Chapter 2
Energy Dissipation and Shock-Induced Phase Changes in Mock Energetic Crystals via In-Situ Raman Imaging

Mahavir Singh*, Seongmin Yoon, and Vikas Tomar

Abstract This study investigates shock deformation and energy dissipation in mock-energetic crystals using advanced full-field time-gated Raman spectroscopy. The experimental approach involved multiple timestamps to assess shock dissipation along the [1 0 0] direction, aligning with the direction of shock propagation. A total of 1200 experiments were conducted to integrate and construct Raman spectra over four timestamps, each of a duration of short time windows of less than 10 ns, allowing for detailed observation of shock-induced changes. Flyer velocities of 1.2 km/s were utilized to generate high-pressure conditions of up to 5 GPa and temperatures exceeding 1000 °C within the sample. The motion of the aluminum flyer was monitored, and its planarity was confirmed using high-speed optical imaging, which demonstrated the necessity of maintaining a uniform impact to prevent non-uniform deformation during shock propagation. Further energy density dissipation was computed to be $6.67 \pm 2.11 \times 10^7$ kJ/m^3 and was found evenly distributed in the shocked region. The shock loading caused formation of a metastable phase which disappeared leading to micromechanical crushing and spall type failures in the crystals. For an impact velocity of 1.2 km/s the shock velocity was found to be 6.25 km/s. This comprehensive analysis provides insights into the elastic-plastic transitions and phase changes occurring in solid materials and a method to determine shock response in materials especially energetics which are opaque to PDV/VISAR wavelengths.

Keywords Shock deformation · Time-resolved Raman Spectroscopy · Energy dissipation · Energetic Crystals · Metastable Phase

Introduction

Understanding material response under shock compression is crucial for aerospace, defense, and materials science applications. Extreme pressures and temperatures induce complex deformation mechanisms, including elastic-plastic transitions, energy dissipation, and phase transformations [1, 2]. Characterizing these processes is essential for designing materials that endure high-strain-rate conditions. Traditional shock compression studies use photon Doppler velocimetry (PDV), X-ray diffraction, and laser interferometry to measure stress-wave propagation, but these methods lack real-time molecular-level resolution. Ultrafast spectroscopic techniques, particularly time-resolved Raman spectroscopy, have advanced the study of shock-induced structural changes at atomic and molecular scales [3].

Organic crystalline materials, including energetic compounds and their mock equivalents, undergo significant chemical and structural changes under shock loading [4]. Internal energy redistribution, metastable phase formation, and molecular rearrangements impact mechanical integrity and failure modes. Raman spectroscopy enables in-situ observation of these transformations, providing insights into energy localization and dissipation [5]. Sucrose, a widely used model system for shock studies, shares structural similarities with polymer-bonded explosives and has well-characterized Raman-active modes [3, 5]. Shock compression induces distinct phase transitions in sucrose, marked by shifts in CH and CH2 vibrational modes, which help quantify local stress and temperature distribution [5, 6]. This study employs multi-point time-gated Raman spectroscopy to capture the spatial and temporal evolution of shock waves in sucrose crystals, integrating ultrafast optical diagnostics for characterization of energy dissipation and phase transformations.

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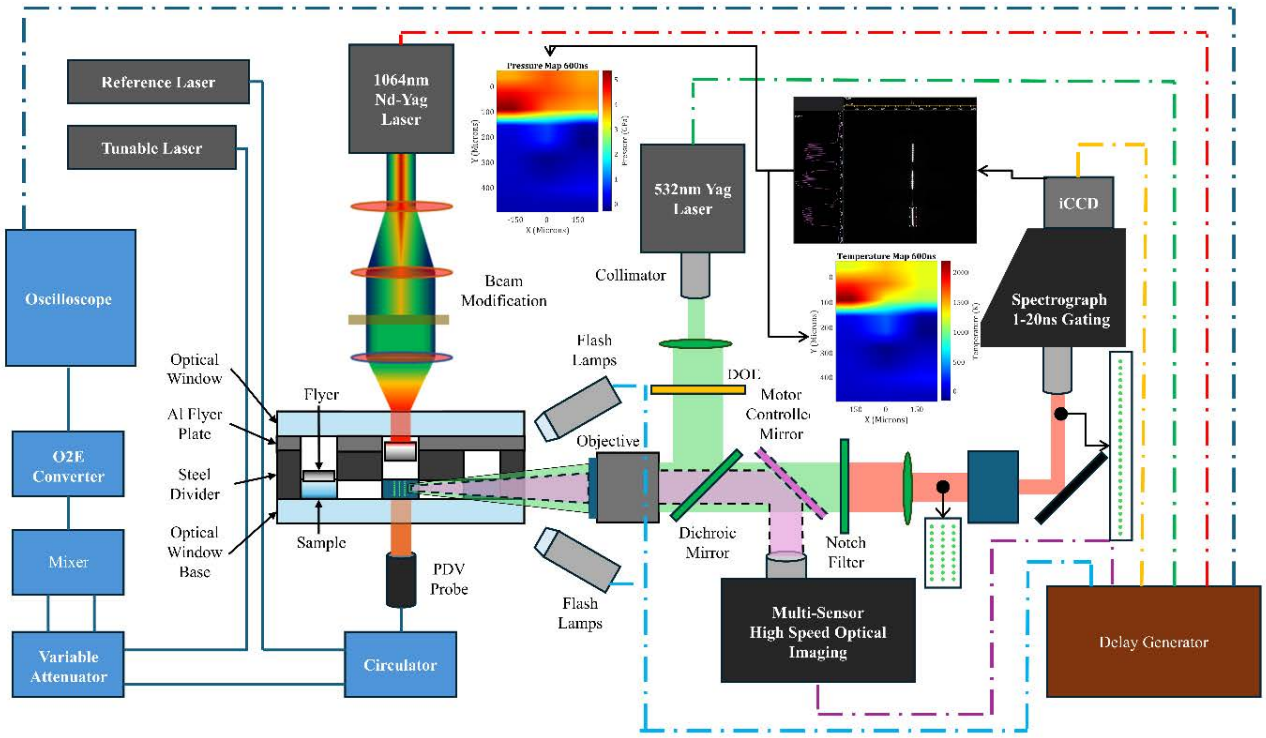

Fig. 1 Schematic of the experimental setup utilizing multi-point Raman spectroscopy to measure pressure and temperature distribution [6] in a shocked material, induced by a laser-driven flyer impact system. The peak shifts are subsequently used to calculate the energy dissipation density in the elastic shocked region.

Materials and Methods

Sucrose crystals were selected due to their well-characterized shock response and structural similarity to polymer-bonded explosives. The crystals were carefully polished and mounted to ensure uniform impact conditions. The experiments were conducted on samples with controlled thickness to achieve repeatable shock loading conditions. Shock waves were generated using a laser-driven flyer plate impact system, consisting of a 1064 nm pulsed laser with an 8 ns pulse width (figure 1). The specimens were mounted under a glass plate with an aluminum foil layer, from which aluminum flyers of 1.5 mm diameter were launched at velocities of 1.2 km/s. A precision alignment system ensured consistent impact positioning across multiple experiments.

Raman Spectroscopy and Optical Imaging: A custom-built multi-point, time-gated Raman spectroscopy system was employed to capture molecular-scale changes during shock loading. The system utilized a 532 nm pulsed laser with a 10 ns gate width. The laser beam was divided into 27 beams, which propagated in parallel and struck the front surface of the sample through a 532 nm dichroic mirror (figure 1). High-power switchable microscopic objectives enabled the acquisition of high-resolution Raman spectra at different spatial locations. After data collection, the 27 beams were optically modified to enter the spectrograph and an iCCD camera within a 10 ns gating window. The experiments were repeated 300 times to construct a complete Raman spectrum in the 2950–3050 cm^{-1} range, ensuring high-resolution data around the CH and CH2 intra-molecular vibration bands.

Results and Discussion

Raman spectra were recorded at four distinct timestamps corresponding to the undeformed state, elastic wave arrival, elastic-plastic deformation, and post-shockwave stages. The elastic peak shifts were analyzed to determine pressure and temperature variations, with the CH and CH2 peaks at 2990 and 3018 cm^{-1} used to characterize the shocked state of the material. These peaks were calibrated to quantify changes in pressure and temperature during shock compression.

Pressure and Temperature Distributions: Using Equation 1, the pressure and temperature distribution as shown in figure 1, within the shock wave were mapped by shifts in the CH2 symmetric and asymmetric stretching modes. The maximum recorded pressure was 5 GPa, with a corresponding temperature exceeding 1000 °C. The spatial distribution of temperature closely followed the pressure gradients, suggesting strong thermomechanical coupling during shock propagation. The constants C1-C4 are correlation constants for pressure and temperature against the peak shifts of the CH and CH2 peaks.

$$dP = \frac{C_4 \cdot d\omega_1 - C_3 \cdot d\omega_2}{C_1 \cdot C_4 - C_2 \cdot C_3} \quad dT = \frac{C_2 \cdot d\omega_1 - C_1 \cdot d\omega_2}{C_2 \cdot C_3 - C_1 \cdot C_4} \tag{1}$$

Shock-Induced Energy Dissipation and Phase Transitions: Utilizing the linear dependence of pressure and temperature given in Equation 1, and assuming the shock deformation is fast enough for the heat loss to be purely adiabatic in nature, the total energy dissipation density was derived using Equation 2.

$$E = \iint \frac{\partial^2 E}{\partial\omega_1 \partial\omega_2} d\omega_1 d\omega_2 = \frac{\rho_0}{C_3}\left(match\omega_1 - \omega_{1(0)}\right) \left[\frac{-bC_1 + \frac{aC_3}{K}}{C_2.C_3 - C_1.C_4}\left(\omega_2 - \omega_{2(0)}\right) + \frac{bC_1C_3}{K\left(C_1.C_4 - C_2.C_3\right)^2}\left(\omega_2^2 + 3\omega_{2(0)}^2 - 4\omega_2\omega_{2(0)}\right)\right] \tag{2}$$

The energy density was found to be 6.67 $\pm$ 2.11 $\times$ 107 kJ/m^3 within the elastic shocked region, with a uniform distribution. Additionally, Raman spectroscopy data revealed a sequential transformation of the material under shock loading. Initially, an elastic precursor wave propagated through the sucrose crystal, as indicated by small, reversible shifts in Raman peaks. As the shock wave intensified, a transition to an elastoplastic state was observed, characterized by peak broadening and the emergence of new vibrational modes. Subsequently, a metastable phase was observed, which disappeared after the shock wave passed. Under these deformations, the crystal underwent micro-mechanical crushing failure, converting into a pool of very fine powder. The fast and localized deformation caused the specimen to retain its original shape.

Conclusions

This work successfully demonstrates the capability of time-gated Raman spectroscopy in visualizing and quantifying shock-induced transformations in sucrose crystals. The experimental setup enabled high-fidelity measurements of pressure, temperature, and energy dissipation with unprecedented temporal and spatial resolution. The results contribute to a deeper understanding of elastic-plastic transitions and phase changes in mock energetic materials, providing a foundation for shock wave dynamics in energetic materials.

References

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