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Extreme dynamic compression with a low energy laser pulse

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Abstract. Here we review the scaling of laser drive energy with pulse duration for sub-ns dynamic compression experiments. Via a related scaling argument, we also show that the throughput of time-resolved pulsed x-ray dynamic compression experiments (such as those performed at x-ray free electron lasers) varies as the inverse square of the time scale of the experiment. The strong variation of throughput with the scale of the experiment should be a significant consideration in the design of such experiments – to obtain high throughput, the time scale of compression should be no longer than required (via material equilibration) to achieve the desired final material state.

Relatively low energy can be used for short time scale (< 1ns) compression experiments[1-5], enabling high repetition rates at average powers available from even commercial laser systems. As we show below, for time-resolved pulsed x-ray experiments, short time scale experiments at higher repetition rates (but the same average drive power) can obtain substantially higher x-ray scattering throughput than available to longer time scale experiments.

This is a great practical advantage of short time scale experiments. Multiple shot, high rate data acquisition is a necessary element of several broad scientific topics, including the characterization of protein structure and conventional time-resolved physical chemistry studies of transition states. Compared to single shot experiments, high data rate experiments can explore larger phase spaces of material behavior (over a wider range of control samples), can discern subtle changes in behavior (via high signal to noise obtained with averaging), can explicitly obtain statistical information relevant to error analysis, inhomogeneous variation of sample environments and stochastic material behavior, and can observe unlikely events (such as, potentially, the homogeneous nucleation of melting in a superheated metastable state).

Traditionally, laser driven dynamic compression experiments have been designed to approach 1D compression (to more easily interpret data) by employing near-planar compression profiles. Analytically, the deviation of an actual experiment from planar compression is quantified by[6,7]:

$$\left|\frac{\delta v}{v}\right| < \left|\frac{\delta r}{r}\right| \propto \frac{1}{A^2} \qquad 1)$$

where δv/v is the deviation of the compression wave speed (for a curved wave front) from the wave speed in the 1D case, δr/r is the fractional change in wave front curvature over the duration of the experiment, and A is the aspect ratio (typically the sample width/thickness). Critically, the deviation of the wave speed from that obtained in 1D compression is only a function of the aspect ratio of the experiment, and is independent of the time scale of the experiment. Thus, if the compression time scale is sufficient to obtain the final state of interest, *the deviation of the material state from what would be obtained in an ideal 1D experiment will be no worse than that obtained in a longer time scale experiment with the same aspect ratio*. Assuming similar final states are achieved, experiments at short time scales have the same deviation from planarity and exhibit similar systematic errors due to geometry as longer time scale experiments.

At constant aspect ratio, the volume of compressed material varies as the third power of the duration of the experiment – the thickness varies linearly with the compression time and, at constant aspect ratio, the two transverse

spatial dimensions of the experiment each vary linearly with the sample thickness (and the compression time) as well. For a given final state, the compression energy is roughly proportional to the compressed volume, so the compression energy also varies as the third power of the duration of the experiment. In particular, if a compression time of 1 ns is required to obtain a given final state, 1000x more energy than necessary would be required to compress to the same state over 10 ns. Typically, ~Mbar pressures can be obtained with <mJ pulse energy applied over some hundreds of picoseconds[1-7]. This pulse energy is readily available from common commercial chirped pulse amplification systems operating well into kHz repetition rates. As such, short time scale experiments are well suited to high repetition rate and low energy.

Further, the x-ray scattering throughput of high repetition rate compression experiments may be substantially higher than longer pulse experiments, primary due to the third power scaling of energy with drive duration shown above. The scattered intensity per shot increases at most linearly with sample thickness (which is set by the drive pulse duration), but the energy per shot increases as the third power of the sample thickness (at constant aspect ratio as described above). So, the scattered x-ray intensity per unit laser drive energy varies as the inverse square of the scale of the experiment. For a drive laser with a given average power at variable repetition rate, *a 10x shorter experiment will have 100x higher throughput* – this is a strong incentive to keep experiments as short as possible with a high repetition rate.

The scaling of other relevant experimental quantities also benefits from shorter time scales. Generally, low energy/high repetition rate experiments use less sample area per unit signal, generate less volumetric debris per unit signal, and, compared to high energy lasers with comparable average power, high repetition rate lasers (with commensurate smaller pulse energy) typically obtain better shot-to-shot repeatability. Effectively, higher thoughput can be obtained for short duration experiments because the probe overlaps a larger fraction of the compressed material volume. X-ray free electron lasers can obtain ~micron scale beam diameters – a scale comparable to the scale of sub-ns dynamic compression experiments. Finally, high repetition rate lasers also achieve higher average power overall than high energy, low repetition rate lasers due to thermal scaling issues in conventional high energy laser amplifiers.

Sub-ns drive pulses can be obtained with chirped pulse amplifier technology used in femtosecond Ti:Sapphire regenerative amplification, with the great added benefit that pulse shaping technology used for ultrafast lasers is mature, and laser systems which already achieve transformational specifications for high repetition rate dynamic compression experiments are commercially available.

While small scale experiments may still be subject to issues like shot to shot pointing jitter (due to small drive spatial profiles), this is typically accounted for in other contexts (such as biomolecule structure characterization) by characterizing the actual drive on a shot-to-shot basis, where a given diffraction pattern is directly correlated to the conditions of the sample at the probe position ("binning").

Summarizing, there are great practical advantages to using high repetition rate/low energy dynamic compression for the study of material equations of state (where possible) and transient states upon rapid compression.

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