Explanation of anomalous shock temperatures in shock-loaded Mo samples measured using neutron resonance spectroscopy

Damian C. Swift,* Achim Seifter, David B. Holtkamp, Vincent W. Yuan, David Bowman, and David A. Clark

Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

Neutron resonance spectrometry (NRS) has been used to measure the temperature inside Mo samples during shock loading. The temperatures obtained were significantly higher than predicted assuming ideal hydrodynamic loading, a discrepancy which we now explain. The effects of plastic flow and nonideal projectile behavior were assessed. Plastic flow was calculated self-consistently with the shock jump conditions: this is necessary for a rigorous estimate of the locus of shock states accessible. Plastic flow was estimated to contribute a temperature rise of 53 K compared with hydrodynamic flow. Simulations were performed of the operation of the explosively driven projectile system used to induce the shock in the Mo sample. The simulations, and related experiments, indicated that the projectile was significantly curved on impact, and still accelerating. The resulting spatial variations in load, including radial components of velocity, should increase the apparent temperature that would be deduced from the width of the neutron resonance by 160 K. These corrections are sufficient to reconcile the apparent temperatures deduced using NRS with the accepted properties of Mo, in particular, its equation of state.

A long-standing challenge in shock wave experiments on condensed matter is the difficulty of measuring the temperature of the shocked state before it is destroyed by release waves. Most measurements have been made using photon emission spectroscopy (pyrometry),[1,2] but many substances of interest to science and engineering (e.g., metals) are opaque in the relevant region of the spectrum: infrared through visible for shocks up to the terapascal regime. Emission from an opaque substance comes from material within the photon skin depth of the surface, which generally cannot be maintained at the pressure of the initial shock long enough to allow useful emission spectra to be collected. A transparent window can be placed in contact with the sample to maintain an elevated pressure at the surface until released from the free surface of the window or the projectile. However, the mismatch in shock impedance must be taken into account, along with the effect of heat conduction.

Neutron resonance spectroscopy (NRS) has been investigated as a fundamentally different technique for measuring the temperature inside a dynamically loaded specimen, irrespective of its photon opacity[3] though the initial experiments exhibited a large discrepancy from the temperatures expected, from subtle causes which are explained here. Nuclear resonances are characterized by the energy and linewidth in the rest frame of the nucleus, i.e., with respect to the speed of incoming neutrons relative to the nucleus. The resonance is manifested as the variation of attenuation with neutron energy. In a NRS measurement of a shocked sample, neutrons in a band of energies interact with a volume of material. The transmitted spectrum is a convolution of the resonance in the rest frame of a nucleus with the velocity distribution of the nuclei in the sample volume, which depends on the sample temperature. The specimen is chosen to have a measurable resonance within the energy range of the incident neutrons. The spectrum of neutrons is measured after passing through the specimen; the temperature can be inferred from the width of the resonance. The resonance is also shifted in energy by the net relative motion of the specimen with respect to the neutrons, so the spectrum can also provide a measurement of mean material velocity inside the sample: a shift in the centroid of the resonance. In contrast, almost all velocity measurements are made optically (usually by the Doppler shift) at the surface of the specimen.

To perform a single-shot NRS measurement of a shocked state, it was necessary to collect a statistically significant neutron spectrum within the time for which the sample was in the shocked state: on the order of 1 μs for well-understood shocks as generated by the impact of disk-shaped projectiles.[4] To achieve the necessary neutron intensity, it was necessary to design a dedicated ^{238}U spallation target and moderator, and to induce spallation neutrons using a pulse of 800 MeV protons which had been accumulated in the proton storage ring of Los Alamos' LANSCE accelerator.

In any shock loading experiment, a common difficulty is the synchronization of measurements with the shock event. It is particularly challenging to synchronize an impact event, which follows a long delay during acceleration and coasting of the projectile, with a diagnostic pulse from a particle accelerator. The latter is generally not designed to be triggered externally with a short latency or high *a priori* precision in delivery of the diagnostic pulse. For the trial NRS experiments, the projectile was accelerated by detonating solid chemical explosive, using the "Forest Flyer" design,[5] which gave an uncertainty in impact time from the trigger signal of $o(100)$ ns. The Forest Flyer produced a nonisotropic distribution of high speed fragments. In order to protect the LANSCE beamline from damage and breach of vacuum, the shock experiment was tilted so that the projectile and shocked region in the sample were inclined at 55° with respect to the neutrons. The inclination was important in interpreting the NRS measurements, as discussed below. The separation between the projectile and the target was 20 mm, not 15 mm as reported before.[3]

NRS experiments have been performed on the reaction

products of detonating chemical explosive, and on Mo, as it is a standard reference material for high pressure work.[3] The temperature inferred was significantly higher than expected from shock calculations using the best available equations of state for Mo.[3] The Mo was doped with 1.7 at. % of [182]W, and the projectile was accelerated using explosive drive. NRS temperature measurements were made on two nominally identical shock experiments. Assuming the shock states were identical except for a small pressure variation, these measurements could be combined with a root-mean-square uncertainty (product of the probability distribution function for each measurement), as was done previously: 875 ± 46 K. The separate measurements differed by around the sum of their neutron-counting uncertainties, which is consistent with the assumption made. Explosively driven experiments often exhibit some round-to-round variability. Variations in surface velocity of $O(10\%)$ were observed in similar experiments,[11] and impactor curvature or variations in the timing of the neutrons with respect to the shock may lead to variations in NRS temperature, as discussed below. Round-to-round variability results in a larger uncertainty. If present, it would place a weaker constraint on the measured temperature: 875 ± 100 K at 68% confidence level, based on the difference between and uncertainties of the two measurements. These uncertainties can be reduced in future NRS measurements. Nonidealities in the loading are more likely to cause an increase than a decrease in the apparent temperature, so the lower measured temperature may be more accurate. The equation of state (EOS) of Mo is a standard in shock experiments.[6] Although the shock temperature has never previously been measured, it is closely constrained by theory, and uncertainties in the EOS are believed to be too small to explain the discrepancy.[7] Here, we explain the temperature discrepancy by taking account of plastic heating in the sample, and by considering details of the shock loading system used, which induced perturbations in the shock state that affect the apparent material temperature as measured by NRS.

In an ideal planar impact experiment, the strain applied to the sample is uniaxial. Uniaxial strain applied to a solid induces shear stresses, and for Mo at the ~60 GPa pressures of the NRS experiments, the shear stresses induce plastic flow. Compared to shock temperatures in a material without shear strength, plastic flow causes additional heating. The previous theoretical temperatures[3] were calculated from the scalar EOS and neglected the effects of plastic heating.

We have calculated the shock Hugoniot of Mo with and without the contribution from plastic work. Plastic work increases the thermal contribution to the EOS, generally increasing the pressure for a given compression. The effect of material strength, i.e., of elastic stress and plastic flow, was treated self-consistently in a numerical solution of the Rankine-Hugoniot equations for shock compression.[8] This is necessary for a rigorous calculation of heating. Material strength was treated using the Steinberg-Guinan model.[9,10] The effect of plastic work was estimated to be 53 K at a shock pressure of 63 GPa,[11] a significant contribution, but not enough to reconcile the temperature discrepancy (Fig. 1). This analysis depends on the accuracy of the strength model for Mo at these pressures on the Hugoniot. Evidence for the
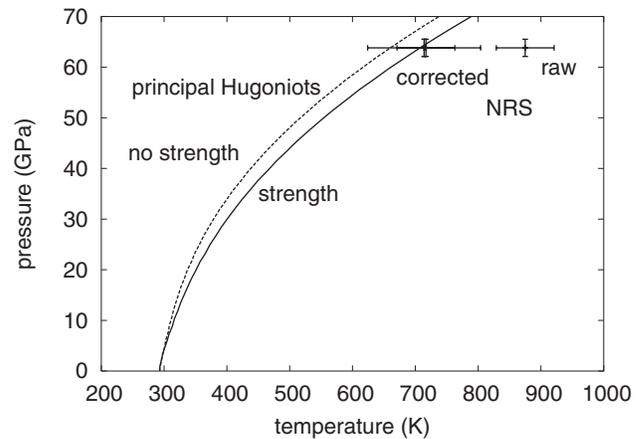

FIG. 1. Temperatures inferred from neutron resonance data, compared with the shock Hugoniot for Mo with and without strength. For the temperature corrected for shock curvature, the smaller error bar is the product distribution (identical state preparation); the larger is the sum distribution (variation in states).

magnitude of the strength is provided by surface Doppler velocimetry measurements made of these experiments: the onset of release from the peak velocity is marked by an elastic release wave of amplitude consistent with the flow stress calculated using the Steinberg-Guinan model.[11] The magnitude of plastic heating may be estimated simply by multiplying the elastic component of stress by the change in volume as the sample is compressed. At 63 GPa, the compression on the principal shock Hugoniot of Mo is 0.85, which is almost entirely plastic. Mo exhibits work hardening, pressure hardening, and thermal softening, so the integrated plastic work depends on the precise deformation history. However, Steinberg-Guinan flow stresses Y for Mo fall in the range $1.6-2.8$ GPa. Thus, without accounting for the precise deformation path, plastic heating should be approximately $60-100$ K.

Pyrometry measurements have been made of the temperature of Mo on release from shocking to similar pressures, with release into a LiF window (~25 GPa residual pressure) and into vacuum. These temperatures were also higher than calculations which neglect plastic heating. Pyrometry is prone to other systematic errors, such as thermal emission from the shocked window or from gas or glue compressed in the gap between the sample and the window, and enhanced plastic heating from the deformation of surface features such as machining marks. The total power in thermal emission varies with the fourth power of temperature, and pyrometry measurements are often more accurate at shorter wavelengths, where the power varies with higher powers of temperature. Pyrometry is, therefore, prone to inaccuracy from spatial or temporal variations in temperature, whereas NRS measures the average temperature. However, the inclusion of plastic heating in Mo brought the calculated surface temperatures into significantly closer agreement with the pyrometry data.[11]

The speed of the projectile was not measured in the NRS or pyrometry experiments, so the shock state could not be inferred directly from the published Hugoniot data for Al and

Mo. The shock pressure was inferred from the peak free surface velocity observed in each experiment, using mechanical EOS $p(\rho,e)$ derived from Hugoniot data.[10] For a given projectile speed or for a given free surface velocity, the shock pressure depends on the shear modulus and flow stress assumed in the projectile and sample. Thus, the "experimental" pressures as well as the calculated temperatures were adjusted when strength was included.

The pressures induced by the detonation of the chemical explosive in the Forest Flyer were much higher than the flow stress of the Al projectile. According to continuum dynamics simulations, the design used for the Mo NRS experiments was subject to hydrodynamic flows which deform and damage the projectile.[5] In particular, the case profile is likely to produce curvature of the projectile. The projectile would also be accelerating on impact. If the projectile is still accelerating, there must be a gradient of pressure and compression through it. On impact, these gradients induce a shock wave with a driving pressure which increases with time, leading to an increasing particle velocity. Unless this variation is taken into account, the broader peak could be attributed to a higher material temperature. If the projectile is not flat, and particularly if there are radial variations in its speed at the time of impact, there will be radial variations in pressure, temperature, compression, and particle speed. At different azimuthal positions in the sample, a radial component of particle velocity presents a different relative speed to neutrons traveling at an angle to the sample axis. This variation in relative speed will broaden the neutron attenuation peak, and appear as a higher material temperature, if not taken into account. Qualifying experiments were performed on the Forest Flyer system as used in the NRS experiments, but they were based on arrival time measurements and did not probe the detailed shape or density distribution of the projectile. Proton radiographs were subsequently obtained of a similar Forest Flyer design, initiated with a plane-wave lens, showing curvature of the projectile in close agreement with the simulations in the region of the projectile affecting the W-doped Mo.[5]

The simulations were used to estimate the variation of compression, temperature, and particle velocity in the doped Mo as a function of time. In principle, the time-dependent fields could be used to simulate the neutron attenuation with the time-dependent neutron spectra. A slightly simpler procedure was adopted here, giving the neutron attenuation as a function of energy at a series of instants in time.

Given the spatial fields of mass density $\rho(\vec{r})$, temperature $T(\vec{r})$, and velocity $\vec{u}(\vec{r})$ at some instant of time t, the spectral attenuation α was

$$\alpha(E) = \int \frac{\sigma f \rho(\vec{r})}{A} \frac{\exp\{-[(E'-E_r)/\delta]^2\}}{\sqrt{2\pi}\delta} d\vec{r}, \quad (1)$$

where E is the neutron's kinetic energy, σ the natural cross section, f the dopant mass fraction, A the atomic weight of the dopant, E_r the resonance energy,

$$E' = \frac{1}{2}m_n|\vec{u}_n(E)-\vec{u}|^2, \quad (2)$$

where m_n and $\vec{u}_n$ are the mass and velocity of the neutrons, and

$$\delta = 2\sqrt{\frac{E'k_BT}{A}}, \quad (3)$$

where k_B is Boltzmann's constant.

$$\vec{u}_n(E) = \hat{u}_n\sqrt{\frac{2E}{m_n}}, \quad (4)$$

where $\hat{u}_n$ is the direction vector of the neutron beam.

The spectrum $\alpha(E)$ calculated at any instant of time was interpreted as an apparent temperature by fitting a Gaussian, from which the apparent temperature was calculated using Eq. (3). The calculated attenuation spectrum was reproduced very well by a Gaussian, so it would not be possible to distinguish spatial variations by variations in the shape of the resonance.

Spatial variations in temperature accounted for 24 K of the apparent NRS temperature; spatial variations in velocity (including radial components) accounted for 124 K. The contributions did not combine linearly: the combined effect was 156 K. The resulting apparent temperature was consistent with the Mo NRS measurements. The magnitude of the pressure, temperature, and contributions to the apparent NRS temperature varied with time and position within the sample, because of reverberations in the projectile originating with the loading history applied at launch. The measured NRS temperature, when corrected for projectile curvature by subtracting 156 K, is consistent with the theoretical shock Hugoniots, and lies closer to the calculation using plastic flow (Fig. 1).

It can be seen that the effect of projectile flatness on the apparent temperature should be $o(100)$ K in these experiments by considering the variation in the component of material velocity along the neutron path inclined at 55° with respect to the axis for a material speed in the shocked state of ~ 1 km/s and the Forest Flyer projectile curvature of $\sim 5 \times 10^{-3}$/mm. The resulting shock curvature gives a speed variation of about ± 120 m/s along the axis of the neutrons, which equates to an effective temperature for ^{182}W atoms of 105 K when integrated around the azimuth. This simple estimate ignores the detailed contributions from neutron speeds and finer spatial variations which were included above, but is of the same order as the rigorous simulation.

We conclude that the NRS measurements of shock temperatures were consistent with the published EOS and constitutive behavior of Mo, taking into account the sensitivity of NRS to radial flow induced by the explosively driven projectiles used. Plastic flow was calculated consistently with the shock jump relations. Using published plasticity data for Mo, plastic flow was estimated to raise the material temperature by 53 K compared with the hydrodynamic shock Hugoniot, in the pressure regime of the experiments. This goes a long way to reconciling the measurements with temperatures expected from the various equations of state for Mo, but does not explain the whole discrepancy. The Forest Flyer system as described is likely to exhibit significant spatial and temporal variations in loading as applied to the sample. The overall effect was estimated to be 160 K, dominated by the contribution from spatial variations in velocity.

The temperatures inferred from NRS seem entirely plausible given the combined contribution of hydrodynamic shock, plastic work, and spatial variations in loading. The difference between the apparent temperatures was similar to the apparent heating caused by spatial variations in the projectile, and may reflect the variability of the explosively launched projectile design. The plastic work and nonideal projectile contributions to apparent temperature were all estimated using *a priori* modeling, with no adjustments made to improve the match to experiment.

Although the NRS technique for measuring shock temperatures has been known for some years, little further development has occurred because of the temperature discrepancy in Mo. Despite this lack of effort in development, the per-shot uncertainty in temperature in the Mo experiments was similar to that attained by mature pyrometry measurements. There is much scope for future developments of the loading system and the NRS diagnostic. Flatter shocks could be induced with improved explosive launchers, propellant guns, electromagnetic guns, or lasers. Electromagnetic and laser loading would allow the sample to be much closer to the neutron source, and the elimination of explosive products means smaller neutron losses from collision with H atoms. Other loading histories could be readily explored, including ramp and multiple-shock compression, and release from a

shock. More recent NRS temperature measurements have incorporated a filter at a known, fixed temperature with resonances bracketing those of the sample, e.g., Ag for ^{182}W, improving the measurement of the background during the shock experiment. NRS measurements could be made without doping with an element of different atomic number, possibly using the natural isotopic composition of a material, and correcting for the known resonance of unshocked material. Temperature uncertainties of $20-30$ K appear readily possible. Other neutron sources than spallation may be suitable, such as laser-accelerated protons followed by (p,n) reactions. It may be possible to construct a neutron detector giving spatial resolution. Eventually, the resonance signal could be precise enough to allow moments of the density of phonon states to be measured in a shocked material, as has been demonstrated statically.[12] Now that the previous NRS results are understood, the technique can be used with more confidence for the wide range of temperature measurements of interest in material dynamics.

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*Present address: Lawrence Livermore National Laboratory, CA 94551; dswift@llnl.gov

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