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Laser-Generated Shock Waves and Applications to Advanced Materials

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The use of lasers for the generation and application of high-pressure shock waves offers unique advantages and challenges. In contrast to impact systems, the range of pressures and strain rates is substantially greater using laser drive. The ability to change the temporal shape of the drive pulse allows a variety of strain-rate conditions to be obtained. In addition, high time-resolution in situ diagnostic methods are relatively simple to implement. Lasers can be at a disadvantage compared to impact methods in terms of shock generation, simplicity of the states achieved, the difficulty of characterizing bulk properties, and sample size. I will review the physics of laser-driven shock physics, diagnostic methods, and applications, with an emphasis on material physics. I will also present some views on important new directions for this area of research.

Laser-Generated Shock Waves

One of the main uses of lasers in materials applications has been the attempt to determine thermodynamic properties such as equations of state. Recently, short pulse lasers have been used successfully to study rapid chemical reactions and the approach to thermodynamic equilibrium in shocked organic materials. While this is an exciting development and relates to our interest here, it is beyond the scope of this paper. What I want to focus on is the use of intense laser irradiation as it relates to structure and mechanical properties.

Lasers generate shock waves in materials by ablation of surface material. Although a number of methods exist that couple laser energy into solids, I will confine these remarks to the case of direct illumination of the target sample by laser energy. When high-power laser energy is absorbed, the irradiated area rapidly reaches very high temperatures, as high as 1 keV. This causes extremely rapid expansion of the surface material, and momentum conservation implies a strong disturbance which travels into the solid material. The details of laser-matter interactions are described in detail in texts on inertial-confinement laser fusion, and I will present just a brief synopsis relevant to the current topic. Other methods of rapid local energy absorption, such as electron beams and heavy ions, have a similar effect on solid targets.

Referring to Fig. 1, we assume that the laser has been incident on the target for some brief time, say 1 ns. In addition, we have assumed that the laser is incident on the target from vacuum. The laser energy in absorbed in a continuous fashion in the expanding plasma up to the critical surface, defined as surface for which the plasma frequency is equal to the laser frequency. At the critical surface, the electron density is

\[ n_e = \frac{\pi e^2 m / e^2 \lambda^2}{\sqrt{2 \pi m}}, \]

where \( m \) is the electron mass, \( e \) its charge, and \( \lambda \) the laser wavelength. The parameter \( n_e \) is of fundamental importance in the study of laser-matter interactions. Energy from the absorption region is transported into the target by electron thermal conduction. Deeper into the target is the so-called ablation front, where the sign of the material velocity changes sign. In other words, on one side of the ablation front the material is expanding away from the target; on the other side the material is being compressed and is moving in the opposite direction. The compressed material generally forms a strong shock wave. A simple way of viewing this is using a rocket model, in which the expanding material is the exhaust (with a velocity \( > 10^6 \text{ km/s} \)), and momentum conservation leads to a very large local thrust on the solid target.

For a typical Nd:YAG laser used in high-power applications, with \( \lambda = 1.06 \mu \text{m} \), \( n_e = 10^{21} \text{ cm}^{-3} \). This is substantially less than solid density, typically on the order of \( 10^{23} \text{ cm}^{-3} \), which has several implications. Thermal conduction between the critical and ablation regions is less efficient the longer this distance is, or the lower the value of \( n_e \). In addition, a
Figure 1: Schematic of the laser-matter interaction generating a shock wave. The critical surface is the region in the expanding plasma for which the electron plasma frequency is equal to the laser frequency. At the ablation front, the local velocity vector changes sign, i.e., to the left the material expands into the laser-plasma absorption region, to the right the material is in compression. The compression generates a shock wave in the solid material at a depth deeper than the ablation region.

A variety of laser-plasma interactions can occur in the underdense plasma ($n_e < n_c$) which can generate highly energetic electrons which penetrate and "preheat" the sample. In general, higher shock pressures and less preheat over longer times are obtained with shorter wavelength laser irradiation; this is often obtained using harmonics of Nd-doped hosts.

The pressure obtained in this way can be described over wide ranges of intensity with a simple scaling relationship. A useful rule of thumb is $P \approx 40(I_{15}/\lambda)^{2/3}$, for pressure $P$ in Mbar (100 GPa), laser wavelength $\lambda$ in $\mu$m, and intensity $I$ in units of $10^{15}$ W/cm$^2$. From this we can see that an intensity of about $4.2 \times 10^{12}$ W/cm$^2$ will generate an ablation pressure of 1 Mbar. This is rather easily achieved with today's highly efficient lasers; for a spot diameter of 100 $\mu$m and a pulse length of 2 ns, a pulse energy of only about 0.7 J is needed.

However, an additional important factor must be noted. The expansion of the material from the ablation front and through the critical surface can cause the distance between them to increase with time, lowering the pressure at the ablation front. Even a relatively constant intensity pulse will produce a decaying shock. After the laser decreases in intensity the ablation pressure also decreases. These effects, as well as two-dimensional effects arising from the plasma expansion, lead to a strongly time-dependent ablation pressure. When the pressure falls, a rarefaction wave travels towards the shock front, eventually overtaking it. All these effects tend to produce a decreasing pressure at the shock front as it travels. It is, in principle, possible to overcome all but the last effect with appropriate temporal shaping of the laser pulse. A rather steady shock can be produced over a time about equal to the half-width of the laser pulse. This time dependence is in strong contrast to impact-generated shocks, which tend to be constant in amplitude over $\approx 1 \mu$s intervals. We hasten to point out that side rarefactions are inevitable in experiments using small spatial scales, so that eventually the shock will become spherical and rapidly decreasing in amplitude.

The effects of material expansion can be alleviated for low-intensity pulses ($\approx 10^{10}$ W/cm$^2$) by using a transparent material overlaying the sample which has a high optical damage threshold.

**Effects of scale**

The short time scale of laser experiments enhances the importance of kinetic effects. The strain rates are of the order of $10^{15}$ and temperatures are quenched at roughly $10^{12}$ K-s$^{-1}$. This makes it possible, and indeed simple to quench metastable phases. The question arises, of course, as to whether sufficient time exists to form metastable phases! This will be discussed in the next section on materials applications. The spatial scales are also important, since laser spot sizes can be made as small as about $2 \lambda$, the diffraction limit. This opens up the possibility of producing shocks in a single grain in polycrystalline materials. Of course, the axial spatial extent (along the laser axis of symmetry) of such effects is also limited, since the shock will become spherical after moving about one spot diameter.
Materials applications

One of the most attractive features of laser-shock waves in novel materials is the fact that the local energy density is small enough that sample recovery is trivial for massive target samples. Also, the repetition rate can be as short as fractions of a second to just a few minutes. This means that low-energy high-repetition-rate experiments can exploit signal averaging. Even high-power experiments can be carried out at a rate that is orders of magnitude higher than impact experiments. Unlike most high-pressure experiments, the data rate can far exceed our ability to analyze it.

I think that laser will find two main uses in novel materials applications. First, the laser is ideal for the study of microscopic effects in solids. The way in which solids react to strong shock waves is still an area with little direct evidence at small-length-scales. If we wish to tailor the behavior of materials, it is essential that we understand the effects at the atomic and mesoscopic levels. The laser may be particularly useful for this, because the scale of the experiments make possible unique diagnostic measurements. For example, x-ray diffraction may be used to study plastic effects and the kinetics of phase transitions. Here that fact that the targets are thin and the relative ease of timing the diffraction measurements with the laser pulse are crucial. This is an area that is just beginning to be explored and deserves much attention. We have yet to use such techniques to understand melting. We still have little atomic and mesoscopic experimental understanding of defect generation and motion. It is critical to make real-time in situ measurements. This is difficult in metals, for x-rays are one of the only ways to study the interior. These are sensitive to the bulk crystal structure, but are difficult to interpret on the mesoscopic level. An alternative may be to use spectroscopic methods such as fluorescence to investigate shock-generated defects in wide-band-gap insulating crystals. Materials such as SiO2, Al2O3, and LiF are outstanding candidates which exhibit contrasting bonding types. Spectroscopic methods have been used in ruby to determine the local strain environment in compression and tension. These crystals could be studied over wide ranges of pulse length, pressure, and time-varying loading. And these studies should be carried out with previously defected or shocked materials. The main challenge here is to design well-posed experiments. Another potentially rewarding experiment will be to characterize the recovered material as distance from the incident laser increases. A wide variety of effects will be in evidence, since we can, in a single experiment, reach all states from plasma, through melt, to very weak shocks, with a variety of loading and unloading rates.

The small scale of laser experiments is a two-edged sword. It will be challenging indeed to determine local properties in regions which are small and are surrounded by potentially large gradients. Diagnostic access is limited to a cone surrounding the laser-axis, and crossed beams will be necessary to locate a position in space. This suggests that even with lasers, there are simplicities associated with scale. Larger scales may allow moving the experimental region well away from the interaction region, and with steadier pulses the situation will be improved. However, in the case of long-time kinetic effects, I think that impact experiments will have the major role. Here the difficulty is to synchronize short-pulse experiments with essentially asynchronous impact events.

At the next level of complexity are experiments at the level of polycrystalline samples. Short pulses can be used, but the very large lasers such as NOVA at LLNL or Gekko-2 in Japan can be used to study really macroscopic materials. For example, we can generate 50 GPa in a typical solid over a 1 mm diameter area for 10 ns using just 4 kJ of laser energy. This is well within the range of today's large lasers. Experiments up to 1 ns duration could be done on a table-top! Here the interest will be in longer times so that diffusion-limited effects such as precipitation at grain boundaries become evident.

Up to this point, I've been discussing basic materials science applications. Now it is time to consider the problems and challenges of new materials. I mentioned above the problem of kinetic effects in the problem of metastable materials. While it is certainly clear that melting may occur quite rapidly, and in fact some structural transitions occur quite rapidly un-
Under shock, we still don't know enough to predict what we can make. This is perhaps one of the key issues. Certainly, grain sizes in resolidified material can be quite small, potentially leading to novel plastic properties. It is probable that rapid solid-state chemical reactions will be possible, given the high energy density available. The question of magnetic materials is problematic, since even moderate shocks will raise the material above the Curie temperature.

When we do succeed in making a material with novel properties, the next question to naturally arise is scaling to industrial, or at least macroscopic quantities. The size of the laser experiments, and the typically short time scale will make this very challenging. If the special effects of short time scale are needed, it may be possible to design large scale impact with thin layers. If the effects of space scale are critical, then lasers may well have to be used. But, lasers other than Nd:YAG, such as KrF or CO2, may be needed. But beyond this, it is outside the scope of this report.

It is entirely possible that the rôle of lasers will be best for characterizing the details of small-scale processes, while impact and energetic materials will be the drivers of choice for making new materials in macroscopic quantities.

Summary

The laser-matter interaction responsible for shock wave generation has been briefly described, and a simple scaling relation for pressure in terms of incident energy was presented. We have described some types of laser-solid experiments needed for understanding dynamic material response for short time and high energy environments. In addition, some indications of needed novel materials investigations have been listed. For lasers, these will possibly in the area of basic materials science in service of novel materials preparation.

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References
