Transverse Acoustic Excitations in Simple Liquid

Lattice modes are modes of vibration of a macroscopic part of a system. In the long-wavelength limit, they may be described by the classical theory of elasticity in which the system is treated as a continuum. As the wavelength of the vibrations becomes comparable to the distance between the atoms, the microscopic structure of the system and the forces between individual pairs of atoms become the dominant features in determining the nature of the modes of vibration.

In simple liquids, longitudinal acoustic (LA) phonon modes are easily observed by ultrasonic, optical, or inelastic scattering experiments because density fluctuations occur owing to a strong repulsive force between atoms when they approach each other. However, transverse acoustic (TA) phonon modes cannot usually be detected in liquids by ultrasonic or optical measurements since the shear force over long distances is very weak. Over short distances, however, there may be a cage effect that acts as a restoring force for TA modes. The TA modes can be observed even in liquid metals in the dynamic structure factor $S(Q,\omega)$ measured by inelastic X-ray or neutron scattering (IXS or INS). Although the TA modes in simple liquids have been discussed theoretically for more than thirty years [1], experimental evidence has not yet been obtained. To detect the TA modes, we carefully measured IXS from liquid Ga at 40°C at beamline **BL35XU** [2]. A Ga sample, 50 μm in thickness, was contained in a thin-walled (0.25 mm) single-crystal sapphire cell [3].

Figure 1 shows the $S(Q,\omega)/S(Q)$ result measured at Q = 10.6 nm^{-1}. LA excitations are clearly seen as large peaks at about 17 meV. In addition, small extra excitations can be detected at about 8 meV between the central quasielastic peak and the LA peaks. The solid curves in the figure indicate the best fits and residuals for the single and double damped harmonic oscillator (1DHO and 2DHO, respectively) models; the 1DHO model includes only the LA excitation, and the 2DHO model, both the LA and TA modes. We focus on the second, weak mode as a signature of a TA excitation. As clearly observed in the figure, the 2DHO model reproduces the experimental $S(Q,\omega)$ spectrum well.

Figure 2 shows the dispersion relation of the weak (open circles) and strong (full triangles and full circles) excitations obtained from the 2DHO fits. The solid curve represents the TA phonon excitations obtained by orbital-free *ab initio* molecular dynamics (MD) simulation. This curve almost coincides with the present experimental data. However, the TA phonon

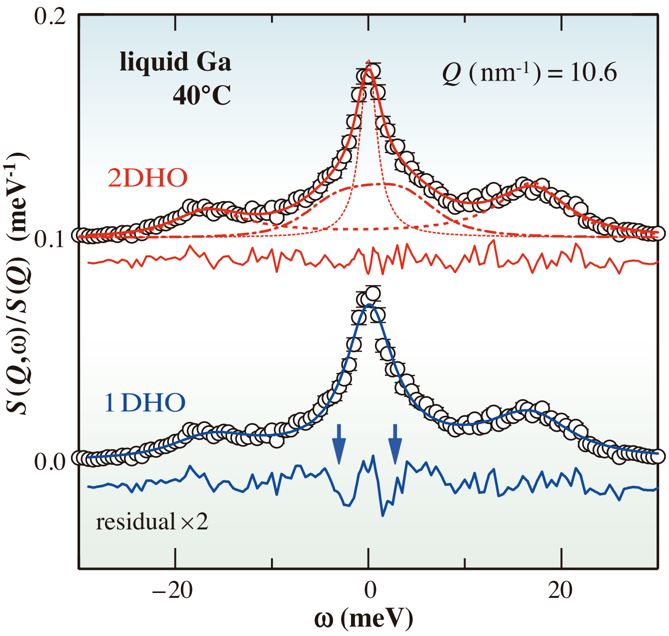

Fig. 1. $S(Q,\omega)/S(Q)$ spectrum at Q = 10.6 nm^{-1}. Solid curves represent the best fits and residuals obtained using the 1DHO (lower panel) and 2DHO (upper panel) models, and dotted, dashed, and chain curves denote the quasielastic, LA, and TA contributions, respectively. See the text for details.

modes cannot be detected in crystals in the first Brillouin zone by IXS experiment, since the direction of TA oscillations is perpendicular to the *Q* direction. On the other hand, the MD simulation reveals that a mixing of the TA and LA excitations occurs in liquid Ga, and the peaks of the quasi-TA branch in the LA excitations detectable by IXS are given by the dashed curve in Fig. 2. This curve almost coincides with the present experimental data, which is clear evidence of the existence of the TA mode in liquid Ga. The velocity of the TA sound is slightly larger than 1050 m/s, indicating a shear modulus of about 6.5 GPa, about 20% of the solid Ga value.

In accordance with the Heisenberg uncertainty principle, the lifetime τ of the phonons can be estimated to be $\tau = \pi/\Gamma_Q$, where Γ_Q is the width of the DHO close to half width at half maximum. Figure 3(a) shows the τ values of the TA (open circles) and LA (other marks) phonons. The τ value of the TA phonons is about 0.5 ps, which is almost the same as that of the LA phonons. Figure 3(b) shows the propagation length *L* of the TA and LA phonons (lifetime times velocity of sound). The *L* value of the TA phonons is 0.4-0.5 nm, which is much smaller than the Ioffe-Regel limit (dashed curve), indicating that the TA phonons are localized. This value may correspond to the size of the cages formed instantaneously (0.5 ps) in liquid Ga.

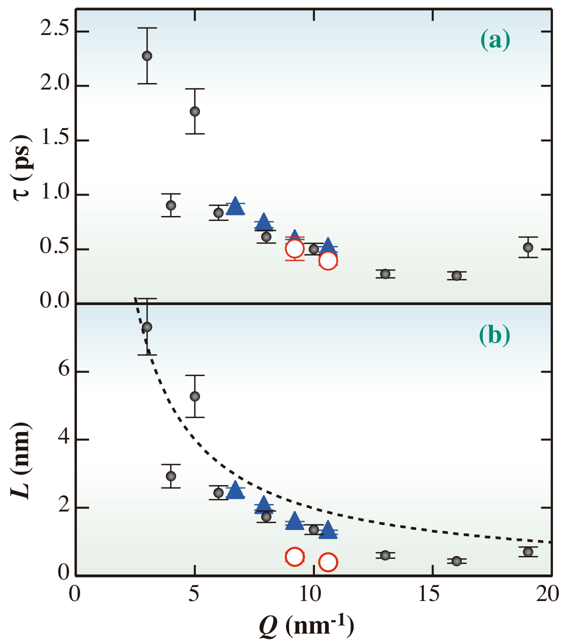

Fig. 3. **(a)** Lifetime τ and **(b)** propagation length *L* of TA (open circles) and LA (other marks) phonons. See the text for details.

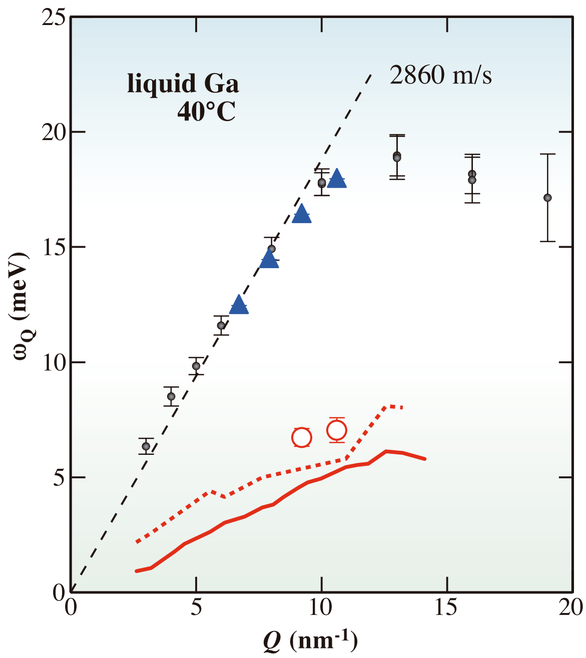

Fig. 2. Dispersion relation of TA (open circles) and LA (full triangles and full circles) phonon excitation modes obtained from the DHO fits. See the text for details.

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References

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