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Report:

Monoatomic liquid metals near the melting point show two kinds of ionic excitations: Brillouin excitations, which turn into sound waves on a macroscopic scale, and the Rayleigh excitation, corresponding to non-propagating density fluctuations arising from temperature fluctuations all over the liquid. In the non-metallic gas phase, the formation of dimers leads to an optical-like vibration, visible in light scattering experiments. Neutron scattering experiments on expanded liquid rubidium, performed by the Marburg group, indicate an onset of these dimer-modes already in the metallic liquid close to the metal to nonmetal transition.

The aim of this experiment was to proof the existence of these excitations in liquid sodium by inelastic x-ray scattering. The advantage of inelastic x-ray scattering over neutron scattering for this kind of experiment is the larger accessible range of momentum and energy transfers and especially in the case of sodium the absence of incoherent ionic scattering.

First we characterized the background signal from the sapphire sample cell and the scattering from helium gas inside the autoclave vessel. Then, the scattering by sodium at different temperatures was investigated. Fig. 1 shows the inelastic x-ray spectra, obtained at the same momentum transfer $k = 1.0 \text{ \AA}^{-1}$ for different temperatures. In the solid (lower part in fig. 1), the longitudinal phonon lines at $\omega \pm 13 \text{ meV}$ are the dominant excitations. For liquid sodium close to the melting point ($T = 105^\circ$ in fig. 1) the Rayleigh line is visible as a central peak in addition to the phonon-like excitations (Brillouin lines) For higher temperatures the central line increases rapidly in intensity, whereas the fraction of intensity from the Brillouin excitations is reduced (upper spectrum in fig. 1).

As the dimer-modes and the Brillouin modes are expected to appear roughly at the same positions, a knowledge of the dispersion and the intensity of the Brillouin modes is essential.

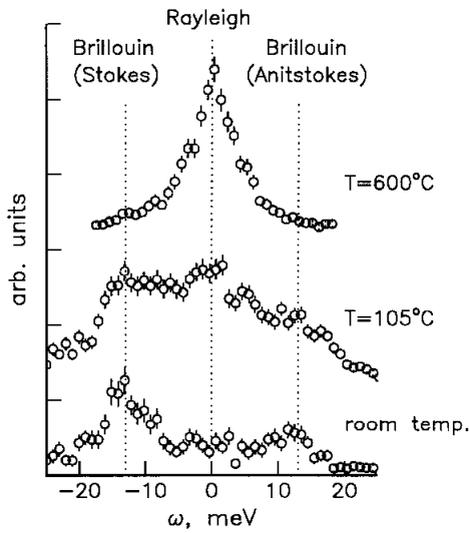


Figure 1: Spectra of solid and liquid sodium at $k = 1.0 \text{ \AA}^{-1}$ for various temperatures.

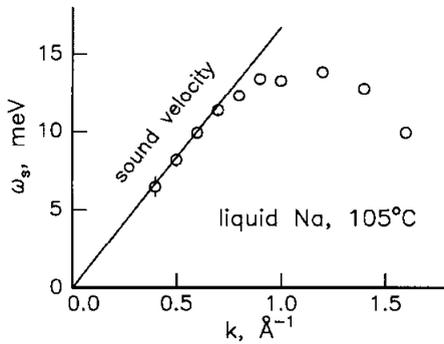


Figure 2: Sound dispersion in liquid sodium near the melting point.

Since no neutron data for the dispersion exist, we started the measurements close above the melting point. Figure 2 shows the Brillouin dispersion obtained by a fit of one central and two asymmetric lorentzians to the data. For small momentum transfers k the slope in the dispersion is equal to the macroscopic sound velocity. At higher k -values, a typical bending of the dispersion is observed, similar to the bending of a phonon dispersion in a solid.

However, in the temperature range investigated so far (100–600°C), no significant observation of a dimer-mode was made. The Brillouin-mode, which is essential for the understanding of a possible dimer mode, could be characterized close the melting point. Further experiments at higher temperatures and lower densities have to follow.