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Estimating the pressure of laser-induced plasma shockwave by stimulated Raman shift of lattice translational modes

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The current paper investigates stimulated Raman scattering (SRS) when laser-induced plasma is formed in heavy water by focusing an intense pulsed 532 nm Nd:YAG laser beam at room temperature. An unexpected low-frequency SRS line attributed to the lattice translational modes of ice-VII (D_2O) is observed. The pressure of the plasma shockwave is estimated using low-frequency SRS line shift. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4736410]

Laser-induced breakdown (LIB) refers to plasma production by focusing an intense pulsed laser beam on substances. LIB is a complex phenomenon with various optical, electronic, thermal, and mechanical effects. Studies of LIB in liquids have focused on initial nonlinear optical processes,[1–3] shockwave generation,[4] cavitation phenomena,[5] and so on. Laser shockwave compression can cause structural change in a liquid substance and new substance formation.[6–8] The pressure of the shockwave is important to understand new substance formation. The peak-pressure of the shockwave during plasma formation on picosecond or nanosecond time scale has been measured using hydrophone and time-resolved photography.[9,10] However, we provide a simple method using stimulated Raman shift with lattice translational modes to calculate the pressure of laser-induced plasma shockwave in heavy water.

The stimulated Raman experimental set-up consisted of a second-harmonic generation Nd-YAG laser source (532 nm) that operated in switching mode (1 Hz repetition frequency, 12 ns pulse length, energy up to 200 mJ), a spectroscopic system (Ocean optics), and a 10-cm-long quartz cell (Fig. 1(a)). The focal point of the laser was at the middle of the cell. The focal length of the focusing lens was 100 mm. The purity of the sample (prepared by Sigma-Aldrich Co.) was 99.9%. The high-pressure cell used in this experiment was based on the diamond anvil cell (DAC) having two diamonds with 500 μm culet size (Fig. 1(b)). The sample with a small ruby chip ($\sim$10 μm) was loaded in a 200 μm hole drilled through a 250-μm-thick T301 gasket preindented to 80 μm thickness. Then, the cell was carefully pressurized with small steps and allowed to stabilize for a few minutes after each pressure change before Raman spectra were taken. Pressure was calibrated using ruby fluorescence. No pressure-transmitting liquid medium was used in the experiment. The high-pressure spectra of heavy water were obtained at 514.5 nm excitation from an Ar ion laser attenuated to 3.6 mW on a Renishaw InVia Raman spectrometer (Spectra Physics163-M42). The excitation beam was positioned on a 5× objective lens into DAC. All experiments were performed at room temperature. For a Gaussian laser beam with energy up to 140 mJ at 532 nm, considering a laser filament diameter of 100 to 200 μm and pulse duration of 12 ns, the maximum irradiance could be estimated at approximately 10^{12} W cm^{-2}. This estimation did not consider the energy loss caused by the heavy water-laser beam interaction and the Kerr self-focusing effect. The irradiance at 130 mJ laser energy was beyond the breakdown threshold of heavy water.[11]

Strong coherent radiations around 610 nm and 470 nm are produced when a 532 nm laser pulse at 100 mJ to 130 mJ energy is focused into bulk heavy water. These coherent radiations are only the results of the stimulated Raman scattering (SRS) of Stokes line and anti-Stokes line for *D–O* stretching vibration ν_1 in heavy water. Similar conclusions have been drawn by other researchers.[12] However, when laser pulse energy is above 130 mJ, an unexpected result can be obtained. A stimulated Raman line 288 cm^{-1} in forward direction begins to form (Fig. 2).

The point group of liquid heavy water molecule is C_{2v}, which has three fundamental vibration modes, namely, $v_1(A_{1g})$, $v_2(A_{1g})$, and $v_3(E_g)$, at approximately 2500, 1178, and 2623 cm^{-1}, respectively.[13] Only the symmetrical vibration mode (v_1,v_2) is strongly active in Raman scattering. The Raman spectra of heavy water molecule do not exhibit a low-frequency vibration mode. The 288 cm^{-1} stimulated Raman line seems unaccountable. However, ice phases exhibit abundant low-frequency lattice modes, such as ice-VI, VII, and VIII.[14]

The 288 cm^{-1} low frequency is attributed to lattice translational modes of ice-VII. Ice-VII, found by Bridgman,[15] is cubic [space group $Pn\bar{3}m(O_h)$] with two molecules per unit cell on site symmetry $\bar{4}3m(T_d)$ (O site $\bar{4}3m$, H site $3m$) and has a proton-disordered paraelectric structure.[14] The density of ice VII is 1.52×10^3 kg/m^3. When the irradiance of the laser reaches 10^{12} W·cm^{-2}, heavy water will breakdown and ionize, generating laser-induced plasma and

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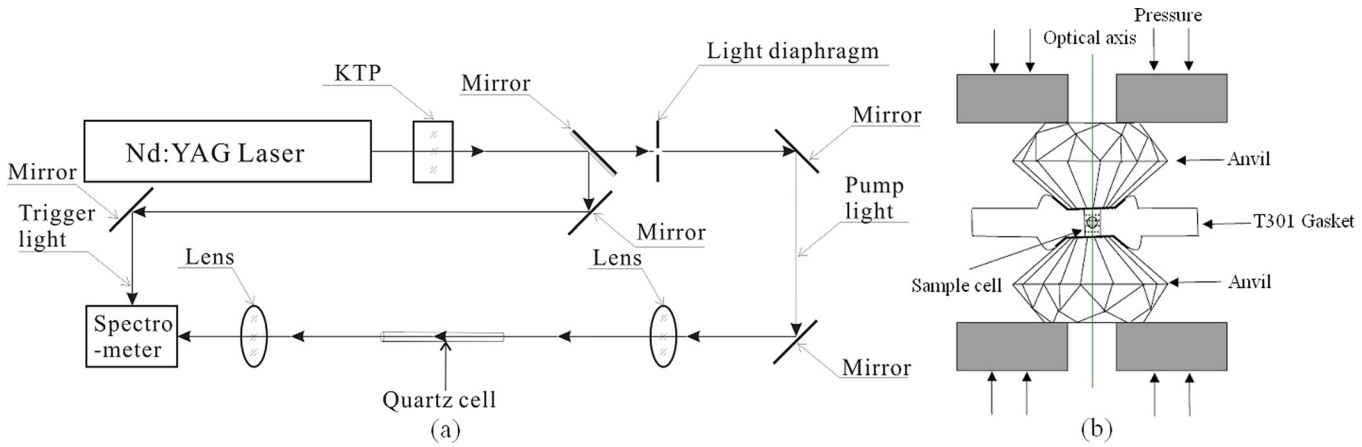

FIG. 1. (a) Schematic diagram of the experimental setup to measure forward spectra of heavy water SRS. (b) Sketch of the diamond anvil cell.

resulting in shockwave formation.[16,17] Laser-induced breakdown mechanism in water and heavy water, as well as in other liquids, has been well studied.[7,18–20] The breakdown starts after absorption of the focused laser light by persistent molecules. The absorbed energy forms high-pressure vapors, which support avalanche ionization in that area. The vaporization process is explosive and generates a powerful shockwave in the surrounding liquid. Ice VII structure is formed by the shockwave, as the trajectory of the shockwave passes through the stable pressure-temperature range of ice VII.[21] The 288 cm^{-1} low-frequency Raman line is attributed to the lattice vibration of heavy water. The Raman shift of lattice translation mode line also linearly increases with an increase in the pumping energy (Figs. 3 and 4).

The relation of material pressure P and μ_p is as follows: $P - P_0 = \rho_0 \mu_S \mu_P$, where μ_p is the particle velocity, and μ_s is the shockwave velocity. P_0 and ρ_0 are the unshocked material pressure and density. P is the material pressure of the laser-induced shockwave.[22] The Raman spectra of heavy water in the DAC up to 14 GPa have been determined to check the shockwave pressure (Fig. 5). Compared with the Raman shift at different pumping energies, the shockwave pressure changes from 3 GPa to 7 GPa under the experimental conditions. Rybakov *et al.* also observed ice-VII only

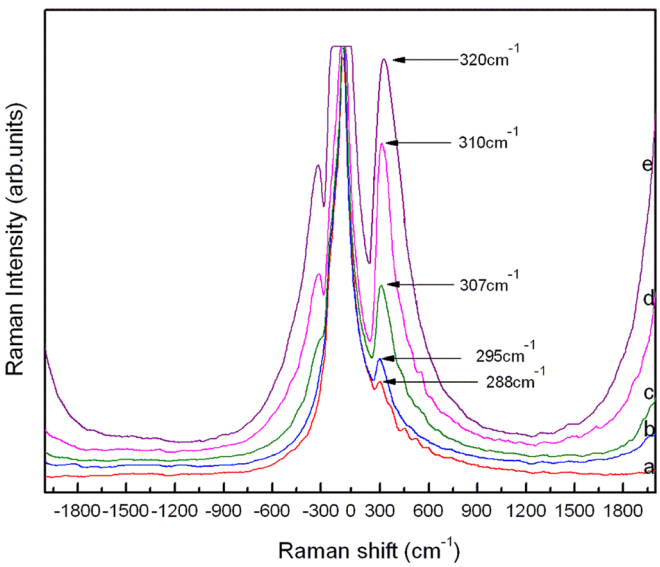

FIG. 3. Spectrum of stimulated Raman at different pumping energy. (a) 130 mJ (b) 135 mJ, (c) 140 mJ, (d) 145 mJ, and (e) 150 mJ.

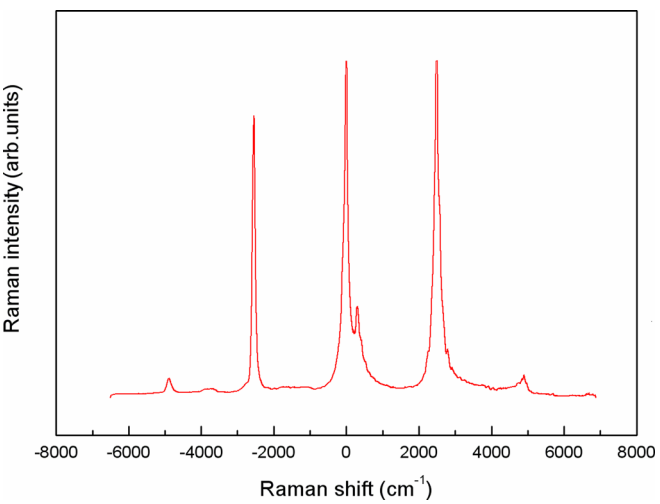

FIG. 2. SRS spectrum of heavy water at 140 mJ energy.

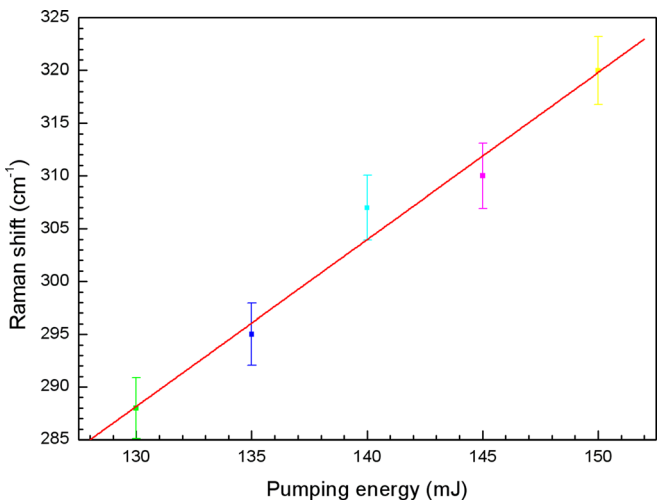

FIG. 4. Raman shift at different pumping energy.

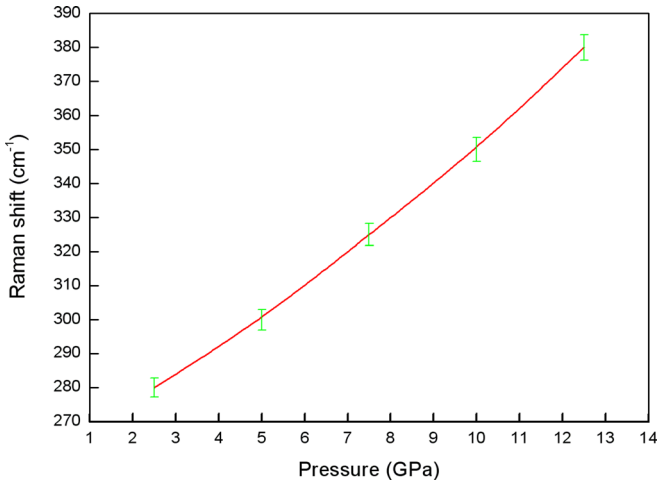

FIG. 5. Lattice mode shift as a function of pressure.

under particle velocities between 0.75 and 2 km/s.[23] Under the particle velocity 0.75 and 2 km/s, the material pressure by laser-induced shockwave is between 3 GPa to 11 GPa. The pressure result of Rybakov *et al*. is in agreement with the experimental results 3 GPa to 7 GPa. Vedadi *et al*. also performed molecular dynamic simulations of water subjected to shockwave.[24] They confirmed that the water phase transforms into an ice-VII-like structure at a particle velocity of 1 km/s (shockwave pressure is 4 GPa). Their theoretical simulation strongly supports our conclusion in the present study.

From the normal Raman spectrum, we can find that the intensity of lattice vibration is too weak to accomplish the SRS of lattice vibration.[25,26] Generally, generating SRS in practice when the Raman cross-section is very small is difficult. The observed lattice translational mode in the SRS may have resulted from the following factors. First, the SRS is a third-order nonlinear process. Hence, its inducement depends on the third-order nonlinear susceptibility, which is mainly affected by electronic nonlinear polarizability.[19] Under plasma conditions, which indicate the generation of a high electric field, the electronic nonlinear polarizability is drastically enhanced. Second, the shockwave would preferentially excite phonon and lower energy molecular vibration, producing highly nonequilibrium vibrational populations.[27–29] Thus, the SRS assigned to the lattice translational modes of ice-VII is easily discovered. The shockwave pressure changes as the laser power changes.

In conclusion, the low-frequency SRS line attributed to the lattice translational modes is observed when laser-induced plasma shockwave is generated. The lattice translational mode SRS indicates that an ice-VII structure is formed in the focal volume of pumping beam in heavy water. Compared with static compression, the shockwave pressure is estimated using SRS of the lattice translational modes.

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