

## OBSERVATION OF FAST SOUND IN EXPANDED FLUID MERCURY ACCOMPANYING METAL-NONMETAL TRANSITION

The metal-nonmetal (M-NM) transition has been an important subject in physics. Since the band picture of electronic states was established, it has been predicted that a divalent metal should transform to an insulating state with volume expansion. Among divalent metals, fluid Hg is the most suitable element for the study of the M-NM transition with volume expansion along the saturated vapor pressure curve because its critical constants are within those experimentally accessible (critical data of Hg:  $T_C = 1478$  C,  $P_C = 1673$  bar,  $\rho_C = 5.8$  g.cm<sup>-3</sup>). The M-NM transition in fluid Hg was first found in electrical conductivity and thermopower data by Hensel and Frank in 1966 and fluid Hg was determined to undergo the M-NM transition with decreasing density,  $\rho$ , from 13.6 g.cm<sup>-3</sup> under ambient conditions to 9 g.cm<sup>-3</sup> near the critical point. We first measured the dynamic structure factor  $S(Q, \omega)$  of fluid Hg at densities from 13.6 to 1.0 g.cm<sup>-3</sup>, including the M-NM transition region at 9.0 g.cm<sup>-3</sup>, using a high resolution inelastic X-ray scattering (IXS) technique [1,2].

This work was carried out at the high-resolution IXS beamline **BL35XU** [3]. Backscattering at the Si (11 11 11) reflection was used to provide an incident beam of 21.747 keV energy and 0.8 meV bandwidth, which was incident onto the sample. Details of the experiments have been described in the literature [1,2].

Figure 1 shows selected IXS spectra,  $S(Q, \omega)/S(Q)$ , at 9.0 g.cm<sup>-3</sup>. The resolution function is shown by a dashed curve at the bottom. The IXS spectra at 9.0 g.cm<sup>-3</sup> have a single peak and the side peaks are not distinct from the central one. Note that the central peaks at 37.1 and 46.8 nm<sup>-1</sup> are very broad at the M-NM transition at 9.0 g.cm<sup>-3</sup>. The spectra were modeled as the sum of the Lorentzian at zero energy transfer and a damped harmonic oscillator (DHO) for the sound mode, with a statistical occupation factor. The optimized model function after convolution using the resolution function is denoted by the bold curves in Fig. 1. Figure 2 shows the  $Q$  dependence of the optimized excitation energy  $\Omega_Q$  (squares) at 9.0 g.cm<sup>-3</sup> obtained from a DHO term together with adiabatic sound velocity (dash-dotted line) obtained by the ultrasonic measurements. Also shown is a dotted line with a slope of 1500 ms<sup>-1</sup>.

As can be seen in Fig. 2, the positive deviation of  $\Omega_Q$  from the dash-dotted line of the adiabatic sound

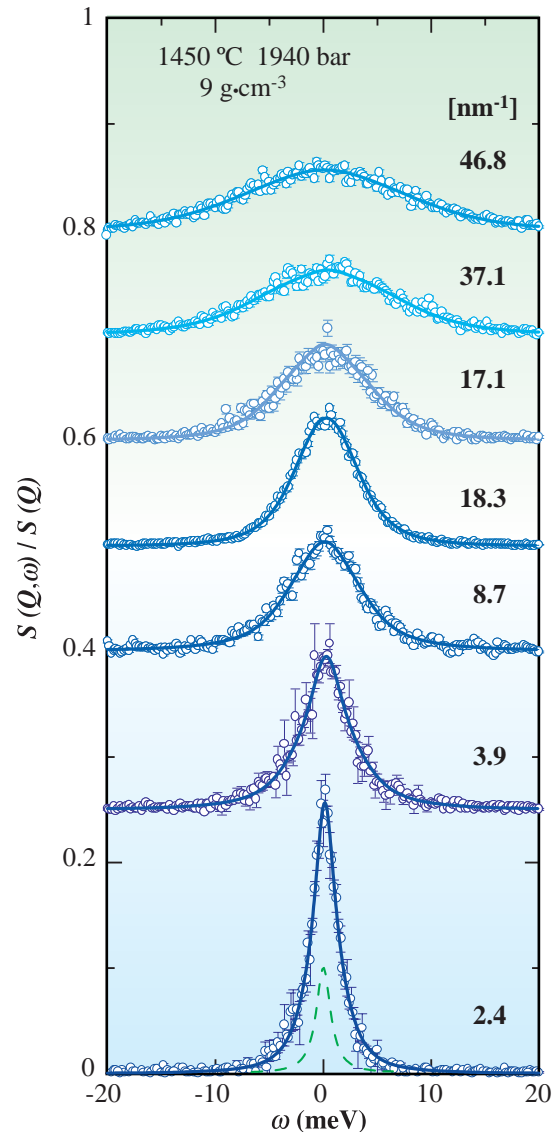


Fig. 1. IXS spectra (open circles) and fits (bold solid curves) of fluid Hg at a density of 9.0 g.cm<sup>-3</sup> at 1450°C and 1940 bar. After subtracting the background, the experimental data were normalized to their integrated intensity.

velocity becomes very large at low  $Q$  and the effective velocity,  $v_s(Q) = \Omega_Q / Q$ , at 2 - 4 nm<sup>-1</sup> is estimated to be 1500 ms<sup>-1</sup>, which is triple the adiabatic sound velocity. We investigated the effective velocity estimated at low  $Q$  as a function of  $\rho$  and compared it with the adiabatic sound velocity. In the metallic region (13.6 - 9 g.cm<sup>-3</sup>), the effective velocity does not decrease much with volume expansion and becomes three

times as large as the adiabatic sound velocity in the M-NM transition. With further volume expansion to the insulating state, the effective velocity becomes small and approaches the adiabatic sound velocity. Thus, a large positive dispersion, fast sound, is observed only in close vicinity of the M-NM transition.

We now discuss how fast sound is induced in the M-NM transition. The average number of nearest neighbors within the first coordination shell is reduced with volume expansion, while the nearest neighbor distance remains unchanged. On the basis of an assumption consistent with these experimental results, Franz [4] succeeded in explaining that the M-NM transition occurs at the correct density of  $9 \text{ g}\cdot\text{cm}^{-3}$ . She pointed out the importance of the reduction and fluctuation of the coordination number in the M-NM transition. When the coordination number is substantially decreased, Hg atoms are considered to move more easily within interatomic distances and fluctuations in the nearest neighboring coordination number are enhanced. Then, a medium-range fluctuation between the metallic and insulating region appears, as schematically shown in Fig. 3. We consider that such fluctuations have a relatively large correlation length of around 1 nm in the M-NM transition and microscopic pressure fluctuations are enhanced due to the deformation of a pair potential

that reversibly changes between the metallic and insulating states in a short period. We conclude that these fluctuations intrinsic to the M-NM transition were attributed to fast sound.

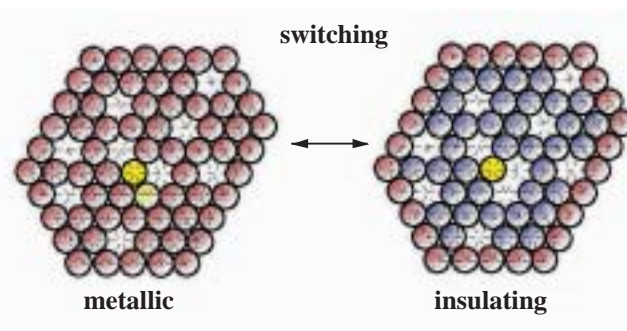


Fig. 3. A schematic illustration to relate the M-NM transition to the observed fast sound. Importance of fluctuations in the coordination number was first pointed out by Franz [4]. When a yellow atom in the left picture loses a neighboring atom (a half-tone yellow atom) and the coordination number becomes three, atoms in the medium range surrounding the central atom (denoted by half-tone blue in the right picture) should transform to an insulating state. Our IXS results suggest that such fluctuations between metallic and insulating states occur reversibly in coincidence with free atomic motion under volume expansion. The central atom experiences fluctuations in pressure due to the deformation of a pair potential resulting mainly from the repulsive part. Switching of the potential on a picosecond time scale results in the high-frequency pressure fluctuations, i.e., fast sound.

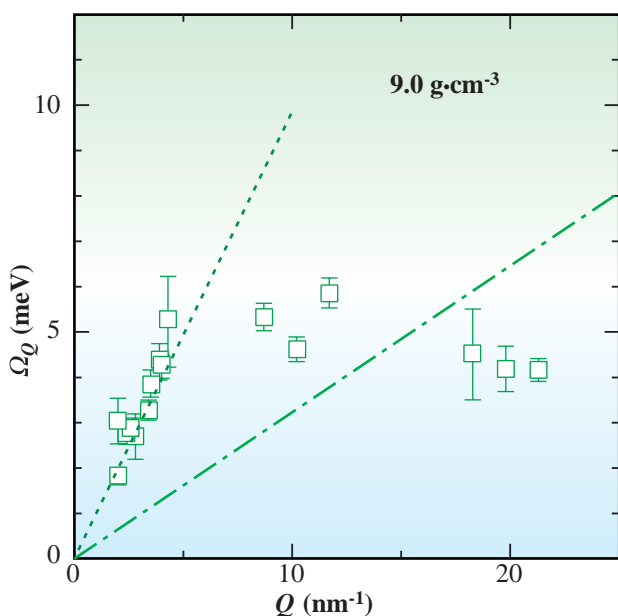


Fig. 2. Excitation energy,  $\Omega_Q$  (squares) as a function of  $Q$  at  $9.0 \text{ g}\cdot\text{cm}^{-3}$ . Dash-dotted and dotted lines correspond to the adiabatic sound velocity at  $9.0 \text{ g}\cdot\text{cm}^{-3}$  and a slope of  $1500 \text{ ms}^{-1}$ , respectively.

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## References

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