Relative Consistency of Equations of State by Laser Driven Shock Waves

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An experiment shows that equations of state of solid matter at pressure $P = 10$–50 Mbar can be studied by using lasers with pulse energy $E \approx 100$ J. Laser beams smoothed by phase-zone plates produced high quality, planar shock waves in two-step, two-material targets, allowing simultaneous measurements of the shock velocities in the two materials. By the use of the impedance-matching technique, the relative consistency of the equations of state of these materials can be tested, or a relative equation of state data can be measured. Pressures higher than 35 Mbar were achieved in gold.

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A knowledge of the equation of state (EOS) of materials at pressures in excess of 10 Mbar is important in several branches of physics [1], including astrophysics and inertial confinement fusion research. Hydrodynamic and thermodynamic codes usually employ EOS data (such as, e.g., those provided by the SESAME tables [2]) which were mainly produced by calculations using theoretical models. Simpler model EOS's are also widely used [3]. Only a few experimental data (see [1,4,5], and references therein) are, however, available to validate EOS calculations and discriminate between different theories in this high pressure regime.

Pressures above a few Mbar can only be achieved through the use of dynamic shocks. EOS data in the tens of Mbar domain were produced by nuclear weapon driven experiments [4,5]. Even higher pressures can be generated in the laboratory by pulsed lasers [6]. In earlier experiments pressures up to 100 Mbar were reached by shock waves generated in a laser irradiated solid [7,8] or in a target foil impacted by a laser accelerated foil [9]. However, such shock waves were not adequate for accurate EOS measurements, where spatial uniformity, constant velocity, and low preheating of the material ahead of the shock wave are essential [6,10]. Recently, two experiments have demonstrated high quality, planar shock waves induced by laser generated thermal radiation (indirect laser drive). Pressures as high as 750 Mbar were achieved using laser pulses of 25 kJ (at wavelength $\lambda = 0.53$ μm) and a foil impact technique [10]. In another experiment, 20 Mbar planar shocks were produced by employing 2.2 kJ laser pulses [11].

It should be observed that in all the quoted laser driven experiments the generated pressure was determined indirectly by the measurement of the shock velocity D and the use of an EOS. A direct measurement of an EOS point, instead, requires the experimental determination of an additional parameter of the shocked material [12], such as, e.g., the fluid velocity U, as was done in some nuclear explosion driven experiments [5]. Recently, the feasibility of such simultaneous measurements of D and U in shock wave experiments driven by laser generated thermal radiation has been proved [13]. However, a very high power laser is required, in order to maintain a constant ablation pressure for a sufficiently long time (a few nanoseconds). An intermediate approach between the usual indirect method of pressure determination and the direct measurement of an EOS point is possible (and, in fact, was used in accurate nuclear explosion driven experiments [4]). The method is based on the impedence-matching technique and consists of measuring, on the same shot, the shock velocity in two different materials. This allows for testing the relative consistency of the EOS of the two materials, or, alternatively, for measuring an EOS point for one of the materials, using the EOS of the other material as a reference. (In passing, we notice that the impedance-matching technique was used largely in the past to intensify laser driven shock waves [8].)

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In this Letter we report the first demonstration of such a technique at pressure $P > 10$ Mbar (actually up to 35 Mbar in gold), using direct-drive laser irradiation, and two-step, two-material targets. The experiment was performed at the Laboratoire pour l'Utilisation des Lasers Intenses (LULI), Ecole Polytechnique. Experiments employing the same principle were performed in the past [14], but the achieved pressure was well below 10 Mbar, and the errors in the measurement of the shock speed were of the order of $\pm 10\%$. Instrumental to the success of the present experiment was the production of a smooth, nearly one-dimensional shock wave [15], obtained by the use of the beam smoothing technique based on the so-called phase-zone plates (PZP) [16]. As a result of the good coupling of laser energy to matter (a feature intrinsic in the direct-drive approach at submicron laser wavelength) and of the reduced level of preheating (as compared to indirect drive), it has been possible for the first time to perform EOS studies at pressure $P = 10$–35 Mbar by using laser pulses of relatively modest energy ($E \approx 100$ J at $\lambda = 0.53$ μm).

Our experiment is based on the impedance-matching technique [12] applied to a double-step target, with the structure sketched in Fig. 1. The target is made of a "base" foil made of a material A, which is irradiated by the laser on one side, and supports, on the opposite side, two steps made, respectively, of the same material A and of a different material B. Using rear-face, time-resolved imaging we experimentally determine the velocity of the shock propagating through the two steps D_A and D_B (corresponding to particle velocities U_A and U_B), respectively. If the EOS's (and hence also the Hugoniot curves) of the two materials are known, it is then possible to test their relative consistency. It requires then the coincidence of the experimental point (P_B, U_B) and of the one (P'_B, U'_B) deduced from (P_A, U_A) by using impedance-matching equations associated to EOS tables. Alternatively, a relative EOS measurement can be performed for material B by taking the EOS for material A as a reference [12].

EOS experiments aim at discriminating between different theoretical models. In most cases the pressure deviations between the models do not exceed 10% [6], which sets an upper limit of about 5% to the experimental accuracy required in the measurement of the shock velocity. In our case there are three main sources of possible errors in the determination of D: the quality of the shock itself (requiring flatness over a wide region), the sweep speed (ps/mm) of the streak camera, and the knowledge of the step thicknesses.

In our experiment the shock emergence from the target was inferred by detection of the emissivity of the target rear face in the visible region. This was imaged by a photographic objective onto the slit of a visible streak camera with 5 ps time resolution. We performed the calibration of the streak sweep speeds with an etalon made up with a series of short laser pulses (FWHM = 100 ps). The relative error in the speed used for our experiments was lower than 1%. The system magnification was $M = 22$, allowing a 5 μm spatial resolution, which was checked by imaging a suitable grid. We note that, according to diffraction theory, the elementary speckles produced by the PZP's are smaller than the quoted resolution, and cannot be evidenced in our experimental results.

Three of the six beams of the LULI laser (converted into its second harmonic, $\lambda = 0.53$ μm) with total laser energy $E_{2\omega} \approx 100$ J were focused onto the same focal spot. The temporal behavior of the laser pulse was Gaussian with a FWHM of 600 ps. A fourth beam, also converted into 2ω, was used as a temporal fiducial. Each beam had a 90 mm diameter and was focused on target with an $f = 500$ mm lens. An equivalent plane diagnostic was implemented for controlling the laser focal spot. The system employed an imaging objective (Olympus 50 mm, 1/1.2, the same as the one used to image the target rear side onto the streak camera slit) and a charge-coupled-device (CCD) camera (12 bits, 512×512 pixels). A removable mirror allowed the image to be directed to the CCD or to the streak camera, so that we had an almost "on line" control of the focal spot shape, which could be easily checked between two shots on targets. Finally, an active x-ray pinhole camera looking at the target on the laser side, at 22.5° with respect to the laser beam plane, was used to check plasma formation and to image the focal spot in the x-ray domain.

In the optical smoothing by "classical" random phase plates (RPP), the envelope of the focal spot is given by the diffraction on the elementary pupil of this plate (square, hexagon, etc.). This generally produces a spatial distribution of the laser energy close to a Gaussian curve (in fact, $\sinh^2$ for a square pupil). In the case of our experiments, such a distribution would entail an increase of uncertainties on the results of measures due to variations of intensity, and hence of shock velocity, across the focal spot. In order to produce an uniform spatial distribution, instead of RPP's, we have used the

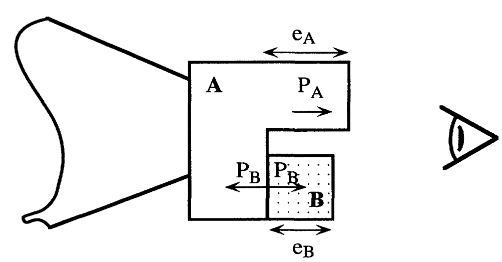

FIG. 1. Double-step targets with common base and two steps of different materials, A and B. From the shock traveling time in these steps, obtained with a visible streak camera, the shock velocities, D_A and D_B, are determined. P_A and P_B are the corresponding pressures.

optical smoothing technique of PZP, which is built on the association of the main lens with a Fresnel lens array having a randomly distributed phase shift (0 or π). Each lens of the array has consecutive elements whose phase shift differs by π. The PZP's allow a flat top distribution of laser intensity to be produced at a given distance from the focal plane of the main focusing lens [16]. Note that, even if we were able to produce very high quality shock waves [15], our experiment conditions were not optimal for the application of the PZP technique due to the long focal length of the focusing lens (500 mm) and to the small laser beam diameter (90 mm) [16]. Indeed our PZP's design is a compromise between the laser intensities required to achieve the desired shock pressure ($P \geq 10$ Mbar in aluminum) and the size and flatness of the focal spots. In our case we had only five elements per Fresnel lens, which is not enough to have very sharp edge width. We were able to produce a measured focal spot consisting of a 200 μm top-hat intensity distribution with Gaussian edges. The total focal spot FWHM was 400 μm, which corresponds to a laser intensity $I_L \leq 10^{14}$ W/cm^2. Our results imply that even higher quality shock waves could be produced using larger beams provided by other existing laser facilities.

In order to reduce one of the possible sources of experimental errors, one needs high quality, well characterized targets with the structure described above. In addition to the accurate knowledge of the step thicknesses, sharp step edges are required. Also, the spacing between the two steps must be small compared to the flat top-hat portion of the laser focal spot. Our targets have been developed in collaboration with the Laboratoire des Cibles of the Centre d'Etudes de Limeil-Valenton. The fabrication of the target is made of three stages: first, the base material is deposited; a mask is then applied to this base order to deposit the first step. Then, a second mask is applied, which is mechanically and optically guided, to ensure that the steps do not overlap and that their separation is limited to no more than 50 μm. The second step is then deposited. The overall quality of the targets was checked by electron microscopy.

In the first set of experiments, we used stepped targets made of aluminum (base and one of the steps) and gold (the other step). The step heights were determined with an absolute error smaller that 0.05 μm. Since the thicknesses of the aluminum and gold steps were about 4 and 2 μm, respectively, this ensured a relative error of about 1% for the aluminum step and about 2% for the gold step. The EOS's of these materials have been intensively studied, and differences between either various EOS calculations or experimental results are within a few percent [17]. This allows us to check the overall sensitivity of the method.

Figure 2 shows a typical streak image of a two-step target. Here we can clearly determine the shock velocities in the two materials, the flat part of the focal spot being $\approx$200 μm while the spacing between the two steps

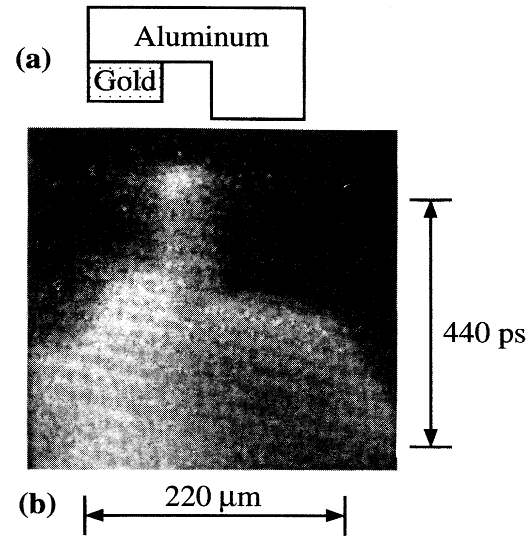

FIG. 2. Experiment on a double-step target. Optical smoothing is realized with PZP's giving a measured focal FWHM spot diameter of 400 μm. (a) Sketch of the target; the aluminum base thickness is 9.5 μm, the steps of aluminum and gold are 4.25 and 1.85 μm, respectively. (b) Streak camera record of visible light emitted by the rear side of the target.

is $\approx$45 μm. In order to check that the shock pressure is constant inside each step we performed laser shots on targets with different thicknesses of the aluminum base. 1D simulations, performed with the hydrodynamic code FILM developed at Ecole Polytechnique, have shown that in our irradiation conditions ($3 \times 10^{13} < I < 10^{14}$ W/cm^2) the base must be thicker than approximately 8 μm. We also checked [15] the quality of the shock wave generated with the PZP smoothed beams using simple foil targets; we found typical variations of $\pm$5 ps for the shock breakthrough time across the 200 μm flat region of the focal spot. In order to ensure that 2D effects are quite negligible under these irradiation conditions, the experimental results and the FILM simulations were compared with simulations made with the 2D hydrodynamic Lagrangian code DUED [18], developed at ENEA Frascati. With all of the above errors taken into account, the shock velocities were determined with a maximum error of $\pm$4.5% in aluminum and $\pm$6% in gold. These errors can be reduced to $\pm$2% by improving the target fabrication, by using a higher resolution streak camera, and by enhancing the step heights (time measurements errors on the streak signal being absolute). In this last case, an increase of laser energy (by a factor of 2–3 only) would be sufficient to ensure constant shock speeds in these higher steps.

In the case shown in Fig. 2, the experimental aluminum shock velocity is $D_A \approx 24.5$ μm/ns (corresponding to $P = 10$ Mbar), and the gold shock velocity is $D_B \approx 13.9$ μm/ns (corresponding to $P = 22$ Mbar). A summary of the experimental data is presented in Fig. 3,

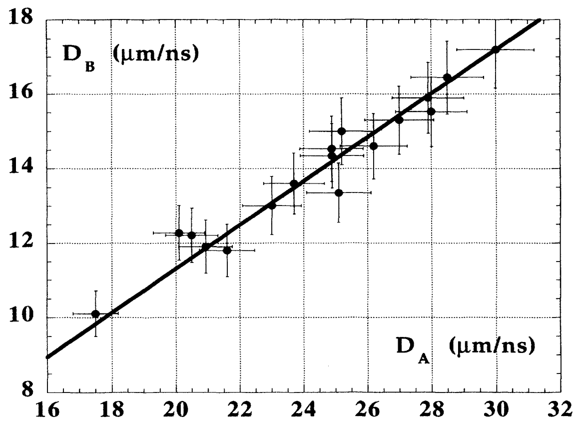

FIG. 3. Experiments with aluminum-gold targets. The shock velocity in gold, D_b, is plotted versus the shock velocity in aluminum. Error bars are 4.5% and 6% for aluminum and gold, respectively. The solid line represents the theoretical SESAME data.

where the shock velocity in gold D_B is plotted versus the aluminum shock velocity D_A. The corresponding pressures range from 4.5 to 16.5 Mbars in aluminum, and from 9.5 to 37 Mbars in gold, depending on the input laser energy. The data are in good agreement with the SESAME EOS [2] (solid line), with most points lying in a 3% wide error strip around the theoretical prediction.

In conclusion, the reported experiments have shown the possibility of obtaining quantitative measurements on shock waves in a solid sample directly irradiated by optically smoothed laser beams. The pressure regime $P = 10-50$ Mbar can be explored by employing laser facilities of relative small size ($E \approx 100$ J). By the use of PZP's, which allows the production of high quality, flat shock fronts, we have shown that the proposed two-step technique allows simple tests of the consistency of EOS tables. In addition, this technique allows relative EOS measurements of a material, assuming a reference EOS for the other target material. In particular, aluminum can be taken as a reference for pressures up to 25 Mbar [19], which allows EOS studies of high impedance materials up to 50 Mbars. Such a method could be applied [20], for instance, to a better knowledge of the doped plastics EOS (such as CH-Br) used in experiments on Rayleigh-Taylor instabilities [21]. Moreover, applications to astrophysical problems [22] are also conceivable by using suitable laser pulses.

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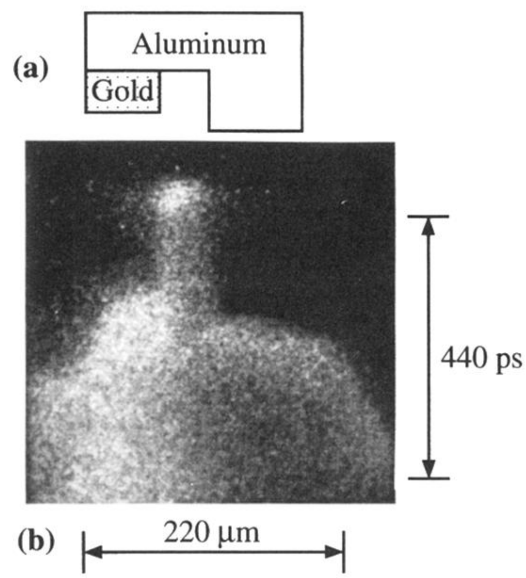

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