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Pressure Dependence of Poisson's Ratio of Glassy Baltic Amber Studied by Brillouin Scattering Spectroscopy

ブリルアン散乱法によるガラス状バルト琥珀のポワソン比の圧力依存性

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1. Introduction

The magnitude of the elastic moduli is governed by both the atomic bond energy and the atomic packing density, while the Poisson's ratio is related to the degree of symmetry of the structural units existing at the molecular scale. The fragility of a glass-forming liquid is intimately linked to a very basic property of the corresponding glassy states: Poisson's ratio, or the ratio of longitudinal and transverse sound velocity, V_l and V_t, respectively.[1)] Fragility index m of liquids is defined as the apparent activation energy of shear viscosity η or structural relaxation time τ a at the glass transition temperature T_g. Analysis of a large number of glasses, including covalent and hydrogen-bonded, van-der-Waals and ionic glasses, shows a correlation between V_l/V_t and m.

Regarding presure dependence of fragility, it was reported that three kinds of intermediate small-molecule glass-formers do not show a pressure dependence of fragility within experimental error.[2)] The defect diffusion model explains these experimental findings, supporting a clustering mechanism in the glass-forming process.

The pressure dependence of Poisson's ratio of a typical strong glass, SiO_2 shows the remarkable increase up to 13 GPa.[3)] However, in theintermediate glycerol, at low pressure it increases gradually, while at pressure higher than 1 GPa it becomes constant.[4)] Although pressure dependences of the Poisson's ratio have been studied for a few glass-forming systems, not much is known about the influence of pressure on Poisson's ratio.

In many polymer glasses, the fragility increases as molecular weight increases. Amber is a unique example of a glass that has been aging for a very long time below its glass transition temperature, thus reaching a state which is not accessible under normal experimental conditions. It is very fragile and its fragility index was reported to be about 90.[5)] As fragility increases, the intensity of fast relaxation process increases while the intensity of boson peak

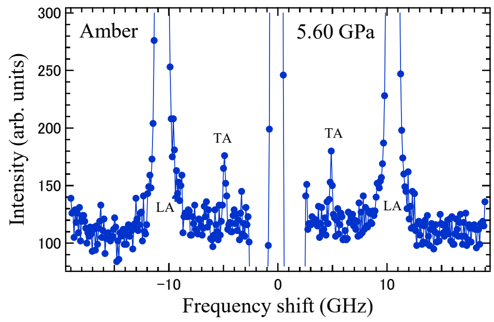

Fig. 1 Observation of LA and TA peaks in Brillouin scattering spectra of Baltic amber at 5.60 GPa.

decreases. The boson peak of Baltic amber is not yet observed by Raman scattering and THz time domain spectroscopy reflecting the very high fragility index.[6)] Recenly amber has attracted much attention as an ideal fragile glass.[7)] In the present study the pressure dependence of poisson's ratio of Baltic amber is investigated by maicro-Brillouin scattering.

2. Experimental

Brillouin scattering spectra of Baltic amber were measured in a symmetric scattering geometry, with the scattering angle of 50-degree, using a high-contrast 3 + 3 passes tandem Fabry–Perot interferometer with a free spectral range of 25 GHz for longitudinal acoustic (LA) and transverse acoustic (TA) modes. The exciting source was a diode-pumped solid state laser with a wavelength of 532 nm. The pressure dependence was measured using a symmetric diamond anvil cell.[8)] A piece of amber glass sample was grounded first, then loaded into a 150 μm stainless steel gasket hole. The pressure of a sample was determined by the standard Ruby fluorescence method. No pressure medium was added. Both compression and decompression curves were measured between ambient and 12 GPa. Brillouin scattering experiments were performed at Sector-13 of APS, Argonne National Laboratory.

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3. Results and Discussion

The pressure dependence of Brillouin scattering spectra of Baltic amber is shown in **Fig. 1**. The intensity of a TA mode is much weaker than that of a LA mode. Up to the present, there is no report on the observation of the TA mode of Baltic amber within our knowledge. The pressure dependence of the LA mode velocity V_l is in a good agreement with the previous study.[9)] The LA mode velocity V_l increases as the pressure P increases, and it obeys the following empirical equation.

$$V_l = V_0(1+P/P_0)^n \tag{1}$$

Here, V_0=2.7 km/s,[10)] P_0=0.1 GPa, n=1/4. Eq. (1) is similar to the pressure dependence of the boson peak frequency ν_{BP} predicted in the soft potential model[11)]

$$\nu_{BP} = \nu_0(1+P/P_0)^{1/3} \tag{2}$$

The difference of index n=1/4 from 1/3 of Eq. (2) may be caused by the pressure dependence of medium range order, because V_l/ν_{BP} is proportional to the medium range order length.

The Poisson's ratio σ is calculated by the following equation

$$\sigma = \frac{V_l^2 - 2V_t^2}{2(V_l^2 - V_t^2)}. \tag{3}$$

Here V_l and V_t are the phase velocity of longitudinal acoustic (LA) and transverse acoustic (TA) modes, respectively. The pressure dependence of Poisson's ratio of Baltic amber up to 12.0 GPa is shown in **Fig. 2**. It is found that the Poisson's ratio σ of Baltic amber is nearly constant against pressure within experimental uncertainty. Considering the correlation between fragility and Poisson's ratio, the fragility of amber may also keep constant against pressure.

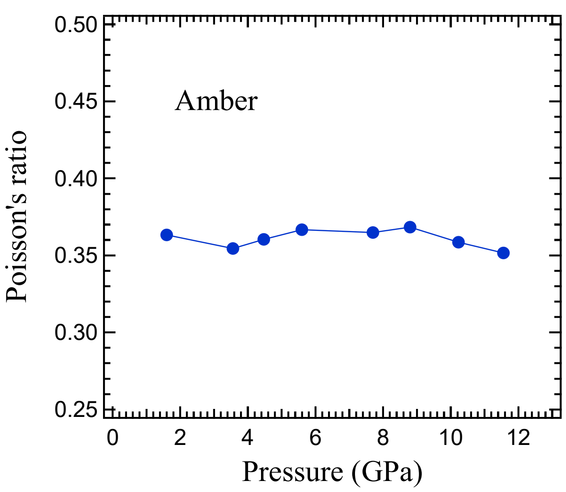

Fig. 2 Pressure dependence of Poisson's ratio of Baltic amber.

4. Conclusions

Baltic amber is natural polymer glass with high fragility index. The pressure dependence of sound velocities and Poisson' ratio of glassy Baltic amber was measured up to 12.0 GPa using a Brillouin scattering spectrometer and a diamond anvil cell. It is found that the Poisson's ratio of Baltic amber is nearly constant against pressure.

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