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APPLICATION OF HIGH-POWER LASERS TO EQUATION-OF-STATE RESEARCH AT ULTRAHIGH PRESSURES

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APPLICATION OF HIGH-POWER LASERS TO EQUATION-OF-STATE RESEARCH AT ULTRAHIGH PRESSURES

ABSTRACT

The application of high-power pulsed lasers to ultrahigh pressure equation-of-state (EOS) experiments is discussed. It is shown that pressures along the principal Hugoniot between 1 and 10 TPa can be produced with existing lasers used for inertial-confinement fusion research. The relevance of measurements in this pressure regime to improving our understanding of condensed matter physics is also discussed. New experimental techniques as well as potential experimental problems are described, and EOS experiments on the Janus and Argus laser systems are proposed.

INTRODUCTION

The capability of high-power lasers to generate ultrahigh pressure shock waves has been recognized for many years. When a laser pulse is focused onto the surface of a solid target, a partially absorbing plasma is rapidly formed. Laser energy is absorbed in the low-density regions of this expanding plasma (electron density $< 10^{21} \text{ cm}^{-3}$ for Nd-glass-laser irradiation) and transported by electronic thermal conduction into the solid target. Material is blown off of the target into the lower density plasma, and this ablation drives a shock wave into the target ahead of the ablating surface. As irradiation continues, the critical surface, where the local plasma frequency equals the laser frequency, moves away from the initial target surface, while the ablation front moves into the target at a velocity less than the shock velocity. Upon termination of the laser pulse, pressure drops rapidly at the ablation surface, and a rarefaction wave propagates into the target, eventually overtaking and attenuating the shock.

In the early studies by Kidder¹ and others,² simple but useful relationships were derived relating the shock pressure, P , to the absorbed laser intensity, I .^{*} Assuming a one-dimensional problem in which the blow-off velocity is subsonic and the plasma is an ideal gas with heat capacity ratio, γ , equal to 5/3, one can write:

$$P = (0.055)(A/Z)^{1/3} I^{2/3} \quad (1)$$

where P is in TPa (1 TPa = 10 Mbar), I is in 10^{12} W/cm^2 , A is the atomic weight of the ions, and Z is the degree of ionization. Energy absorbed in ionizing the atoms has been ignored in this formulation.

Equation (1) implies that the shock pressures produced by laser irradiation are relatively insensitive to material density, as opposed to pressures generated by conventional plate-impact or impedance-matching techniques. More importantly, Eq. (1) predicts that a laser producing an intensity of 10^{12} W/cm^2 can generate pressures in the 100-GPa range. We will show that with the very high-power lasers at LLNL, it is reasonable to consider experiments up to the 10-TPa range.

The proper assessment of the properties of laser-generated shocks and the design of experiments, of course, require a much more detailed understanding than Eq. (1) can provide. For example, the absorption of laser light may be highly dependent on intensity and material, and may be affected by the temporal structure of the pulse as well. Preheat effects due to high-energy photons and electrons produced during the laser-light absorption process play a strong role in high-intensity laser compressions and must be understood. Finally, the temporal nature of the pulse and the evolution of the thermal properties between the critical surface and ablation front strongly regulate the lifetime and the spatial distribution of the shock wave. To more

*For a review of the laser-matter interaction problem up to 1975, see Ref. 3.

properly understand these phenomena, a sophisticated hydro-code with radiation and charged particle transport, such as LASNEX,⁴ must be used.

In this report we discuss the potential application of the high-power glass-laser systems at LLL to ultrahigh pressure equation-of-state (EOS) research. The extreme energy densities attainable with the Janus, Argus, and Shiva lasers are well known in the field of laser-induced-fusion research. We shall show that these lasers also offer the possibility of performing accurate equation-of-state experiments at shock pressures in the 0.1-to-10-TPa

range, where present theoretical models are least reliable and experimental data are extremely limited.

The first part of this report surveys the present status of theoretical EOS for materials at extreme pressures and temperatures. The remainder of the report is an assessment of the feasibility of EOS experiments using the LLL laser systems. We shall address both specific experiments, which may be implemented immediately using presently developed facilities and diagnostics, and more general concepts, which may be developed over the next decade.

PRESENT STATUS OF THEORETICAL AND EXPERIMENTAL EOS RESEARCH

High energy-density conditions occur in many situations in astrophysics, geophysics, and man-made laser-driven compressions. A study of the phenomena associated with these conditions requires the knowledge of the properties, such as the equilibrium state and transport behavior, of many materials over extreme ranges of temperature and density.

Theoretical approaches to the necessary equilibrium thermodynamics are of three general types. First, fundamental theories, based on our understanding of the underlying physics, attempt to be rigorous and complete, but they are correct only in some asymptotic limits of temperature and density. Examples are the Saha ionization equilibrium theory, valid at low densities, and Coulomb fluid theory valid at high densities. Second, approximate theoretical models are simplifications of first-principles theories, where known complications are either neglected or approximated. An example of a contemporary approximate theory is the Augmented-Plane-Wave (APW) model for electronic band structure of solids. A third approach involves phenomenological or totally empirical models, in which parameters are adjusted to agree with experimental data. A global EOS model may require all three approaches in various temperature-density regions, together with procedures for combining them in a thermodynamically consistent way.

The difficulty of accurate thermodynamic characterization of a material may be demonstrated by reference to the schematic diagram shown in Fig. 1. Here the useful range of several theoretical models has been indicated. Important factors unrepresented in this illustration include the complex-

ities of atomic number-dependence and molecular structures. The eight theoretical regions shown can be divided into three classes as follows:

Class 1. Theoretically precise

- Ideal and weakly coupled plasma
- Saha ionization equilibrium
- High density Coulomb fluid

Class 2. Theoretically approximate, ranging from accurate to inaccurate depending on T, ρ

- Dense partially ionized plasma
- Multiphase and neutral fluid
- Condensed matter

Class 3. Theoretically intractable at present

- Strong pressure ionization
- Hot, charged, dense plasma-liquid.

The theories listed as precise are supported by laboratory or astronomical experimental data in the appropriate T, ρ regions. The approximate theories of the second class generally approach the rigorously correct theories in their appropriate T, ρ limit. For example, the dense partial ionization model has the Saha ionization equilibrium as its low ρ and high T asymptote. To some degree these theories have experimental support. The intractable regimes represent the thermodynamic region where present modeling is least reliable.

The range of data from contemporary experimental techniques is also indicated in Fig. 1. These techniques include:

- Static compression, using high-pressure presses, such as diamond anvils, for measurements on the isotherm up to ~ 0.1 TPa.^{5,6}
- Explosive and gas-gun-generated shock-wave compression for measurements on or near the principal Hugoniot at pressures up to ~ 0.5 TPa.

Fig. 1. Schematic diagram of energy-density plane of EOS surface for fictitious but realistic material. The eight following are the theoretical regions discussed in the text: (1) ideal and weakly coupled plasma, (2) Saha ionization equilibrium, (3) high-density Coulomb fluid, (4) dense, partially ionized plasma, (5) multiphase and neutral fluid, (6) condensed matter, (7) strong pressure ionization, (8) hot, charged, dense plasma-liquid.

- Isobaric expansion ⁷ for measurements in expanded hot liquid metals at temperatures up to ~ 1 eV.

- Magnetic flux implosion for measurements along the isentrope to pressures of 0.5 to 1 TPa. ⁸

Hugoniot measurements have also been made in the 1-to-5-TPa range by Al'tshuler *et al.* ⁹ in the Soviet Union, and by Ragan *et al.* at LASL, ¹⁰ both

using a nuclear explosion as the driving source. Recent revisions in the standards used in the Soviet experiments have changed the data by as much as 50%. ¹¹ Because of their questionable accuracy, these data are not included in Fig. 1. The data point by Ragan *et al.* is not included because their experimental technique was applicable only to molybdenum.

We see that the range of validity of present theoretical models forms one outer boundary about a portion of the EOS space shown in Fig. 1, whereas another boundary is formed by the range of present accurate experiments. Intermediate to these regimes lies a large and poorly characterized region. This is the region where one must presently resort to statistical atom theories (Thomas-Fermi) or interpolation techniques.

The purpose of the experiments proposed in the following sections is to bridge this large gap be-

tween experimentally and theoretically understood portions of the EOS surface. The emphasis in this report will be on Hugoniot measurements, because methods for studying the Hugoniot at ultrahigh pressures appear most soundly developed at present. It is also important to realize that, because the Hugoniot traverses a central portion of the EOS space shown in Fig. 1, accurate data along this thermodynamic path can provide important tie points for interpolations between the various theoretically understood regions.

SHOCK PRESSURE ACHIEVABLE WITH LLL HIGH-POWER LASERS

The pursuit of fusion by inertial confinement has led to the development at LLL of the world's most powerful lasers. We will be discussing four LLL laser facilities^{1,2}; each in its time has been, is, or will be the world's most powerful. The smallest laser is Janus, constructed in 1974 and capable of delivering a maximum of about 200 GW of power in Gaussian pulses as long as 300 ps FWHM in each of its two arms. Argus, completed in 1976, can deliver up to several terrawatts on target in pulses potentially as long as several nanoseconds. Shiva, which began operation in December 1977, has twenty beams, bundled into two branches, and is expected to produce about 10-15 TW in each branch. Shiva Nova, scheduled for the early 1980's, will produce 100 TW in each of two branches.

We have used the LASNEX code⁴ to study the irradiation of 1-mm-diam disks of solid D_2 , LiH, Fe, and U by the aforementioned lasers. The maximum pressures achievable in these materials are summarized in Fig. 2. The pulse characteristics are listed in the figure caption. Figure 2 also shows the maximum shock pressures possible in these materials using conventional laboratory techniques. It is seen that even in a Janus EOS experiment, pressures could be produced that would exceed those possible by any other present laboratory technique. With the more powerful lasers, interesting regimes of present theoretical uncertainty could be experimentally addressed for the first time.

As exciting as the prospect of doing experiments in a presently inaccessible regime of EOS space might be, the foregoing discussion remains little more than a curiosity unless accurate diagnostic techniques can be developed. It is easy to appreciate that the diagnostic requirements for accurately characterizing a laser-driven, shock-wave event are

Fig. 2. LASNEX-calculated peak shock pressures achievable in various materials by irradiation with different LLL lasers (pulse characteristics used in these calculations were 50-J/250-ps FWHM for Janus, 10³-J/1 ns FWHM for Argus, 10⁴-J/1 ns FWHM for Shiva, and 10⁵-J/1 ns FWHM for Shiva Nova; all pulses were Gaussian).

extremely severe. The necessity of small targets and the short duration of typical laser pulses (no more than a few nanoseconds) mean that any measurement must be made with high spatial and temporal resolution. For example, an accurate in-target velocity measurement in a 1-mm-diam target irradiated by a 1-ns pulse would require time resolution of ~ 10 ps and spatial resolution of $\sim 1 \mu\text{m}$. We will see that such techniques are presently available and that the design of clean, accurate experiments appears feasible.

In the following section we discuss some of the properties of laser-generated shocks that must be properly understood before meaningful EOS experiments can be performed.

Then in the section entitled "Equation-of-State Diagnostic Techniques" diagnostic requirements for ultrahigh pressure EOS experiments are discussed, and an assessment of the applicability of various techniques to laser experiments is made. The section entitled "Previous Experiments" reviews the rather small amount of previous experimental work in this field. In the sections, "EOS Feasibility Experiment (1 TPa) Using Janus Laser" and "Argus EOS Experiment Designed to Reach 2 TPa," we propose experiments that could be performed on the Janus and Argus lasers in the near future. And in the final section we discuss other potential laser-EOS experiments.

STRUCTURE OF LASER-GENERATED SHOCK WAVES

Any EOS experiment requires a determination of the initial and final states of the material being studied. In a shock-wave experiment this means that the conditions ahead of and behind the shock front must be measured. In the simplest experiments the state of the material ahead of the shock is the ambient state and is measured before the experiment; if the initial state of the material is changed by some phenomenon propagating ahead of the shock, additional diagnostics are required to characterize this new initial state. This often proves to be too difficult experimentally, and the alternative is the design of an experiment with sufficient care that the initial state is unperturbed during the experiment. This is one requirement of a so-called "clean" experiment.

All of the state variables cannot in general be measured in an EOS experiment. Usually, only one or two variables are measured and are related to the other variables of the problem through the conservation laws of mass, momentum, and energy. In order to apply these laws, the geometry of the shock wave must be known. The simplest shock-wave experiments are one-dimensional (i.e., the shock wave is planar), and the characteristic thickness of the shock front is negligibly small. In the following we will therefore be striving toward experiments that satisfy these conditions.

The foregoing considerations require that early experiments must study the structure of laser-generated shocks. Potentially important phenomena that must be characterized include:

is essentially spherical at distances from the surface that are comparable to the diameter of the irradiated spot. The achievement of a planar shock therefore requires targets that are thin relative to the spot diameter. Shock planarity can be verified experimentally and will be discussed under "Equation-of-State Diagnostic Techniques." Geometrical effects may also be studied with two-dimensional LASNEX calculations.

Effects of Spatial Variation in Laser Intensity

The ideal laser pulse exhibits a super-Gaussian spatial variation of intensity: i.e., $I/I_0 \approx \exp[-(r/r_0)^n]$, where r_0 is the spot radius and $n \geq 5$. Thus, I is very uniform across the irradiated spot. In reality, however, the pulse may contain "hot spots," small regions of higher intensity, due to beam breakup in the laser amplifier chains. Theoretically, the thermal conductivity of the laser-generated plasma is sufficiently high to smooth out these hot spots as long as $(v_{osc}/v_{therm}) \ll 1$, where v_{osc} is the velocity of the plasma electrons in the electric field of the laser radiation, and v_{therm} is their thermal velocity. This condition is satisfied for $I \geq 10^{14} \text{ W/cm}^2$, which covers the intensity regimes we'll be discussing for Janus and Argus experiments. At higher intensities, the effects of a non-uniform laser profile on the shock structure must be assessed carefully.

Absorption Mechanisms that Depend on Angle of Incidence

At high intensities, a significant fraction of the absorbed laser energy may be due to resonant

Geometrical Effects

The rapid heating of a planar surface by the absorption of laser energy produces a shock wave that

absorption,¹³ which varies with angle of incidence. If the target is not situated in the Gaussian waist of the incident pulse, this mechanism could lead to a nonuniform shock structure. These effects should not be significant at intensities below $\sim 10^{14}$ W/cm², but may become important in higher-intensity experiments designed to probe beyond pressures of ~ 2 TPa.

Suprathermal Electrons

Absorption of laser light by collective mechanisms can produce electron distributions that are highly non-Maxwellian, and the production of very-high energy electrons is particularly favored. Experimentally, this is a phenomenon of great importance, because the long mean-free paths of such suprathermal electrons permit them to penetrate far into the target, preheating material ahead of the shock and making shock compression more difficult. This problem has been of considerable concern to laser-fusion researchers. A principal concern in an EOS experiment is that preheating may significantly alter the thermodynamic state ahead of the shock front, so that the initial conditions of the problem are effectively unknown.

Clearly, targets must be designed such that the region of study lies beyond the range of suprathermal electron preheat effects. Here LANEX proves invaluable. In the section entitled "EOS Feasibility Experiment (1 TPa) Using Janus Laser," we will see that the temperature profile of the shock may be strongly modified by suprathermal electron preheating; this result suggests a potentially useful means for exploring preheating effects experimentally.

EQUATION-OF-STATE DIAGNOSTIC TECHNIQUES

A shock compression is fully described by the pressure p , density ρ , and internal energy E ahead of and behind the shock front, as well as the shock propagation velocity u_s and material velocity u_p . The initial state p_0, ρ_0, E_0 is usually known. Equations of conservation of mass, energy, and momentum relate the remaining five unknowns. Thus, two of these five parameters must be measured to determine completely an equation-of-state point, $p = p(\rho, E)$.

To appreciate the kinds of diagnostic tools that might apply to these experiments, we should first

Radiative Preheating from Shock Front

The very high pressure shock waves discussed here are characterized by temperatures ranging from several eV to several hundred eV. Thermal radiation frequencies from these shock fronts may therefore exceed the plasma frequencies of most metals, permitting the thermal radiation to propagate ahead of the shock front and couple into the uncompressed matter. The consequence would be an effective broadening of the shock front. Such phenomena are well documented in studies of shocked gases, in which, due to the long photon mean free paths, radiation-coupled-shocks with characteristic widths as great as a millimeter are possible.¹⁴ Of course, these effects would be greatly reduced in a metal; however, shock widths of even a few microns would be a serious problem in targets that are no more than a few tens or hundreds of microns thick.

Thermoelectric Transport Across Shock Front

The large temperature gradient across the shock front may provide the means for electrons in the shock-compressed material near the front to move into the uncompressed material, travelling at roughly the Fermi velocity, which may typically be an order of magnitude higher than the shock velocity; these electrons represent another possible preheat mechanism.

The above phenomena must be considered in the successful design of any laser-generated shock-compression experiment. These effects will be addressed in the following sections on diagnostics and proposed experiments.

discuss briefly how target dimensions are determined, because target size and laser-pulse length together define the spatial and temporal scales of the problem.

The dimensions of flat disk targets are determined by the following constraints:

- Laser-irradiated area must be small enough to permit sufficiently high intensity, I , and thus high pressure ($\propto I^{2/3}$, approximately), to be attained.
- Diameter of irradiated area must be large compared to disk thickness so that shock-wave propagation is essentially one-dimensional.

- Target thickness must be large enough to permit study of the shock in a region that has not been preheated by fast electrons or x rays produced near the critical surface.

- Target thickness (and, hence, shock transit time) must be sufficiently small that, after the laser power is turned off, rarefaction waves from the ablation surface do not significantly attenuate the shock. For later discussion we refer to this characteristic thickness as the "rarefaction range."

For significant measurable pressures (>0.5 TPa) to be produced with a Janus pulse (≈ 300 ps), the above considerations require a target less than 500 to 1000 μm in diameter and 30 to 50 μm thick. Shock velocities for the pressures discussed in this report are expected to range from 20 to 50 $\mu\text{m}/\text{ns}$. The transit times for such shocks through Janus targets may therefore be no more than a few ns; this defines the total duration of the experiment. An accurate temporal characterization of any material property on this time scale therefore requires unusually precise resolution, on the order of a few picoseconds. Diagnostic tools used in conventional shock-wave experiments, (e.g., self-shorting electrical pins) respond much too slowly (a few hundred picoseconds) to be of use in these experiments. We must therefore consider whether material properties can be measured with meaningful accuracy on the time and spatial scales defined above.

Unfortunately, in an EOS experiment it is usually not possible to measure accurately p , ρ , and T directly. What can be readily measured is u_s , and we discuss below how this may be done in laser EOS experiments. We also discuss a potential method for measuring u_p .

Shock Velocity Measurements in Metals

This technique uses an ultrafast streak camera to measure the temporal dependence of the brightness of a metallic target surface (see Fig. 3). The thermal radiation produced by the high-temperature shock front will become visible to the streak camera only when the shock has advanced to within a few skin depths, δ , of the surface. As the shock progresses to the surface, the surface brightness will increase until the shock breaks out of the free surface, at which time rarefaction and rapid cooling ensue. The rise time of the signal recorded by the camera will be approximately δ/u_s . For the optical-frequency components of this signal, which will be recorded by the streak camera, $\delta \sim 0.1 \mu\text{m}$. In aluminum, for example, at 2 TPa, $u_s \sim 30 \mu\text{m}/\text{ns}$. The arrival time of the shock at the free surface could therefore be determined to within ~ 3 ps by accurately recording the onset of this optical signal. This uncertainty is comparable to the time resolution (~ 10 ps) of the streak cameras presently used in laser fusion research at LLNL.¹⁵

The feasibility of studying the high-temperature shock front on a 10-ps time scale would permit us to assess experimentally some of the phenomena discussed in the previous section (suprathermal electron effects, radiative heating from the shock front, etc.), because if they occur, they would add temporal structure to the radiated signal. Obviously, if we cannot see such effects on this time scale, they play no significant role in the problem. We can also study the spatial distribution of the thermal radiation with spatial resolution near 10 μm by appropriate magnification in the streak camera imaging optics. Thus, by studying the tem-

Fig. 3. Optical measurement of shock arrival times at metal surfaces (because visible thermal radiation produced by shock heating will be seen by the streak camera only after shock has advanced to within a few skin-depths of the free surface, camera will record a rapidly increasing signal).

poral and spatial history of the thermal radiation emitted across the rear face of an irradiated disk, uniformity of the shock can be assessed.

By measuring the arrival time of the shock at two precisely measured steps on the rear surface of a laser-irradiated disk, a shock velocity may be obtained (see Fig. 4). For a step height of $25\text{ }\mu\text{m}$ and $u_s = 30\text{ }\mu\text{m}$, an average shock velocity may be measured with an accuracy of $\sim 2\%$, assuming that the steps are smooth to within a few tenths of a micron and that their separation can likewise be measured to within a few tenths of a micron; this is readily achievable.

With the long pulses (6 to 10 ns) that are possible only on the higher-energy lasers, Argus and Shiva, targets may be made larger, because the rarefaction range increases. For a 10-ns pulse a step height of 75 to 100 μm could be used, and a consequent shock velocity measurement accurate to $\sim 1\%$ may be expected. This approaches the accuracy possible with the best present techniques used in conventional experiments at much lower pressures.

Shock Velocity Measurements In Transparent Solids

A technique for measuring shock velocities in initially transparent targets was first suggested by van Kessel and Sigel.^{16,17} The method, sketched in Fig. 5, consists of backlighting the target normal to the direction of shock propagation with a small visible-light laser. As the shock propagates through the target, material is ionized by the high pressure and temperatures, and the target becomes opaque to the backlighting source. An ultrafast streak camera is used to obtain a temporal and spatial record of the opaquing of the target. This record permits a shock velocity to be determined. The accuracy of this technique is limited by time and spatial resolution of the camera. For example, $1\text{-}\mu\text{m}$ spatial resolution and 10-ps time resolution are required to determine the shock velocity of a $25\text{-}\mu\text{m/ns}$ shock to 2% accuracy.

One problem, inherent in this diagnostic, is the question whether the ionization front can be

Fig. 4. Sketch of idealized streak-camera photograph showing optical signals corresponding to shock breakout at two surfaces on stepped target.

Fig. 5. Schematic of technique for measuring u_s in initially transparent materials and an idealized streak photograph obtained.

associated with the shock front and not with preheating effects. Resolution of this question requires either a good ionization model (which demands very low Z) for the target and a quantitative understanding of preheating effects or an additional diagnostic. A possible diagnostic is sketched in Fig. 6. Here the experimental configuration is identical to that in Fig. 5, except for a thin metal disk at the bottom of a narrow notch facing the oncoming shock wave. By observing the onset of motion of the disk, the arrival time of the shock can be ascertained.

Two spatially resolved dimensions of the shock propagation may be obtained by using a mode-locked dye laser, with pulse lengths of about 5 ps at ~ 2 -ns intervals, as the backlighting source. In this

method, the streaking rate of the camera is slowed down and the slit opened up to a few hundred microns. If the streaking rate is slow relative to the 5-ps pulse duration, then a series of two-dimensional images is produced on the film. This technique would prove especially useful for studying the geometrical effects mentioned in the previous section.

Particle Velocity Measurement

A possible technique for measuring the particle velocity behind the shock front employs the ultrafast x-ray streak camera developed in the LLL Laser Program.¹⁸ This instrument, presently used

Fig. 6. Possible experimental technique for differentiating between shockwave and preheat effects in irradiated transparent targets. In (a) ionization front is caused by preheat, which creates high temperature, but does not produce high particle velocity necessary to cause rapid motion of inserted disk; rapid acceleration of disk occurs only at later time, when shock has arrived. In (b), the ionization front is generated by shock, so that motion of disk commences simultaneously with arrival of shock.

for space-time studies of x-ray emission from imploding laser-fusion targets, offers simultaneous temporal resolution of 15 ps and spatial resolution of $6 \mu\text{m}$. It has a nearly energy-independent response to photons in the 0.3-to-8-keV range. An experimental arrangement for particle velocity measurement is shown in Fig. 7.

The target is composed of two adjoining slabs, chosen to have greatly differing photon cross sections in the energy range of the x-ray source, one being essentially transparent, the other opaque to the x rays. For a source energy of 10 keV, suitable examples would be LiH and Al. The x rays could be generated by the beam not used for target irradiation.

A backlighting technique is then used to observe the transit of the target interface across the field of view of the streak camera ($\sim 100 \mu\text{m}$ at $50\times$ magnification). The velocity of the interface is equal to the particle velocity on either side of the interface. It is possible that the opacity of the transparent material will vary sufficiently upon shock compression that the shock front could be also observed in the streaking record. Neglecting diffraction effects, the accuracy estimated for this

measurement would be 6 to 8% for $u_p = 35 \mu\text{m}/\text{ns}$ (consistent with $u_s \approx 50 \mu\text{m}/\text{ns}$).

Fig. 7. Possible particle-velocity diagnostic utilizing ultrafast x-ray streak camera.

Other Property Measurements

The foregoing discussion has focused on velocity measurements in the shock-compressed target. These are the only properties that appear feasible to measure at present, using existing diagnostic tools. However, the long-range view includes the desire (and necessity) to measure additional properties. A direct density measurement would be of great value. Because density is related to shock and particle velocity by $\rho/\rho_0 = (1 + u_p/u_s)$, errors in u_p and u_s can lead to large uncertainties in ρ when ρ/ρ_0 becomes large. One potentially useful density diagnostic involves measurement of the Stark broadening of x-ray lines from seeded targets. Early experiments exploring this technique¹⁹ have been unsuccessful because other effects (e.g., Doppler broadening, opacity broadening) appear to dominate the line broadening at low compressions. At extremely high densities, which might be achievable by isentropic compression with Shiva or Shiva Nova, the Stark widths may become great enough to provide valuable density information.

Temperatures associated with strong shocks are of considerable interest for a complete EOS

model. At shock pressures of 1 to 10 TPa, the atomic shell structure breaks down as electrons become delocalized. Heat capacities in this pressure range on the Hugoniot may therefore exhibit large peaks. Because internal energy is one of the parameters obtained from a shock wave experiment, temperature measurements are necessary to identify these heat capacity peaks.

The time durations of the experiments considered here are 1 ns or less; therefore, any temperature diagnostic must be able to respond rapidly, perhaps in 10 ps. At present, the only instrumentation available with this time resolution works in the visible (optical-streak camera) or x-ray (x-ray streak camera) spectral regions.

As described in Appendix A, each of the techniques to measure temperature in the shock-compressed medium has problems in instrumentation and interpretation. Accuracies of $<10\%$ are probably needed if the measurements are to be significant for EOS modeling. This accuracy appears to be neither trivial nor impossible. The actual temperature determination must, however, be considered as a second generation experiment.

PREVIOUS EXPERIMENTS

Only two experiments aimed at characterizing the properties of a laser-initiated shock wave have been reported in the literature. Van Kessel and Snel²⁰ studied the spatial and temporal development of shocks generated by a 20-J, 5-ns laser pulse focused onto 40- μ m-diam spots on solid-hydrogen and plexiglass slabs. The backlighting shock-velocity diagnostic discussed in the previous section was employed. The observed phenomenon was highly two-dimensional, due to the small irradiated spot (an unfortunate necessity with lasers of only moderate power).

A maximum shock velocity of 38 μ m/ns was found for the hydrogen target. Using an extrapolation of EOS measurements made at much lower pressures,²⁰ a peak pressure of 0.2 TPa was estimated. This figure is rather impressive, because the highest pressure achievable in hydrogen by any other in-laboratory means is about 10 GPa in a single shock. However, the pressure estimated above is in rather poor agreement with that predicted by one-dimensional code calculations that predict a maximum pressure of about 2.1 TPa in this experiment. This substantial discrepancy between theory and experiment is primarily due to the

two-dimensional nature of the shock front. Our LASNEX calculations also indicate that suprathermal-electron preheat probably played an important role in this experiment as well. These results point up the necessity of understanding the phenomena discussed in the section entitled "Structure of Laser-Generated Shock Waves." Nonetheless, this experiment represented the first direct observation of a laser-initiated shock wave and was the first to indicate the potential of similar experiments to high-pressure research.

Billon *et al.*²¹ studied the propagation of laser-initiated shocks in aluminum and CH_2 targets, using both planar and cylindrical geometries. A planar CH_2 target was irradiated with a 30-J/3-ns pulse focused on a 60- μ m-diam spot. The shock-transit time through the 30- μ m-thick target was measured by an interferometry technique, using the first harmonic of the incident laser light (0.53 μ m) as a diagnostic beam and fast framing cameras to photograph the interferograms. The emergence of the shock from the rear target surface was clearly seen about 1.5 ns after the initial interaction with the laser pulse. The estimated mean shock velocity corresponded to a pressure of about 0.17 TPa. A

LASNEX study of this experiment has not been performed, but we can conclude from other LASNEX calculations that suprathermal-electron effects were undoubtedly severe in this experiment.

In the experiment with cylindrical geometry, an aluminum tube, 245 μm in diameter and 67 μm in wall thickness, was symmetrically irradiated normal to the cylinder axis by four laser beams. A total of about 190 J was deposited in about 5 ns. The area of irradiation was not given. The breakthrough of the shock was detected by observing the extinction of light from a probe laser aligned along the tube axis. A symmetric extinction of the probe light was observed, indicating that high thermal conductivity smoothed any discrepancies in nonuniform illumination. The shock transit time corresponded to a pressure of ~ 0.2 TPa.

EOS FEASIBILITY EXPERIMENT (1 TPa) USING JANUS LASER

One of the standard methods of shock-wave EOS measurement is impedance matching. The technique involves the propagation of a shock from a known standard material into an unknown sample (or vice versa). By measuring u_s in both materials and matching boundary conditions, one can determine the EOS of the unknown relative to that of the standard. Because only a measurement of u_s is required, our first series of experiments will be of this type. We now present results of a one-dimensional LASNEX study of an impedance-matching experiment appropriate for the Janus laser.

A possible target for these experiments is shown in Fig. 8. The shock-velocity diagnostic discussed under the heading, "Shock Velocity Measurements in Metals," would be used to measure the history of thermal radiation emitted from each of the stepped surfaces. We selected aluminum and tungsten as the two sample materials for this study; however, the number of feasible and interesting combinations is large. Aluminum is typical of low-Z metals, for which our theoretical understanding is best (another example would be beryllium), whereas tungsten represents the many high-Z metals for which our understanding is rather poor. The maximum pressures achieved by conventional means in these materials are about 0.2 TPa in Al and 0.5 TPa in W.²⁰

A 50-J/250-ps FWHM Gaussian pulse impinged upon a 500- μm -diam spot on the Al surface. About 30% of the incident radiation was absorbed.

These results are in strong disagreement with the higher theoretical predictions of the pressure. This discrepancy has been attributed partially to the effects of lateral thermal conduction; i.e., the area of the ablation front is much greater than the area of the focal spot due to lateral transport of energy between the critical surface and the ablation front. A substantially larger focal spot will eliminate significant effects due to this phenomenon. The much higher powers produced by the LLJ lasers make this possible.

We now discuss the design of two EOS experiments using the Janus and Argus lasers and directed at measuring materials properties in the 1-to-2-TPa range.

Fig. 8. LASNEX-designed aluminum-tungsten impedance-matching target for Janus experiment.

Suprathermal-electron generation in the Al was characterized* by $\alpha = 6$.

*In LASNEX, suprathermal electrons, of velocity v are produced by a source function varying as $v^2 \exp(-v^2/\alpha v_s^2)$, where v_s is the mean thermal electron velocity. The parameter α characterizes the energies of the suprathermals and is chosen such that calculations reproduce the high-energy features of measured x-ray spectra.

Fig. 9. Calculated peak shock pressure vs time for irradiation of target shown in Fig. 8 by a 50-f 250-ps FWHM Gaussian pulse.

Figure 9 shows the calculated peak shock pressure plotted vs time. A peak pressure of about 1.5 TPa in the Al slab corresponds to the peak of the laser pulse at 300 ps. As the laser power decreases, rarefaction waves from the ablation front overtake the shock and p decreases. By the time the shock has reached the Al-W interface ($t = 1.7$ ns), p has decreased to about 0.6 TPa. Because of the higher shock impedance ($\rho_0 u_0$) of W, a 10-TPa shock is produced in the W and a 1-TPa shock is reflected back into the Al at this point. At $t = 2.7$ ns, the shock arrives at the back surface of the W, its magnitude now reduced to about 0.8 TPa. Pressures of this magnitude exceed those achievable in any material by any present-in-laboratory means at 111.

Preheat effects can be obtained from Fig. 10, which shows the rate of energy deposition vs distance X into the target at $t = 300$ ps, the peak of the laser pulse. The two sources of preheating shown are suprathermal electrons and x rays generated near the critical surface in Al. For $X < 20 \mu\text{m}$,

Fig. 10. Calculated rate of preheat energy deposition vs depth into target for Janus aluminum-tungsten impedance-matching experiment (time = 300 ps at peak of pulse).

suprathermal electrons are seen to be the dominant preheating mechanism. Beyond $X = 20 \mu\text{m}$ preheating effects are not important, as can be seen by the plot of temperature T vs X in Fig. 10, where T drops from 1 eV at $X = 10 \mu\text{m}$ to less than 0.1 eV at $X = 18 \mu\text{m}$. This temperature compares with a value at the shock front of about 6 eV. Another potential source of preheat is blackbody radiation from the shock front itself (see "Structure of Laser-Generated Shock Waves"). To assess the importance of this phenomenon, low-energy groups were included in the photon group structure. However, effects of such a precursor did not appear significant at these shock temperatures. Once the shock was traversed $X = 15$ -to- $20 \mu\text{m}$ region, pressure and temperature profiles steepen significantly, their widths apparently determined only by zone thicknesses. We note that our one-dimensional calculations do not include the effects of self-generated magnetic fields, which are expected to enhance suprathermal-electron temperatures. To account for this effect, the first free surface of the target shown in Fig. 8 was positioned at $X = 30 \mu\text{m}$.

As discussed previously a sharp turn-on of the optical signal emitted from each step is expected. The LASNEX calculations support this idea. Figure 11 shows the electron temperatures as functions of time for various target depths. The expected sharp turn-on of thermal radiation* is clearly seen. Near the rear surface ($X = 49 \mu\text{m}$), where zoning is finest, an optical-signal rise time of ≤ 10 ps is calculated. It is interesting to note the early-time temperature rises due to suprathermal-electron preheat. Figure 11 illustrates how a high preheat level may broaden the fast turn-on of the optical signal and lead to a spurious shock-velocity measurement. This result suggests, in fact, an interesting series of experiments to study fast-electron preheat effects by measuring the time dependence of the free-surface brightness of a thin foil.

The 5- μm -step thickness shown in Fig. 8 is chosen as a compromise between the desire to sample u_s at as many depths as possible (since u_s is time-dependent), the necessity to make the shock transit time long compared to the time resolution of the streak camera, and the constraint of keeping the target thin. Also, this thickness should be large compared to the roughness of the step surface ($<0.5 \mu\text{m}$ for a carefully prepared surface). For a 5- μm step we thus expect shock velocity measurements in

*For the several-eV temperatures discussed here, the peak of the blackbody function occurs at wavelengths so much shorter than those to which the camera is sensitive that variations in optical signal are roughly proportional to variations in T (Rayleigh-Jeans limit).

Fig. 11. Temperature histories at several depths in the 50- μm -thick aluminum-tungsten impedance-matching target shown in Fig. 8 (X = distance from initial irradiated surface; time = zero when leading edge of laser pulse first hits target).

a Janus experiment to be accurate to 15% or better.

While a u_s measurement of 15% accuracy is not sufficient to improve significantly our present EOS

models, it is more than adequate to establish the feasibility of laser-shock-wave experiments, to study preheat effects, to test code predictions, and to develop diagnostic capabilities. These would be the

goals of Janus experiments. As mentioned previously, with long pulses (>1 ns) the high-accuracy measurements necessary to provide useful EOS data may be made.

ARGUS EOS EXPERIMENT DESIGNED TO REACH 2 TPa

The ultimate accuracy of a Janus EOS experiment (in which velocities are measured) is limited by the short pulse length. Short pulse length also prevents us from making optimum use of the laser's pressure-producing capability, because rarefaction waves substantially reduce the pressure at the shock front before it has traversed the preheated region of the target (see Fig. 9). These constraints can be overcome in an Argus shock-wave experiment, in which nearly square pulses several nanoseconds long are possible.

Figure 12 shows the time dependence of the LASNEX-predicted shock pressure in a 1-mm-diam Al-W impedance-matching target irradiated by a 3-kJ 3-ns "square" pulse, produced by stacking ten 300-fs 300-ps FWHM pulses at 300-ps intervals. Here we see an 0.8-TPa shock is generated in the Al and 1.8-TPa forward and reflected shocks are produced at the Al-W interface. Note that, because of the longer pulse length, this higher pressure experiment is actually achievable with a lower intensity than that proposed for the Janus experiment.

An assessment of preheat effects reveals a preheated region of about $90\text{ }\mu\text{m}$. At this depth, LASNEX calculations indicate suprathermal electrons heat the material to about 0.1 eV. A possible impedance-matching target is shown in Fig. 13. The high powers and long pulse lengths achievable with Argus permit the use of a significantly larger target than that required by Janus. Note that step thicknesses for the Argus target are $25\text{ }\mu\text{m}$, compared to the $5\text{-}\mu\text{m}$ steps of the Janus target. Shock velocity measurements accurate to about 2 to 3% should therefore be possible in a 3-ns-pulse Argus experiment. With a 6-to-10-ns square pulse, shock velocity measurements accurate to about 1% appear possible. The accuracy of an Argus EOS experiment, therefore, should approach that possible with conventional techniques (i.e., HE and two-stage gas guns), which operate at much lower pressures.

The transit time for the shock across the four steps of the Argus target shown in Fig. 13 is about 5 ns, which exceeds a streak-camera writing time (~ 1.5 ns), if 10-ps time resolution is required, it therefore becomes necessary to time-delay a portion of the optical signal. This delay could be achieved

by mounting the target directly against a coherent bundle of low-dispersion optical fibers, as illustrated in Fig. 13. Outside the target chamber, the bundle may be separated and the signals routed through appropriate optical delays.

We summarize this section by noting that in an Argus EOS experiment:

- Pressures approaching 2 TPa can be produced in a simple impedance-matching target. Pressures this large have not been previously produced by any conventional means.

- Accuracy of an Argus experiment should approach that possible with the best present in-laboratory technique, the two-stage gas gun, which is limited to lower pressures.

Figure 14 compares possible Al-W impedance matching experiments performed with Argus, Janus, and the H Division two-stage gas gun. It is seen that the capabilities of the LLL high-pressure research effort could be significantly extended into interesting and relevant regimes by the successful development of the experiments we have discussed.

Fig. 12. Peak shock pressure vs time in Argus impedance-matching experiment.

Fig. 13. Aluminum-tungsten impedance-matching target for Argus EOS experiment.

OTHER POTENTIAL LASER-DRIVEN EOS EXPERIMENTS

This report has concentrated on shock-compression experiments, because they appear to be the most feasible candidates for first-generation experiments. However, laser irradiation of matter can produce a rather rich variety of material states, and it is important to consider other potential EOS experiments of interest.

Other Shock-Wave Experiments

These include multiple-shock, expanded initial state shock, and blast-wave experiments. Multiple-shock experiments would be of interest because they permit study of material states closer to the principal adiabat than the principal Hugoniot. Valuable temperature data could be obtained from such experiments. The production of the multishocked state *should be readily achievable with the multibeam lasers at LLL*. Expanded initial state shock experiments would be of interest in exploring the

high-temperature side of the principal Hugoniot. True blast-wave experiments should be possible with the very short pulses (down to ~ 30 ps) possible with the LLL lasers. Thus, the attenuation of the laser-produced shock should be described by self-similar relations very early in the experiment. An integral blast-wave experiment has been proposed by Grover.²² It is estimated that pressures above 1 TPa could be measured, if Argus were used as the energy source.

Isentropic Compression

States near the principal isentrope can be achieved by properly tailoring a high-energy laser pulse so that the irradiated material actually experiences a long sequence of shocks. Experiments in this regime would be highly desirable, because they could provide direct tests of the APW band-theory calculations discussed in the section entitled "Pres-

Fig. 14. Comparison of various aluminum-tungsten impedance-matching experiments (solid curves represent Al and W principal Hugoniot; dashed curves represent reflected Hugoniot at Al-W interfaces).

ent Status of Theoretical and Experimental EOS Research."

Expansion Experiments

The efficiency with which x rays and high-energy electrons are produced by laser-matter interaction suggests potentially important expansion experiments in which material is heated nearly uniformly throughout its volume. The resultant material states would approximate an isochoric heating followed by isentropic expansion of the hot plasma. Such EOS paths in the multi-eV region

would provide experimental data in the dense partially ionized plasma regime (see Fig. 1).

Accelerated Flyer Plates²³

Experiments of this type would use laser-driven ablation to accelerate a flyer-plate, which would subsequently impact the target, generating a high-pressure shock wave. The potential advantage of this technique is that the target would not be subjected to suprathermal-electron preheat, nor would there be concern about structure in the shock profile due to variations in the laser-pulse profile.

CONCLUSIONS

We have seen in this report that the LLL high-power lasers offer the potential to extend significantly the present range of EOS experiments. Pressures in the 0.5-to-30-TPa range could be achieved with facilities either presently in operation or planned for the next five years. Thus, a pressure-density regime of great theoretical uncertainty could be addressed for the first time by laboratory experiments.

To illustrate the potential value of accurate ultrahigh-pressure, shockwave experiments, we

show in Fig. 15 a plot of shock pressure vs density for beryllium. Shown here are the available experimental data, extending up to ~ 0.1 TPa, as well as the principal Hugoniot calculated from two theoretical models, one the well-established TFD-based EOS of Cowan,²⁴ the other a recent shell-structure model from Graboske and Wong.²⁵ Note the asymptotic agreement between the models—at low pressure where the models can be normalized to experimental data and at extremely high pressures (~ 100 TPa) where first principles theory becomes

Fig. 15. Principal Hugoniot of beryllium calculated from two theoretical models, TFD-based EOS of Cowan and shell-structure model of Graboske and Wong.

reliable. However, in the intermediate range 1 to 100 TPa, the two models predict densities for a given pressure that differ by as much as 30% for this simplest of metals. This difference is illustrated in Fig. 16. Clearly, shock wave data of a few percent accuracy in this pressure range would be invaluable in assessing the validities of these models.

Fig. 16. Fractional difference between calculated densities of beryllium along principal Hugoniot between TFD-based and shell-structure EOS models as a function of pressure.

APPENDIX A

TEMPERATURE MEASUREMENTS IN LASER-SHOCKED MATERIALS

Conventional pyrometric techniques for determining the temperature of gray body radiators operate best on the high-frequency side of the black-body spectrum. In this spectral region the radiant intensity increases as a high power of temperature. However, the intensity also drops off with increasing frequency like $\exp(-h\nu/kT)$; therefore, operating at frequencies too far above the blackbody peak results in a small signal-to-noise ratio and inaccurate temperatures. Depending on the details of the detector response and the noise spectrum, an optimal energy range to work in is 2 to 5 times the energy of the blackbody peak, or 9 to 25 times kT of the radiator.

For shock experiments between 1 and 10 TPa, the temperatures will be between 5 and 100 eV for various materials. Thus, only at the highest temperatures could the x-ray streaking camera be operated in an optimal spectral range. For all such experiments, the optical streaking camera will operate in the Rayleigh-Jeans tail of the blackbody spectrum, where the radiant intensity is proportional to T . Therefore, meaningful pyrometry with optical radiation will require absolute intensity measurements of accuracy better than 10%. On the other hand, the x-ray measurements may be made with considerably less accuracy, but one must anticipate a noise problem, especially from the expanding laser-generated plasma.

The geometry of the radiation pyrometry experiments will depend on the type of material studied. For an opaque material like a metal with visible radiation, residual temperature of a free surface after shock and release back to zero pressure can be measured.^{26,27} The residual temperature, although interesting, would be confused by the fact that even if the relaxed material is in a homogeneous equilibrium state, it will be an expanding, highly ionized vapor with a varying opacity. In order to measure the temperature associated with a state close to the principal Hugoniot, a metal must be backed with a transparent anvil.²⁸ At the shock pressures associated with the experiments described here, we know of no optically transparent anvil materials. Thus, direct optical pyrometric temperature measurements on metals shock-compressed to >1 TPa will be very difficult without significant increases in the time response of our diagnostics. At the higher pressures, x-ray pyrometry may work adequately, but opacities and noise sources must be well characterized.

The situation may be much simpler for optically transparent samples like solid H_2 or polyethylene. Because in this case the unshocked part of the sample forms a transparent anvil, material on the principal Hugoniot can be observed directly for times as long as 1 ns. If the electrons and ion temperatures behind these strong shocks can equilibrate on such short time scales, the temperatures can be measured with optical pyrometry. Because the electron-ion equilibration time for elastic scattering is $(M/m)^{1/2}$ times the electron-ion scattering time, where M and m are the ion and electron masses, respectively, and, because the electron-ion scattering time is typically 10^{-15} s or less for room-temperature normal-density metals, thermal equilibration may occur on sub-picosecond time scales. Nonequilibrium radiation, such as that associated with inelastic collisions of electrons with ions and subsequent line radiation, must, of course, be accounted for.

Line emission may also be used as a temperature indicator if the participating atomic levels can achieve local thermodynamic equilibrium (LTE) in the short time scales. For example, doping H_2 with a small concentration of He could give rise to 587.5-nm radiation from HeI and 468.6-nm radiation from HeII, which can be used for temperature determinations at 3 to 6 eV. Besides the LTE assumption, problems associated with this measurement include the density dependence of oscillator strengths and Stark broadening.

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