| | Experiment title: | Experiment number: |
|---|---|---|
| | Dynamic structure factors of liquid Si and the other 4B metals | HS1504 |
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**Report:**

Liquid (*l*-) Si has a number of unusual properties, which have prompted several experimental and theoretical studies. Upon melting, Si undergoes a semiconductor-metal transition accompanied by significant structural changes. The molar volume decreases by about 10%, and its coordination number grows from four in the solid state to about 6.5 in the liquid. Despite its metallic nature, however, the structure of *l*-Si is more complicated than that of simple liquid metals like liquid alkalis. Besides the low coordination number, the structure factor maximum comprises a shoulder at the high $Q$ side [1], which cannot be reproduced using a hard-sphere model. These features were interpreted as indications for covalent structures persisting in the liquid metallic state near the melting point. In this report we present the first results of the dynamic structure factor $S(Q,\omega)$ for *l*-Si.

The sample was located in a single-crystal sapphire cell, placed in a high-pressure vessel equipped with Be windows capable to cover scattering angles of 0°-55° [2]. The vessel was filled with 2 bar of high-purity grade He gas. The high temperature of 1480 °C was achieved using a W resistance heater and measured using two W-5%Re:W-26%Re thermocouples. The experiments were performed using x-rays of 17.794 keV (Si(999)) at eight $Q$ values between 3.24-11.8 $nm^{-1}$. A resolution correction was performed by fitting the convolution of a model function and the experimental resolution to the experimental data. As the model function, a Lorenzian was chosen for the central line and a damped harmonic oscillator [3] for the inelastic peaks.

Fig.1 shows the results of $S(Q,\omega)$ at 1480 °C normalized to $S(Q)$ at $Q$ = 3.24-11.8 $nm^{-1}$, where inelastic peaks can be identified. Full circles represent the experimental data with error

bars and the solid line is the best fit of the convolution integral to the data. The experimental resolution function is also given (dashed line). At $Q$ = 3.24 nm$^{-1}$, distinct phonon peaks located at $\omega$ = ± 9.5 meV superimpose the central quasielastic line. With increasing $Q$, the width of both the elastic and inelastic peaks broaden, and especially the phonon peaks get to be highly damped. The spectra in Fig. 1 demonstrate that the dynamics of *l*-Ge is dominated by longitudinal propagating modes, analogous to liquid alkali metals where phonon excitations are well known [4].

The excitation energies $\omega_Q$ are depicted as a function of $Q$ in Fig. 2 by squares. The dashed line represents the dispersion of hydrodynamic sound obtained from recent measurements of the adiabatic sound velocity (3977 m/s). Of particular interest in this dispersion relation is the observation that the positions of the phonon peaks lie on higher energies than the hydrodynamic line. Namely, the so-called *positive* dispersion relation observed in liquid alkali metals [6] is also found in *l*-Si. This finding contrasts with the result in *l*-Ge [5], where no *positive* dispersion was found although it possesses very similar structural and thermodynamic properties.

Further scientific discussions are, however, very limited because of the poor statistics of the present results, *e.g.*, the dispersion relation in Fig. 2 is rather scattered. This was due to a bad quality of W heating wire, which allowed to keep high temperatures only for several hours. A follow-up experiment should be essential to confirm the phonon dynamics in the low $Q$ range, and also to explore localized excitations and the appearance of *optical* modes. Diffusive properties at the $Q$ values of the first peak- (28 nm$^{-1}$) and the shoulder (35 nm$^{-1}$) positions in $S(Q)$, *i.e.*, the *de Gennes* narrowing, are then also to be investigated.


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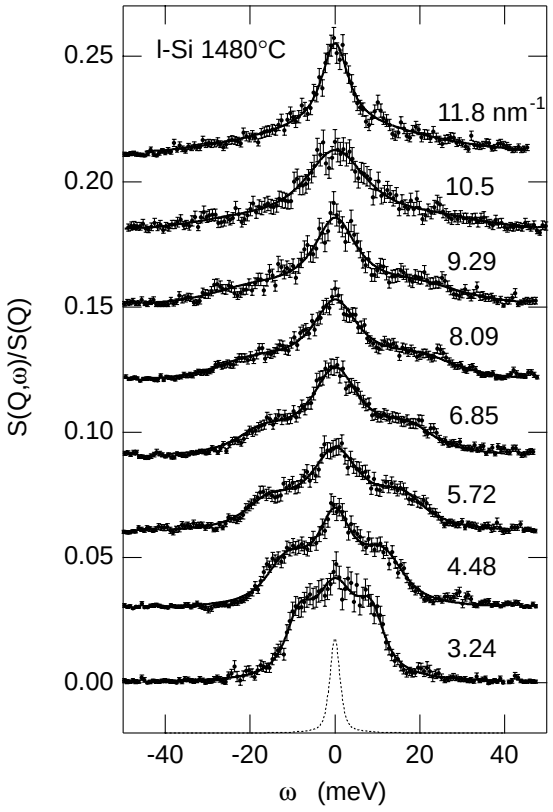


Fig. 1

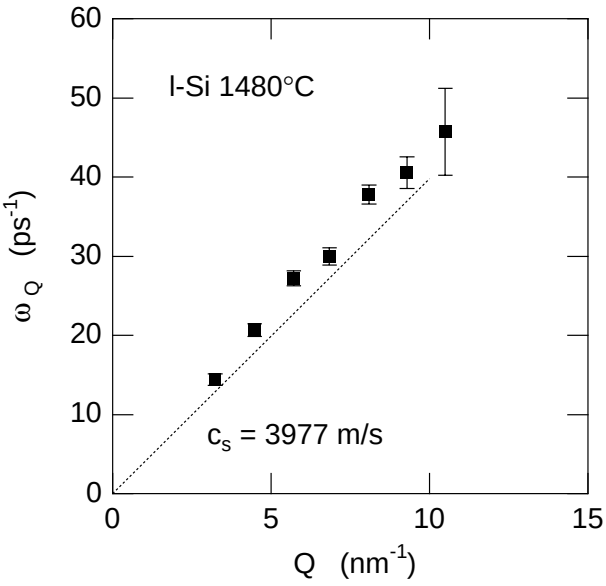


Fig. 2