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Impulsive stimulated light scattering from opaque materials at high pressure

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Abstract

Recent progress in the application of impulsive stimulated light scattering to opaque materials under high pressure is reviewed. Measured elastic constants and sound velocities of polycrystalline hcp ε-iron to 115 GPa are presented.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

A number of experimental techniques are now available for measuring the elastic properties of metals under very high pressure. With the emphasis on iron these include shock wave compression [1], directionally [3, 4] and temperature [5] dependent x-ray diffraction (XRD), nuclear resonant inelastic x-ray scattering (NRIXS) [2, 6, 7], inelastic x-ray scattering (IXS), [8] and Raman spectroscopy [9, 10]. From both a theoretical and experimental point of view, these techniques possess a wide variety of advantages and drawbacks. These may relate to the sample requirements, the maximum pressure attainable, the experimental precision, the cost and complexity of the apparatus, the assumptions involved in extracting useful information from the measured data, etc. A technique that has proven very useful for determining elastic properties of transparent materials in the diamond anvil cell (DAC) is Brillouin light scattering (BLS). Measurements have now been made to at least 70 GPa in the DAC using this technique [11]. Since it is an inelastic visible light scattering process, Brillouin scattering allows the direct measurement of the speed of propagation of acoustic waves. The experimental apparatus is modest and relatively inexpensive. However, it is difficult to apply BLS to opaque materials in the DAC [12]. This difficulty is largely a consequence of the greatly reduced interaction volume compared with transparent materials. The intensity of scattering from bulk acoustic waves can be orders of magnitude greater than that of scattering from surface acoustic waves (SAWs). It is likely, therefore, that the latter signal may be swamped by scattering from the transparent pressure medium or diamond anvils (via multiple different scattering geometries).

2. Impulsive stimulated light scattering

The technique of impulsive stimulated light scattering (ISLS), while possessing many of the advantages of BLS, offers, in principle, a single, well defined, scattering geometry, as well

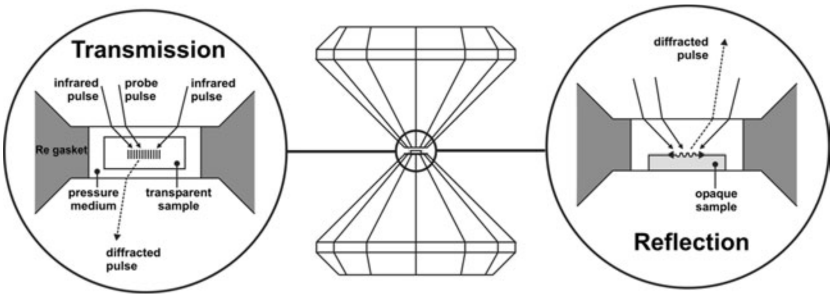

Figure 1. The geometry of the ISLS process in the context of the diamond anvil cell.

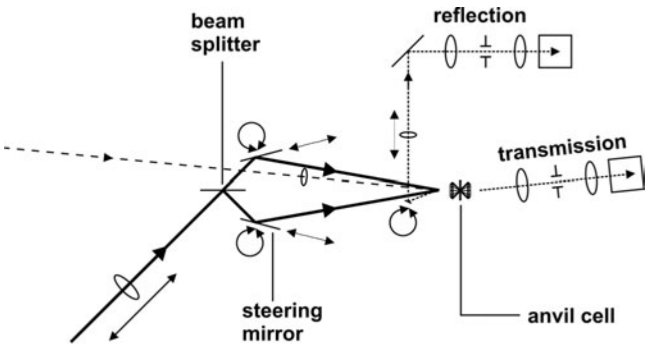

Figure 2. Schematic of optics near the sample position. The thick solid lines represent the path of the excitation pulse(s). A single 1064 nm pulse is split and then recombined in the sample. This symmetric arrangement guarantees temporal coincidence. The steering mirrors may be rotated and moved to allow adjustment of the grating wavevector. The long-dashed line represents the path of the probe pulse after exiting the mechanical delay line (not shown). The short-dashed lines represent the paths of the diffracted signals from opaque ('reflection') and transparent ('transmission') samples.

as a very large signal strength, even in the case of metals [13]. ISLS can be considered to be a light-induced ultrasonics process. In contrast to BLS, the acoustic wave is artificially stimulated [14]. In general, two laser pulses are combined in the sample at a known angle of convergence, thereby giving rise to an interference pattern (figures 1 and 2). Optical absorption, in turn, results in a regular array of hot regions that rapidly expand, and thus coherently excite counter-propagating acoustic waves. To an incident probe beam the acoustic waves appear as diffraction gratings (the exact nature of the coupling depends on the optical properties of the material). Moreover, the total diffracted signal is modulated at some characteristic frequency or frequencies, due to phase differences in the individual diffracted components. Two counter-propagating acoustic waves, for example, would produce a modulation of the signal at twice the acoustic frequency, while a single acoustic wave, together with the static thermal grating (which persists for some time after formation, depending on the thermal transport of the sample medium), produces modulation at exactly the acoustic frequency. To obtain the velocity of the acoustic waves, the common wavevector (which is determined by the convergence angle of the excitation pulses) is combined with the measured frequency. In our system, pulses (of duration $\sim$100 ps) of the first and second Nd:YAG harmonics are used to excite and probe the gratings, respectively. We do not examine the evolution of the gratings in real time (although this can be done in some cases [15]). Rather, the points making up the measured time series (figure 3)

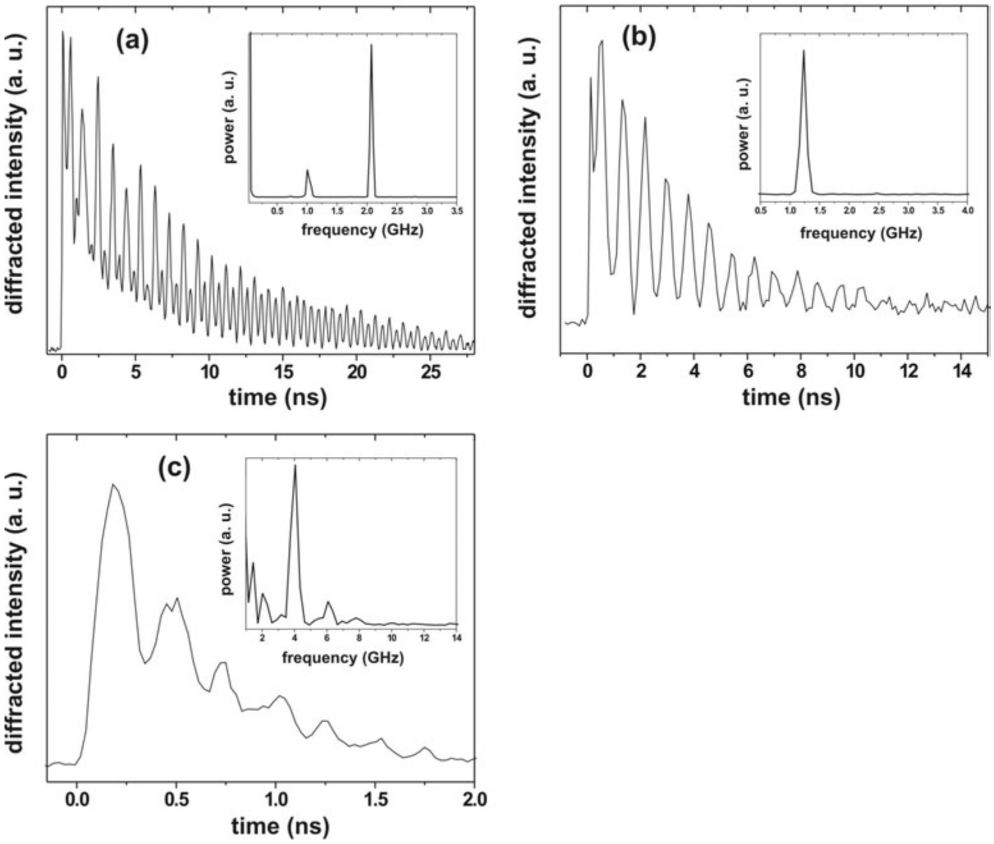

Figure 3. Time series obtained in three different situations: (a) a single crystal of (100) germanium in air with a wavevector along [001], (b) a single crystal of (100) iron in argon at 9.5 GPa with a wavevector along [001], and (c) polycrystalline hcp ε-iron in contact with diamond at 115 GPa. Note the different timescales. The insets are the corresponding power spectra (obtained by taking a fast Fourier transform with a Hanning window). In the case of germanium the Rayleigh frequency and its corresponding double are clear (see the text). In the other two examples the wave in question is essentially a leaky Stoneley wave. Rotation studies on iron single crystals to ~11 GPa have allowed us to determine the elastic constants of iron close to the bcc–hcp transition (to be reported elsewhere).

are due to many thousands of separate grating excitations, with the arrival of the probe pulse in each case increasingly delayed with respect to the excitation pulses. The rate at which gratings are produced is limited by the laser repetition rate (up to 4 kHz in our case). The corresponding time interval ($>$250 μs) is much larger than the grating duration (typically less than 40 ns). The highest frequency we can excite and measure is therefore limited only by the laser pulse width. We have not found sample heating due to the excitation and probe pulses to be a significant factor in our measurements. This observation is supported by calculation [13], and has also been tested as far as possible by obtaining spectra at different incident powers and comparing frequencies thus obtained. We also believe that in the context of the DAC the contiguous transparent medium (see below), particularly in the case of diamond, acts as a 'heat sink'. In any case, we believe uncertainties due to, for example, pressure gradients to be much larger than those due to sample heating.

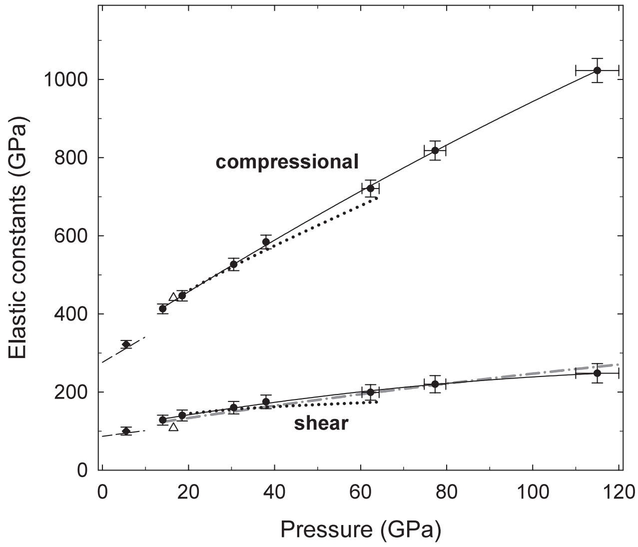

Figure 4. Shear and compressional aggregate elastic constants of polycrystalline Fe as a function of pressure (circles). The solid lines are guides to the eye (second order polynomials). The dashed lines are polynomial fits to IXS data [8]. The grey dashed-dot line is the results of Raman measurements [9]. The values at approximately 5 GPa (solid diamonds) were obtained in the polycrystalline bcc phase and are compared with the results of a neutron scattering study [25] (dashed lines). The agreement (within 3%) attests to the reliability of the ISLS technique. Open upwards pointing triangles: ultrasonic data [4].

3. Surface waves in the diamond anvil cell

Naturally, SAW propagation in the DAC can only take place along a material interface formed by the sample in contact with the pressure medium, or other material such as the anvil. A difficulty that immediately arises is that the SAW velocity is determined not only by the sample but also by the contiguous material, whose acoustic parameters must therefore be known in order to extract those of the sample from the measured velocity. A further difficulty is that unlike a true SAW (e.g. the familiar Rayleigh wave), the wave excited at an interface typically does not have an infinite lifetime, and its acoustic energy radiates rapidly into the surrounding bulk material. (A wave whose energy flow is strictly parallel to the interface is called a Stoneley wave. It exists only when restricted matching conditions of acoustic parameters are met. See [16] for a discussion of the mathematical formalism we have used to calculate interfacial SAW velocities in terms of acoustic parameters and propagation direction, and also [15].) Experimentally, this results in a less well-defined frequency that limits the experimental resolution. Nevertheless, a judicious choice of the additional material may greatly reduce the severity of these problems.

Our research has included measurements on both single crystal and polycrystalline metals under quasi-hydrostatic conditions, and also on polycrystalline metals under non-hydrostatic conditions. In the case of single crystals, the DAC is rotated about its axis in order to determine the dispersion of the surface waves as a function of direction. Figures 3–5 display some of our results.

4. Conclusions

We intend to extend our measurements of the elastic properties of metals to the 1.5 Mbar regime, and also to combine ISLS with laser heating. We suggest that in the latter context

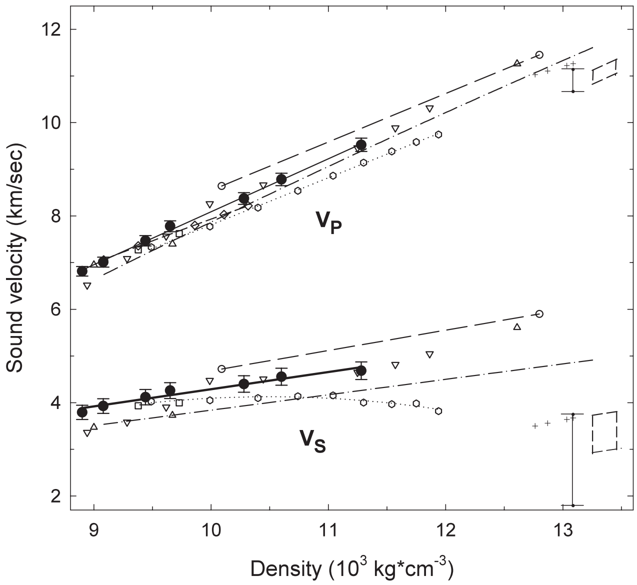

Figure 5. Shear and compressional aggregate sound velocities of ε-iron as a function of density. The solid circles and lines are our data for Fe, with the density calculated according to [24]. Open hexagons: shock wave data [1]. Open upwards pointing triangles: radial diffraction and ultrasonic data [4]. Open downwards pointing triangles and open squares: NRIXS data of [2] and [7], respectively. Open diamonds: IXS data [8]. Dash-dotted lines represent the results of x-ray diffraction study [5]. Open circles and long dashed line: first principles calculations [19]. The results of other calculations [18, 20–23] are close to these data. Crosses: seismic data [26]. Boxes of short dashed lines (see [27] for the temperature correction of the density) and error bars represent bounds for the temperature-corrected (at 4700 and 6700 K) extrapolation of our data to inner core conditions. The short dashed lines and the error bars represent the results of assuming empirical and theoretical results, respectively. These results will be reported in full elsewhere.

ISLS may have the advantage that only a very small sample volume would be involved, and problems of thermal gradients and stability would presumably be reduced.

Another property that may potentially be extracted from ISLS time series is the thermal diffusivity (determined by the lifetime of the static thermal grating). This has been done in the anvil cell in the case of transparent materials [14], and in the case of metals under ambient conditions [17]. We are presently running finite element simulations to separate the contributions of the contiguous materials from that of the sample. Depending on the results of these simulations we may be able to determine the thermal diffusivity of iron in the megabar range. Such information should be of great interest to the geophysics community.

Acknowledgments

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