STIMULATED BRILLOUIN SCATTERING IN SHOCK-COMPRESSED FLUIDS*

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In this Letter we report the first observation of stimulated Brillouin scattering in a shock-compressed material. The experiment was carried out in liquid acetone shocked from a pressure of 1 atm and temperature of 20°C to a pressure of 35 ± 5 kbar, a temperature of 910 ± 30°K, and a compression of $31 \pm 4\%$. The experiment has yielded the first measurement of the acoustic properties of condensed matter in such a highly compressed state.

The experimental arrangement of optical components is shown in Fig. 1 and was basically similar to apparatus used in previous experiments.[1,2] The high-explosive shock-wave techniques are described in detail elsewhere.[3] The sequence of operations during the experiment was as follows: The three lasers were pumped simultaneously. After a preset time during which the lasers approached maximum inversion, the detonator was fired, and the explosive lens converted the point initiation into a plane detonation wave which passed into the sample as a one-dimensional shock wave. At the appropriate time, the Kerr cell was triggered by piezoelectric pins set in the sample. The light pulse from the trigger laser was directed at the bleachable filter. When the bleachable filter became saturated, the oscillator laser began to emit, producing a 20-nsec pulse of high spectral purity (several adjacent cavity modes). This pulse passed through the amplifier, and its final intensity was about 200 MW/cm^2. The light pulse was then focused behind the shock front in the acetone where stimulated Brillouin scattering occurred. The backscattered pulse, shifted down in frequency, was amplified in the oscillator-amplifier system and recorded, along with the initial laser pulse, on the oscilloscope and Fabry-Perot interferometer. The three-laser system was developed to provide timing to approximately 0.1 μsec, single-frequency operation, and high intensity. All three requirements are necessary in carrying out stimulated Brillouin scattering in a shock-wave experiment.

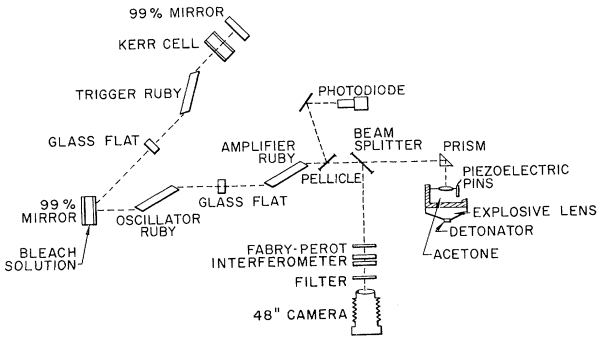

FIG. 1. Experimental apparatus.

The results are shown in Fig. 2. Figure 2(a) is a Fabry-Perot interferogram of the initial laser pulse, showing its spectral purity; Fig. 2(b) shows stimulated Brillouin scattering in the acetone sample at room temperature and pressure. The characteristic doublet can be seen, along with a very faint line which corresponds to second-order multiple Brillouin scattering. Figure 2(c) shows the result of the shock-wave experiment. Line A is the laser line. Line B is stimulated Brillouin scattering from unshocked acetone in front of the shock wave. Line C is stimulated Brillouin scattering from the shock-compressed acetone.

In a static experiment, the Brillouin-Stokes

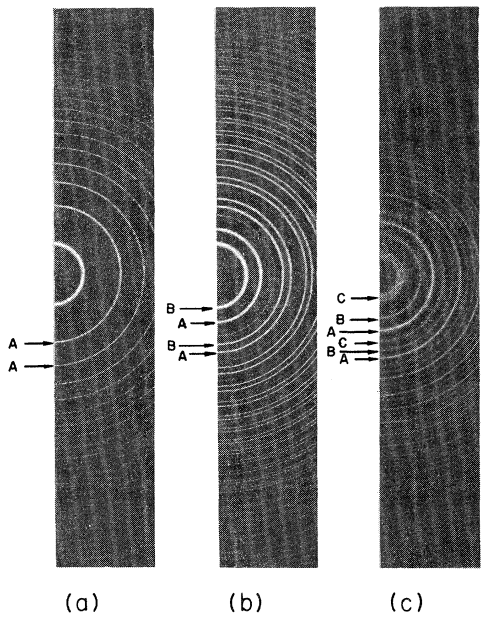

FIG. 2. Fabry-Perot interferograms of stimulated Brillouin scattering.

line B is shifted down in frequency by an amount given by the Brillouin formula,[4]

$$\Delta\nu = \nu_0 \frac{2nv_s}{c} \sin\frac{\theta}{2}, \quad (1)$$

with $\Delta\nu$ the downward frequency shift (which is also the frequency of the acoustic phonons generated), n the index of refraction of the liquid, v_s the velocity of sound in the liquid, and θ the scattering angle. In this case $\theta = 180°$ and $\sin\frac{1}{2}\theta = 1$.

In stimulated Brillouin scattering behind a shock front, the shifted line, C, has undergone three Doppler shifts: one shift due to the acoustic velocity of the stimulated phonons, given by Eq. (1); one because the light wave must cross a moving interface between two media of different refractive indices; and one because the material behind the shock front is moving at a constant velocity, U_p, with respect to the laboratory coordinate system. The total frequency shift, $\Delta\nu'$, experienced by line C, therefore, is given by

$$\Delta\nu' = \frac{2\nu_0}{c}\{n_2 v_s + (n_2 - n_1)U_s - n_2 U_p\}, \quad (2)$$

with $\Delta\nu'$ the downward shift in frequency, n_2 the refractive index in the shocked medium, n_1 the refractive index in the unshocked medium, U_s the shock velocity, and U_p the particle velocity behind the shock front.

The shock velocity was measured by piezoelectric pins placed at known locations in the sample. Particle velocity was calculated from a shock impedance-matching solution with the container by the usual techniques.[3] The refractive index was calculated from the Lorentz-Lorenz equation, which has been shown to be fairly accurate (better than 5%) for shock-compressed liquids at pressures less than about 50 kbar,[5] although failing badly at much higher pressures. The shift of line C corresponds to a phonon frequency of 13.9 GHz and an acoustic velocity of 2.9 ± 0.3 mm/μsec, and agrees, within experimental accuracy, to the calculated value of 3.1 ± 0.4 mm/μsec obtained from hydrodynamic codes. A quantitative comparison of theoretical predictions with the results of more precise experiments currently being carried out will be published elsewhere. Figure 3 is a time-resolved photodiode record of the sequence of events during the experiment, and shows that line C cannot be due to multiple-stimulated Brillouin scattering in the unshocked fluid. The initial laser pulse and the amplified Brillouin pulse are recorded, with a superimposed ripple due to beating of adjacent cavity modes. The very intense second pulse is the sum of both Brillouin scattered pulses, corresponding to lines B and C in Fig. 2(c). These two scattered light pulses originate at essentially the same point in space, and hence the same point in time. A very weak third pulse can be seen which corresponds to higher order multiple Brillouin scattering from the unshocked material. This faint pulse was also present in the photodiode record from the experiment performed under static conditions.

It can be seen from Fig. 2(c) that line C exhibits considerable fine structure. This is probably due to "mode pulling," a phenomenon first described by Brewer.[6] The backscattered light is not amplified in the oscillator at its own central frequency, but at the central frequency of one or more of the adjacent oscillator cavity modes. The Brillouin-scattered light from the unshocked acetone returns as a very narrow line; this line is then "pulled" to the nearest cavity mode. The Brillouin light scattered from behind the shock front is spectrally broadened, because of the steep pressure gradient behind the shock front at the focal point, and upon its return to the oscillator is amplified at frequencies corresponding to several adjacent cavity modes. The spacings of the fine lines in line C were measured and correspond to the cavity modes of the oscillator laser used in this experiment.

The current series of experiments is directed toward developing and testing improved equations of state for highly compressed fluids. With shock-wave techniques, it is possible to achieve temperatures up to several thousand degrees Kelvin and pressures up to one million atmospheres. It is necessary to use shock-wave techniques to carry out these investigations, since at room temperature most liquids freeze at about 30 kbar.

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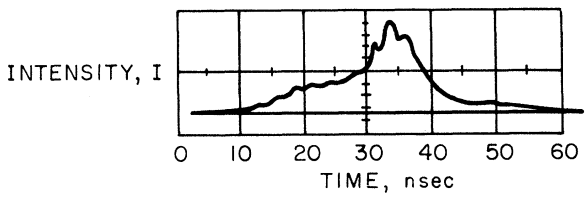

FIG. 3. Photodiode record of shock experiment.

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PARAMAGNETIC RESONANCE TRANSMISSION IN GADOLINIUM*

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Using the spin transmission or selective transmission resonance method previously applied to conduction-electron paramagnetic resonance,[1] we have observed rather unexpected but unique and characteristic resonance lines transmitted through 99.9% purity gadolinium foil at temperatures above the Curie temperature of 289°K. In this type of resonance technique, the sample forms the common wall between two microwave cavities tuned to the same frequency and so arranged that the external magnetic field may be oriented either perpendicular or parallel to the foil with the microwave magnetic field perpendicular to the static field. One cavity is used for excitation of the resonance, and the other cavity, into which leakage must be kept to a minimum, is connected to a sensitive superhetorodyne receiver which is coherent with the excitation generator and therefore provides phase-sensitive detection.

The sample foil is 75 μ thick, and the skin depth at the frequency used in this experiment (9200 MHz) is about 1 μ. Transmitted paramagnetic resonance under these conditions is surprising. If it is assumed that a signal is carried by independent motion of conduction electrons, a conventional estimate of spin relaxation due to spin-orbit[2] or spin-ion core[3] interaction leads to too short a relaxation time to make such a resonance observable. If one assumes that the paramagnetism above the Curie temperature is due only to statistically oriented paramagnetic ion cores with atomic-range order, it is clearly impossible to transmit a coherent phase resonance through a sample thicker than a skin depth. The alternative, which the shape of the resonance seem to corroborate, is to invoke superexchange or itinerant magnetic order with longer range than might be expected.

Figure 1 shows the experimental phenomena when the static magnetic field is parallel to the surface of the gadolinium sample. The bottom two traces show a paramagnetic-resonance absorption signal observed by reflection from the excitation cavity only. Trace (d) shows this strong signal by direct power measurement on a crystal detector, and trace (c) shows the same resonance using field modulation and a lock-in amplifier which gives the derivative. [The signal-to-noise of (c) is poorer than (d)

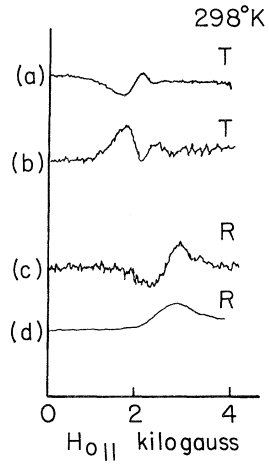
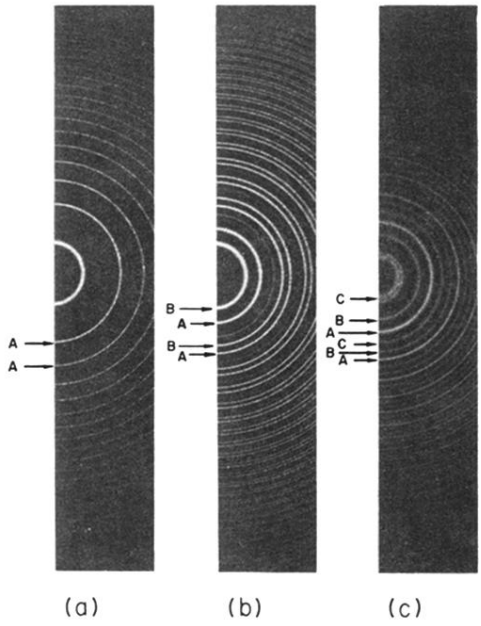


FIG. 1. Resonance traces in 3-mil gadolinium foil at 298°K with static field parallel to sample. (a) and (b) Transmission resonances at two phase settings. (c) Derivative of reflection signal. (d) Direct reflection signal.



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