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High-pressure elasticity of Baltic amber studied by Brillouin spectroscopy

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1. Introduction

Baltic amber (or succinate) is the most renowned form of amber, an amorphous fossil resin vitrificated for a few hundred million years. Due to its lengthy and rare formation process, Baltic amber calls for much attention not only for geologists and paleontologists but also for condensed matter physicists ascribed to its peculiar elastic properties.[1,2)]

The pressure is one of the most important parameters to modify various properties of materials. Especially, understanding the densification behavior of materials in extremely high pressure is essential.[3)] Since Baltic amber is a unique glass produced by vitrification with hyperaging, investing its elasticity modulation induced by pressure would give an important clue to understand macromolecular materials.

In our experiment, we employed high-pressure Brillouin light scattering to investigate the elastic properties of the Baltic amber under high pressure.

2. Experimental methods

Brillouin light scattering experiments were carried out with a multi–pass tandem Fabry–Perot interferometer (TFP–1, JRS scientific instruments) and a 532 nm diode-pumped solid-state laser (Excelsior–532–300, Spectra-Physics). The laser intensity was attenuated to ~6 mW to protect the sample. A photon-counting device and an analyzer attached to the Fabry–Perot interferometer were used. A schematic diagram of the optical setup with a diamond anvil cell (DAC) is described in **Fig. 1**. The DAC was used to apply extremely high pressure to the sample.

The Baltic amber sample was cut into a small piece. It was loaded in a gasket hole of symmetric DAC and filled the entire gasket hole to apply hydrostatic pressure without pressure transmitting medium. Two ruby spheres are also included in the gasket hole with the sample. The ruby fluorescence spectra to measure the pressure inside the gasket hole were collected by a Spectra Pro®–275 (Acton Research Corporation) spectrometer equipped with a PMT detector.

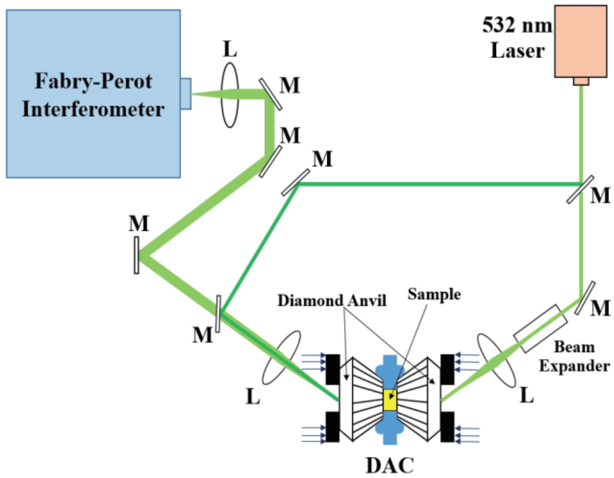

Fig. 1 A schematic diagram of the high-pressure Brillouin light scattering experiment setup. The letter M indicates a mirror and L indicates a lens.

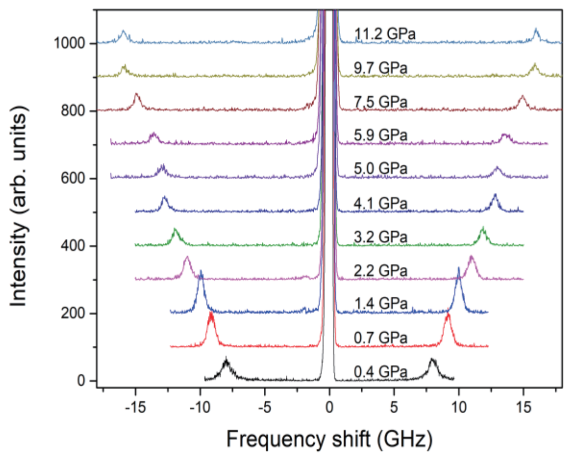

Fig. 2 The Brillouin spectra obtained at various pressure conditions in a forward, symmetric geometry.

3. Results and discussion

Fig. 2 shows the Brillouin spectra of the Baltic amber in a forward, symmetric scattering geometriy under increasing pressure. TA phonon modes were not observed in the entire pressure range. Meanwhile, only an LA phonon mode was observed as well in a backscattering geometry which is consistent with the Brillouin selection rule for isotropic materials.[4)]

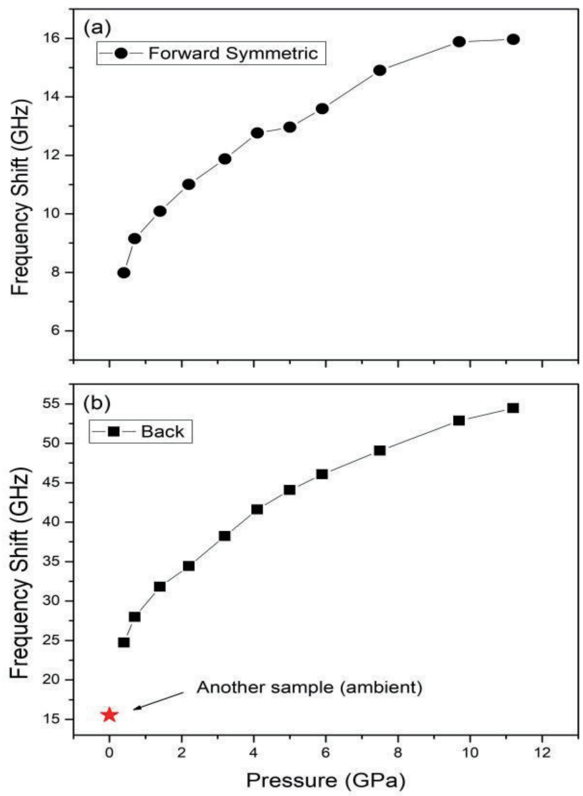

Fig. 3 The Brillouin frequency shift plotted against pressure for (a) a forward symmetric geometry and (b) a backscattering geometry. The red star indicates the value obtained without a DAC.

Fig. 3 shows the frequency shift of LA mode in the Baltic amber in two scattering geometries. The Brillouin frequency shift of the LA mode increased monotonically when pressure was applied, which is a direct signature of densification.

The Brillouin peak frequency in two distinct geometries gives information about the acoustic velocity and refractive index.[5)] The acoustic velocity is plotted in **Fig. 4**. The acoustic velocity rapidly increased upon compression at low pressures below ~2 GPa which indicates the collapse of the free volume and then increased slowly up to ~11 GPa. It is well known that the acoustic velocity is approximately proportional to the boson peak (BP) frequency. According to the theory on BP considering a vibrational instability**,** the pressure dependence of BP is proportional to $(1+P/P_0)^{1/3}$ where P_0 is a constant.[6)] The data points in Fig. 4 were fitted well with the formula $v(0)(1+P/P_0)^n$ with n=0.25, which shows that a similar relation holds. This coincides with the theory of BP as well.

Besides, the refractive index of the Baltic amber was also calculated by the law of refraction of light. Employing Clausius–Mossotti relation with an assumption of mean polarizability in the Baltic amber, the density-pressure isotherm was also calculated. Both of the refractive index and density

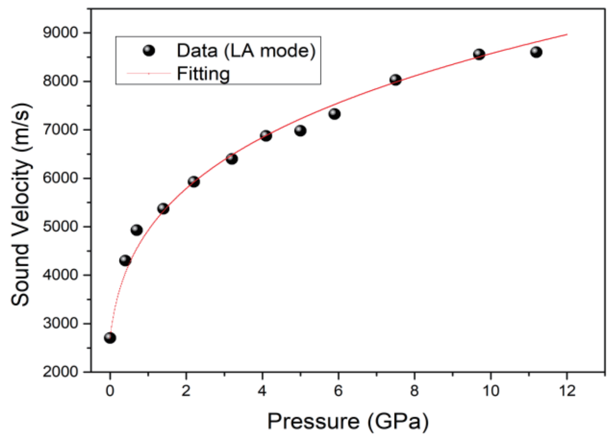

Fig. 4 The acoustic sound velocity of the LA phonon mode in the Baltic amber plotted against pressure. The red line is a fitting based on the theory of boson peaks.

showed a slightly increasing tendency.

4. Conclusion

In conclusion, we successfully observed the pressure-dependent behavior of the acoustic sound velocity of the Baltic amber. When pressure is applied, the acoustic velocity rapidly increased in the low-pressure regime and became nearly saturated at ~ 11 GPa. The refractive index and density of the sample gradually increased as a function of pressure as well.

Acknowledgment

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