materials to construct their EOS [14,15]. The shock pressure can be varied in a controllable way from less than 10 kbar up to ~800 Mbar, by using lasers with different energy, time duration, and focal spot. The strain rates achievable in laser-produced shock waves vary from about $10^4$ to $10^9$ s$^{-1}$. An additional important advantage of laser-induced shock wave is the possibility to synchronize the shock with a variety of optical, x-ray, and electronic diagnostics. EOS of copper up to 20 Mbar, gold and lead up to 10 Mbar, Parylene-C and brominated CH at pressures up to 8 Mbar, have been made using the HELEN laser at AWE, UK [16]. Absolute measurements of the EOS of iron at pressures in the range of 1–8 Mbar have been made using the LULI Laser at Ecole Polytechnique, France [17]. EOS of dielectric materials like sapphire (Al$_2$O$_3$) and lithium fluoride (LiF) up to 20 Mbar was measured using the Omega laser at Univ. Rochester, USA [18]. The Hugoniot of tantalum up to pressures of 40 Mbar was measured with the Gekko/Hyper laser at Institute of Laser Engineering, Japan [19]. EOS of low density foams in the range of 60–130 mg/cm$^3$ up to pressures of 3.6 Mbar was measured with the PALS laser at Prague [20]. In general, determination of EOS involves measurement of the shock and particle velocities in the medium. In these experiments, an intense laser pulse is focused on a foil (whose EOS is to be determined) kept in vacuum. The ablating plasma launches a strong shock wave in the foil material due to momentum recoil of the ablating material. During the duration of the laser pulse, a continuous shock wave is launched in the inward direction from the ablating target surface. If the thickness of the foil is such that the shock transit time is less than the laser pulse duration, there is no shock rarefaction. The shock wave travels with a velocity $u_s$ and particles behind the shock wave attain a velocity $u_p$. If the laser spot size on the foil is much more than the foil thickness, one can assume 1-D shock propagation. The various material parameters in the shocked region like density ($\rho_1$), shock pressure ($P_1$), and kinetic energy of the compressed material ($E_1$) are related to the initial material properties, the shock velocity ($u_s$), and the particle velocity ($u_p$) by the standard Hugoniot relations $\rho_1u_1^2=\rho_0(u_s-u_0)^2$, $\rho_0u_0=\rho_1u_s=P_1-P_0$, $\rho_0\mu_0\{E_1-E_0+\frac{1}{2}u_0^2\}=P_1u_p$. The shock velocity is experimentally measured from the time it takes for the shock wave to reach the rear side of the foil. This, in turn, is measured from the shock luminosity due to compression. These propagation times are usually on ps time scale and hence fast optical streak cameras are required to record the luminosity variation. The particle velocity is determined experimentally using optical techniques like Velocity Interferometer System for Any Reflector (VISAR) or Optically Recording Velocity Interferometer System (ORVIS). These instruments give the free surface velocity of the rear-side of the foil. The particle velocity is generally equal to or slightly less than half the free surface velocity. In absence of instruments like VISAR, it is still possible to obtain the particle velocity independently by using optical shadowgraphy techniques where two pulses with known delay are used for probing the plasma from the tangential
direction and the particle velocity can be measured \cite{21,22}. In addition, it is possible to use chirped pulse diagnostics like chirped pulse reflection \cite{23}, frequency domain interferometry \cite{24} etc., to obtain the shock breakout time and particle velocity more accurately. When it is not possible to determine the particle velocity experimentally, one uses the standard relation \( u_s = u_o + \alpha u_p \) to get the particle velocity from the shock velocity, when \( u_o \) and \( \alpha \) are known for the material \cite{25}.

2b. Experimental Setup and Results

The initial experiments at RRCAT were conducted on a 200 ps Nd:YAG commercial laser system at laser energy of 2 J (max) to obtain the EOS of gold by using Al+Au and Al+Cu layered targets using the impedance matching method \cite{26}. At laser intensities \( \sim 3 \times 10^{13} \) W/cm\(^2\), shock pressures in the range 4.5 to 6.5 Mbar for Al, 9-13 Mbar for Au, and 9-10.5 Mbar Cu were obtained \cite{27}. In order to extend these studies to higher pressures, experiments were then carried out using an Nd: glass laser system at laser energy 10 J with pulse duration of 600 ps, with Al as reference material. The laser output was converted to second harmonic (532 nm) to enhance the temporal contrast. The laser energy was focused on 8 \( \mu \)m Al foils at an intensity of \( 10^{13} \) W/cm\(^2\). An (S-20) optical streak camera with a time resolution of 8 ps was used for these measurements. In order to have a time reference or fiducial, a portion of the laser light was collected at an earlier stage, converted into second harmonic using a KDP crystal, and then passed through an optical fiber up to the entrance slit of the streak camera. The length of the fibre was adjusted such that the time of laser light reaching the end of the fibre is exactly equal to that of the laser light reaching the streak camera (in absence of the foil). The shock velocity was estimated by measuring the shock propagation time defined by the time delay between fiducial and the shock arrival at the rear surface of the foil (figure 1). The streak camera was triggered in advance by the laser pulse using a photodiode to compensate for the inherent time delay of the streak camera circuitry. A shock velocity of 21 km/s was calculated from the measured transit time (figure 2). The corresponding particle velocities are 12 km/s, as estimated from the LASL data-book \cite{25} which correlates the shock velocity and particle velocity by the relation \( u_s = 5.38 + 1.34u_p \). The shock pressure was calculated to be 7 Mbar from the standard relationship \( P = \rho \rho_s u_s^2 \), where \( \rho_o \) is the solid density of aluminium (2.7 gm/cm\(^3\)).

3a. Time Resolved X-ray Diffraction Experiments with Ultra-short Pulse Lasers

EOS studies require long pulse (>200 ps) lasers whereas time resolved x-ray diffraction (TXRD) needs ultra-short lasers (< 1 ps). TXRD is used to probe the lattice strains on sub-picosecond time-scales under extreme conditions of temperature and pressure. The diffraction pattern shifts have been used to determine the average lattice compression following elastic–plastic deformation. The appearance of new diffraction lines has been used to examine solid–solid phase transitions. VANISHING
of the reflected Bragg peak can give information about solid-liquid transition. Although 3rd and 4th generation synchrotron sources can be used for this purpose, characteristic x-rays produced with ultrashort lasers offers a cheaper and compact alternative. The TXRD technique was first demonstrated with a shocked silicon (111) crystal by Kalantar et al. [28] using Nova laser. bcc to hcp phase transition in Fe at 13 GPa shock produced by laser irradiation at $2 \times 10^{10}$ to $1 \times 10^{12}$ W/cm$^2$ (2 – 6 ns) was probed with 1.85 Å iron K-shell x-rays by Hawreliak et al. [8]. X-ray diffraction was used with VULCAN laser to measure shear strains in single crystals of copper shocked to pressures in excess of 100 GPa (1 Mbar) by Murphy et al [29].

In a typical TXRD setup, a major part of the laser beam energy is focused on to a solid target to generate an efficient high brightness, sub-picosecond x-ray probe beam. The line radiation emitted is used as monochromatic x-ray source for probing the transient structural modifications in the crystalline sample induced by the remaining fraction of the laser beam referred as pump laser. The rocking curves of sample are recorded as a function of diffraction angles for different time delays between pump and probe pulse. Commonly, the experiment is performed in Bragg geometry. It is well known that in the case of Bragg reflection, a large number of layers participate in the process and the diffraction pattern is cumulative contribution of all the participating layers. From Bragg’s law, the angle of diffraction is related to the change in inter-planar spacing. The lattice strain can be estimated using $\Delta d/d = - \cot \theta_B$ where $d$ is inter-planar spacing, $\theta_B$ is Bragg angle. In such experiments, dynamics of the lattice deformation is studied from the shift / broadening of the diffracted x-ray peak and the change in reflectivity. In addition to this, exploiting the fact that different x-ray photon energies have different penetration depth (defined as 1/e attenuation length) in the material, keeping the external excitation source parameters invariant, it is possible to infer about the characteristics of shock wave propagation inside the optically opaque samples.

3b. Experimental Setup and Results

Experiments were conducted using the 10 TW Ti: Sapphire laser system. The schematic of experimental setup is shown in figure 3 (a). The $K_\alpha$ x-ray probe was generated by focussing the 45 fs laser pulses onto solid (titanium, iron and copper) targets. A part of the uncompressed (200 ps) pulse was used to irradiate a 500 µm thick flat Si (111) crystal ($d = 6.271$ Å) at an intensity of 6 GW /cm$^2$. The time delay between the 200 ps pump laser pulse and the probing x-ray pulse was adjusted by an optical delay line. A positive delay here means the pump laser pulse is leading the probe x-ray pulse. The diffracted x-ray spectrum was recorded on an x-ray CCD camera. The point x-ray source allowed a direct imaging of a part of the crystal surface on to the x-ray CCD camera. This enabled simultaneous recording of the diffracted x-ray spectrum from the laser irradiated as well as pristine area of the crystal. Figure 3 (b) shows the space resolved CCD image of the diffracted iron $K_{\alpha}$ x-rays from laser irradiated Si (111) surface at a delay of +600 ps. The lower part of the same picture shows the pristine sample where the $K_\alpha$ lines ($K_{\alpha 1}$ and $K_{\alpha 2}$) are clearly identified. The upper part of the picture shows blurring of the $K_{\alpha}$ lines because of the non-uniform lattice compression attributed to cumulative effects of the laser induced compression wave and the associated thermal broadening of the lattice.
The measured rocking curves of Si (111) irradiated by 200 ps laser pulse at a fluence of 2.3 J-cm\(^{-2}\) for various delay times between -300 and +1800 ps are shown in figure 4. The x-ray spectrum shows that the diffraction profiles of Fe K\(\alpha_1\) (6403.8 eV) and K\(\alpha_2\) (6390.8 eV) are well resolved for zero and negative delays. It is observed that the diffraction pattern broadens with increasing time delay up to +1200 ps. After that, the broadening reduces and finally comes back to original state for delays larger than +1500 ps. The x-ray beam is diffracted from the sample at a particular angle within a thin layer of the crystal where the lattice spacing is such that the Bragg condition is satisfied. Different layers strained to different extents under the influence of the compression wave, will diffract at other angles leading to the broadening. Furthermore, at early times, the diffraction will occur from both shocked material and the underlying unperturbed crystal. It may be noted that the broadening of the diffracted signals towards higher angles implies lattice compression induced by pump laser beam. The compression of lattice is either due to the laser ablation of very thin surface of the silicon or stress caused at the front of thermal expansion due to the surface energy deposition [30]. On the other hand, the spread towards lower angles reveals the signature of thermal disordering effect indicating the role of the thermal wave. Nevertheless, the time evolution of the rocking curve signifies the propagation of laser induced shock waves inside the crystal. At later times, the separation of K\(\alpha\) peak signifies the passing of shock waves beyond the maximum probe depth inside the crystal.

The rocking curve of silicon irradiated at same laser parameters (200 ps, 2.3 J-cm\(^{-2}\)) was also measured with Ti K\(\alpha\) (4.5 keV, Bragg angle: 26\(^\circ\)) and Cu K\(\alpha\) (8.05 keV, Bragg angle: 14.2\(^\circ\)). The profiles of the rocking curves are similar to that measured with Fe K\(\alpha\) (4.5 keV, Bragg angle: 18\(^\circ\)). Figure 5 shows the FWHM of K\(\alpha_1\) line radiation as function of delay between pump and the probe pulse for Ti, Fe and Cu. It is observed that the diffraction pattern broadens with increasing delay to reach a maximum and thereafter the K\(\alpha_1\) width decreases and come close to the pristine. The time for the maximum broadening is 650 ps, 1160 ps, and 1870 ps for Ti, Fe, and Cu respectively. FWHM of the rocking curve of irradiated sample shows broadening of 3.2 ± 0.3 times compared to the rocking curves of pristine sample for all the three probe x-ray lines. The high energy x-rays have longer penetration depth and are therefore expected to yield information about the strain over a greater crystal depth.

A quantitative knowledge of the velocity of propagating compression front (shock wave) requires code based on the dynamical diffraction theory applied to the strained and laser-shocked crystal.
Nevertheless, some important features can be realized by approximating the delay time for maximum broadening occurs when shock arrived at depth equals to penetration depth of x-ray probe. The penetration depths for Ti, Fe, and Cu Kα for their respective incident angle are 2.7, 2.9, and 8.3 µm respectively. The mean propagation speed of the laser induce compression wave, deduced from the maximum broadening time of the three different x-ray probe is 12 km/s which is very close to the sound velocity of 9.4 km/s in Si. However, the reduction in intensity of x-ray beam in passage through sample has to be taken into account. Nevertheless, the derived shock velocity values are in broad agreement with those obtained under similar irradiation conditions. Maximum lattice strain was estimated from the shift (up to which diffraction signal appears) of 0.074° which corresponds to a compression of 0.4 %. Hironaka et al. [31] observed a maximum lattice strain of 1.05 % corresponding to the maximum pressure of 2.18 GPa, whereas the maximum lattice strain of 0.4 % corresponds to 0.83 GPa (8.3 kbar).

4. Future Plans

On the EOS front, the particle velocity is being estimated at present indirectly from the relation \( u_s = u_0 + \alpha u_p \) by assuming coefficients \( u_0 \) and \( \alpha \) obtained from literature. At present, the laser system is being upgraded to deliver laser pulses with energies up to 200 J in 400 ps at second harmonic (0.5 TW). After a year or so, we will be in position to carry out EOS studies at high energies and intensities. In the time resolved diffraction studies, in future, we have plans to study time resolved x-ray diffraction on shorter time scales (tens of fs). We are involving more and more material science groups (UGC-DAE-CSR, Indore, Materials and Advanced Accelerator Sciences Division of RRCAT, Laser Materials and Device Development Division, RRCAT) for experiments where the time resolved x-ray diffraction technique could be used for carrying out interesting investigations. Isochoric heating, i.e. heating the matter at solid density (10^{22} \text{ cm}^{-3}) to a very high temperature (1 eV to 1keV), in a very short time (< 1 ps) so that the heating is without change in volume, is one of the least explored areas relevant to structures of planetary and stellar interiors as well as inertial confinement fusion (also called “Warm Dense Matter”). The fast heating can be achieved using 1) x-rays, 2) hot electrons, or 3) protons generated by ultrashort, ultrahigh intensity lasers. With the 150 TW laser to be installed in our laboratory by Feb. 2012, and with our indigenous 50 TW (25 J in 500 fs) under construction, we should soon be in position to start experiments in this new exciting area of research.

References

[1] Celliers P M, Collins G W, Da Silva L B, Gold D M, Cauble R, Wallace R J, Foord M E and Hammel B A, 2000 Phys. Rev. Lett. 84 5564
[2] Remington B A, Drake R P, Takabe H and Arnett D, 2000 Phys. Plasmas. 7 1641
[3] Benuzzi A et. al., 2006 Plasma Phys. & Control. Fusion 48 B347
[4] Godwal B K, Sikka S K, and Chidambaram R, 1981 Phys. Rev. Lett. 47, 1144
[5] Cauble R et al., 1998 Phys. Rev. Lett. 80 1248
[6] Lübecke A et. al., 2010 New J. Phys. 12 83043
[7] Nicoul M, Shymanovich U, Tarasevitch A, Linde D.V, and Sokolowski-Tinten K 2011 Appl. Phys. Lett. 98 191902
[8] Hawreliak J, Kalantar D H , Stölken J S, Remington B A and Lorenzana H E 2008 Phys. Rev. B 78 220101R
[9] Sokolowski-Tinten K, Blome Ch, Dietrich C, Tarasevitch A, von der Linde D, Horn-von Hoegen M, Cavalleri A, Squier J A and Kammler M 2001 Phys. Rev. Lett. 87 225701
[10] Rousse A et. al., Nature 410, 65 (2001)
[11] Uschmann I, Kämpfer T, Zamponi F, Lübecke A. Zastrau U, Loetzsch R, Höfer S, Morak A, and Förster E 2009 Appl. Phys. A 96, 91
[12] Krueer W L 2003 The physics of laser–plasma interactions (Westview, New York)
[13] Reich Ch, Gibbon P, Uschmann I and Förster E 2000 Phys. Rev. Lett. 84 4846
[14] Batani D et. al., 2004 Phys. Rev. Lett. 92 065503.
[15] Batani D et. al., 2002 Phys. Rev. Lett. 88 235502.
[16] Rothman S D, Evans A M, Horsfield C J, Graham P and Thomas B R, 2002 Phys. Plasmas 9 1721.
[17] Benuzzi-Mounaix A, 2002 Phys. Plasmas 9 2466.
[18] Hicks D G, Celliers P M, Collins G W, Eggert J H and Moon S J, 2003 Phys. Rev. Lett. 91 035502
[19] Ozaki N et. al., 2004 Phys. Plasmas 11 1600
[20] Dezulian R et. al., 2006 Phys. Rev. E 73 047401.
[21] Chourasia S, Leshma P, Tripathi S, Murali C G, Munda D S, Sharma S M, Kailas S, Gupta N K and Dhareshwar L J 2010 BARC Newsletter 317 13
[22] Peter G and Janez M, 2011 Optics Letters 36 2782
[23] Chen J P, Li R X, Zeng Z N, Wang X T, Wang W Y, Jiang Y H, Cheng C F and Xu Z Z, 2003 J. App. Phys. 94 858
[24] Benuzzi-Mounax A, Koenig M, Boudenne J M, Hall T A, Batani D, Scianitti F, Masini A and Di Santo D 1999 Phys. Rev. E 60 R2488
[25] Marsh S P 1980 LASL Shock Hugoniot Data (Berkeley, CA, Univ. of California Press)
[26] Zeldovich Ya B and Raizer Yu P, 1967 Physics of Shock Waves and High Temperature Hydrodynamic Phenomena (Academic Press, New York)
[27] Shukla M, Upadhyay A, Senecha V K, Khare P, Bandyaopadhyay S, Rai V N, Navathe C P, Pant H C, Khan M and Godwal B K 2003 Laser & Part. Beams 21 615
[28] Kalantar D H et. al., 1999 Rev. Sci. Instrum. 70 629
[29] Murphy W J et. al., 2010 J. Phys.: Cond. Matter 22, 065404
[30] Kishimura H, Morishita H, Okano Y H, Hironaka Y, Kondo K, Nakamura K G and Atou T 2006 Phys. Rev. B 74 224301
[31] Hironaka Y, Yazaki A, Saito F, Nakamura K.G, Kondo K, Takenaka H and Yoshida M 2000 Appl. Phys. Lett. 77 1967