Inelastic x-ray scattering study of supercooled liquid and solid silicon

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Introduction

Momentum-resolved inelastic x-ray scattering (IXS) technique is one of the powerful methods for the study of dynamical properties of a given system even in extreme conditions like high temperature and high pressure. At the same time, experimental studies of physical and structural properties of liquids have multiplied in recent years with the advent of containerless techniques. These methods reduce the possibility of contamination of specimens and remove external nucleation sites. Therefore, by combining the IXS method with the levitation method, the dynamical properties of stable liquids up to 3000 K and supercooled phase of liquids can be studied.

Silicon is a basic material in the semiconductor industry and has been the subject of a large amount of experimental and theoretical studies over a long time. In the crystalline phase at ambient conditions, silicon is a diamond-structured semiconductor, but upon melting it undergoes a semiconductor-to-metal transition accompanied by significant changes in the structure and density. The coordination number increases from 4 in the solid to about 6.5 in the liquid, and liquid density is increased by about 10%. The principal purpose of the present study was to determine silicon's elastic modulus from the measurement of averaged sound speed determined from IXS.

Methods and Materials

The experiments were carried out at the Advanced Photon Source (APS) beamline 3-ID with a high-resolution monochromator consisting of two nested channel-cut crystals and four backscattering analyzer setups in the horizontal scattering plane 6 m from the sample. The requirements for very high energy resolution and the basic principles of such instrumentation are discussed elsewhere as referenced in [1].

The levitation apparatus was enclosed in a bell jar specially designed for backscattering geometry with a separation of 10 cm between the sample and the detector. Silicon spheres of 2 to 3 mm in diameter were suspended in an argon gas jet and heated with a 270 W CO_2 laser beam. Temperatures were measured during the experiment with a pyrometer whose operating wavelength was 0.65 μm. The temperature gradient on the sample was estimated to be about +/- 20 K. The energy scans were taken for supercooled-liquid and hot-solid silicon at temperature T=1620 K.

Results and Discussion

Sound velocities were determined from the initial slope of the excitation frequencies. Then, the longitudinal moduli for hot-solid and supercooled-liquid silicon were calculated from $L=v_L^2\rho$ using measured velocities. In these calculations, density values were taken from Ohsaka et al. as referenced in [1]. Results are presented in Table 1. together with room-temperature, hot-solid single-crystal measurements, and stable-liquid values. Room-temperature longitudinal moduli were calculated from the values of the single-crystal elastic constants. They were measured between 300 K and 870 K. Since there was no phase transition up to temperature 1620 K for hot-solid silicon, it is reasonable to extrapolate these data to 1620 K in order to compare to our results for the hot solid.

A significant difference (about 20%) is observed between our measurement and the extrapolated single-crystal value of the longitudinal modulus for solid silicon at temperature 1620K. This reduction of the longitudinal modulus may be an indication of the pre-melting. The factor of more than two change in the elastic modulus between supercooled liquid and hot solid at the same temperature can be attributed to the semiconductor-to-metal transition in silicon associated with melting. Also, the longitudinal modulus of the stable liquid is reported in Table 1. About a 10% difference is observed between the modulus of the supercooled and the stable liquid silicon. This can be interpreted as silicon still maintaining metallic properties with a significant increase in the degree of the directional bonding upon supercooling, as found in the x-ray diffraction and ab initio MD studies. All these results are discussed in reference [1].

Table 1. Elastic modulus of the hot-solid and supercooled-liquid silicon measured from our data compared to data for room-temperature single-crystal and stable-liquid taken from the literature.

	Density[e] (gr/cm^3)	Averaged Longitudinal Sound Velocity (m/sec)	Longitudinal Modulus x10^{11}(dyne/cm^2)
300 K[a] RT	2.347	8920[f]	18.68
300 K[b] RT	2.347	8785[f]	18.12
1620 K[b,*] Hot Solid	2.313	8370[f]	16.21[*]
1620 K[c] Hot Solid	2.313	7600+/-200	13.36
1620 K[c] Supercooled Liquid	2.5907	4560+/-100	5.40
1733 K[d] Stable Liquid	2.5718	4370+/-100	4.90

a. from McSkimin (1953) as referenced in [1].
b. from Nikanorov *et al.* (1971) as referenced in [1].
*. Extrapolated from elastic constant data measured at temperature between 300 K and 873 K assuming no phase transition.
c. from present measurement.
d. from Hosokawa *et al.* (2003) as referenced in [1].
e. from Ohsaka *et al.* (1997) as referenced in [1].
f. calculated from longitudinal modulus.

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Reference

[1] A. Alatas, A. H. Said, H. Sinn, E. E. Alp, C. N. Kodituwakku, B. Reinhart, M.-L. Saboungi, D. L. Price, Journal of Physics and Chemistry of Solids 66, 2230-2234 (2005).