## **SUPPORTING INFORMATION FOR:**

## **Enhanced Stimulated Raman Scattering by Pressure- Controlled Shock Wave in Liquid water**

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## **Experimental**

The liquid water has been deionized from triple distilled water. High pressures are derived from equivalent pressure (Fig. S1). The sample was kept in a quartz cell whose width, height, and length were 1, 5, and 10 cm, respectively. The second harmonic of the pulsed Nd-YAG laser with a wavelength of 532 nm was used as excitation laser and focused in the quartz cell. The pulse duration was 12 ns (full width at half maximum [FWHM]), and the repetition rate was 10 Hz. The optical power density and diameter are  $1.4 \times 10^{12}$  Wcm<sup>-2</sup> and 4 mm, respectively. Spectra were obtained by using an objective lens with times 50 long working distance. The output light was accepted on the spectrometer (The spectral resolution was 2 cm<sup>-1</sup>) and the signals were managed by a computer. The experimental setup is shown as Fig. S2.

The scanning speed of spontaneous Raman spectra was 10 cm<sup>-1</sup>/s and the spectral

resolution was 1 cm<sup>-1</sup>. The spectra were measured by Renishaw InVia Raman micro spectrometer. Spectra have been gained by using a 50× long working distance objective lens located in the different samples and detected with a CCD detector. The sample, which has been excited by an Argon laser at 514.5 nm and an output power of 10 mW, was kept in a quartz disk of 10 mm radius.

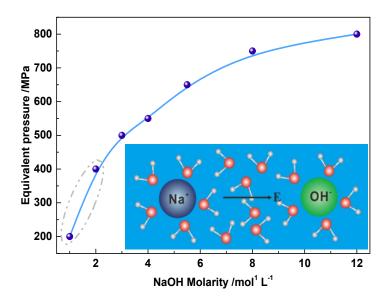


Fig. S1. Equivalent pressure for different NaOH solutions vs Molarity calculated according to Ref [1]. Insert: Schematic diagram of NaOH dissolved in liquid water.

Dissociation of NaOH in water yields the monovalent Na<sup>+</sup> and OH<sup>-</sup> ions. Each Na<sup>+</sup> and OH<sup>-</sup> ion acts as a charge center, generating a radial ion field (E), as shown in insert of Fig. S1 [2]. The ion field softens (lengthens) the O-H bond and stiffens (shortens) the O:H interaction, which has the same effect as high pressure to pure water. Therefore, different concentrations of NaOH produce different equivalent pressures (Fig. S1.). To get better SRS experimental results, the NaOH solutions must be transparent. So, we selected 1 and 2 M NaOH solutions (the dotted line of Fig. S1) and the corresponding equivalent pressures of these concentrations are 200 and 400 MPa.

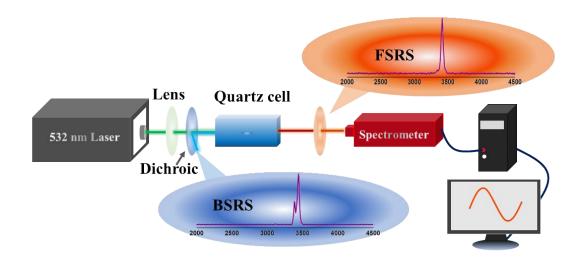


Fig. S2. Schematic diagram of the experimental setup to measure SRS spectra. Insert: SRS spectra of liquid water.

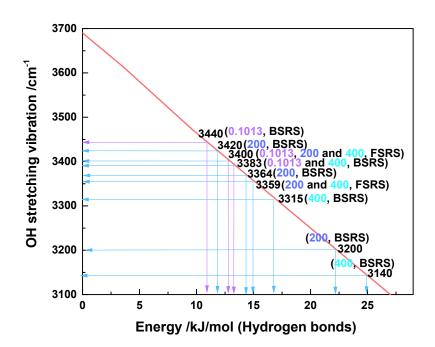


Fig. S3. The relationship between OH stretching vibration and hydrogen bonds energy. Light purple: Atmospheric pressure; Light blue: High pressure.

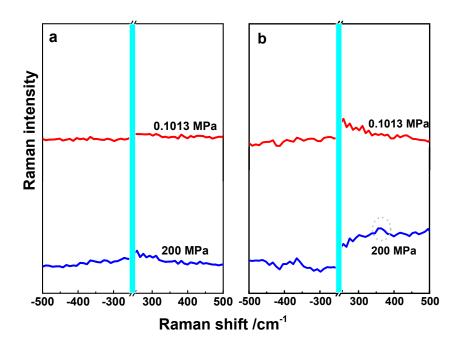


Fig. S4. Low-wavenumber SRS spectra of liquid water in different directions; (a) Forward; (b) Backward.

When LIB process occurs, a small amount of hydrogen is produced in liquid water. However, the SRS peak of hydrogen is difficult to observe at normal conditions. In addition, the Raman peak of hydrogen is mainly concentrated at about 380 cm<sup>-1</sup> [3]. Therefore, we obtain low-wavenumber SRS spectra of liquid water in both directions at atmospheric pressure and 200 MPa as seen in Fig. S4. We find that the peak of hydrogen cannot be observed at atmospheric pressure and 200 MPa. Although this peak may appear in the backward direction at 200 MPa (grey dotted line), this peak is extremely weak. So, we are not sure whether this peak exists or not.

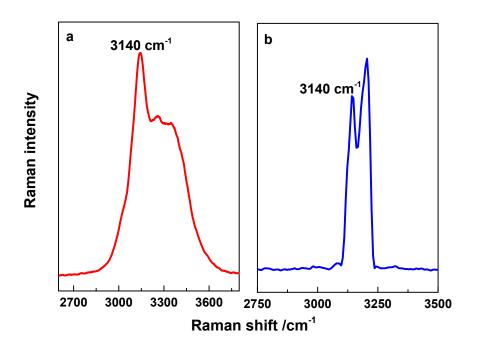


Fig. S5. (a) Spontaneous Raman spectrum of ice Ih at 253 K; (b) FSRS of ice Ih.

Raman shift /cm-1	Assignments
3087	DAA
3231	DDAA
3401	DA
3533	DDA
3642	Free OH

Table S1. Spontaneous Raman peak of liquid water corresponding to mode of vibration (D: donor of hydrogen bonds; A: acceptor of hydrogen bonds); Red ball: O atom; Gray ball: H atom [4, 5].

The temperature of liquid water after LIB is obtained by Equation S1 [6-8] at 400 MPa.

$$T = \frac{hc_0v_k}{K \times \ln\left[\frac{I_s}{I_{as}}(\frac{v_p - v_k}{v_p - v_k})^4\right]}$$
(S1)

Where is  $h = 6.62 \times 10^{-34} \text{ J} \cdot \text{S}$ ,  $c_0 = 3 \times 10^8 \text{ m/s}$ ,  $K = 1.38 \times 10^{-23} \text{ J/k}$ ,  $v_p = 19033 \text{ cm}^{-1}$ . In addition,  $v_k$  represents the Raman shift of the OH vibration peak of water molecules. T is temperature;  $I_s$  and  $I_{as}$  represent the relative strength of Stokes and anti-Stokes peaks, respectively.

## Reference

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