

Studies of collective dynamics in liquid Ge over a wide temperature range using inelastic x-ray scattering technique

*S. Hosokawa*¹, *Y. Kawakita*^{1,2}, *W.-C. Pilgrim*¹, and *H. Sinn*³

¹*Institut für Physikalische-, Kern-, und Makromolekulare Chemie, Philipps Universität Marburg, D-35032 Marburg, Germany*

²*Department of Physics, Graduate School of Science, Kyushu University, Fukuoka 810-8560, Japan*

³*SRI-CAT, Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439, USA*

Liquid (*l*-) Ge shows many unusual properties, which in the past have stimulated intensive experimental and theoretical investigations. Upon melting, it undergoes a semiconductor-metal transition in which the density *increases* by about 4.7 % accompanied by structural changes in the short-range order. Despite its metallic nature, the structure of *l*-Ge is more complicated than that of typical liquid metals like liquid alkalis. Besides the undercoordination, the structure factor $S(Q)$ measured near the melting point (937.4 °C) has a shoulder on the high- Q side of the first peak, a feature that cannot be reproduced using simple hard-sphere models. These features were interpreted as indications that covalent structures persist in the liquid state near the melting point. However, they are believed to disappear with elevating temperature. In this paper, we report the first results of the dynamic structure factor $S(Q, \omega)$ at 980 °C [1] and 1500 °C obtained from high-resolution inelastic scattering experiments using intense x-rays from a third generation synchrotron facility.

The experiments were carried out at the beamline ID28 of the ESRF in Grenoble, France, using a newly developed horizontal diffractometer with an energy resolution of 2.48 meV. The sample was located in a single-crystal sapphire cell [2], placed in a vessel equipped with continuous Be windows [3]. The experimental details are given in elsewhere [1,3]

At each temperature, distinct excitations resulting from propagating modes can be identified. Of particular interest in the phonon dispersion relation is the observation that within the experimental error, the positions of the phonon peaks in the low Q range up to 6 nm⁻¹ lie on the hydrodynamic line whose slope is given by the sound velocity c_s . Namely the so-called *positive* dispersion observed in liquid alkali metals [4] is not found.

The most fundamental approach to a theoretical understanding of the collective dynamics in simple monatomic fluids was obtained through the application of generalized hydrodynamics [5], which has successfully been applied to many liquid metal systems. Using an appropriate potential for *l*-Ge near the melting point, a high frequency limit in the low Q region could be calculated to be about twice c_s , and a big *positive* dispersion would be expected beyond $Q \sim 5$ nm⁻¹. This inconsistency with the experimental results at 980 °C may be due to the fact that *l*-Ge near the melting point does not belong to the class of liquids that can be approximated as simple monatomic fluids, i.e., three-body correlations or a distinct covalent nature dominate the collective dynamics. The discrepancy between experiment and theory is also found at a higher temperature of 1500 °C. It should be noted that a recent *ab initio* molecular dynamics simulation [6] showed that the covalent nature in *l*-Ge should be weakened at high temperatures and $S(Q)$ should gradually resemble that of a simple liquid metal.

[1] S. Hosokawa, Y. Kawakita, W.-C. Pilgrim, and H. Sinn, *Phys. Rev. B* **63**, (2001), in press.

[2] K. Tamura, M. Inui, and S. Hosokawa, *Rev. Sci. Instrum.* **70**, 144 (1999).

[3] S. Hosokawa and W.-C. Pilgrim, *Rev. Sci. Instrum.* **72**, 1721 (2001).

[4] W.-C. Pilgrim, S. Hosokawa, H. Saggau, H. Sinn, and E. Burkel, *J. Non-Cryst. Solids* **250-252**, 96 (1999), and references therein.

[5] J. P. Boon and S. Yip, *Molecular Hydrodynamics* (McGraw-Hill, New York 1980).

[6] R. V. Kurkarni, W. G. Aulbur, and D. Stroud, *Phys. Rev. B* **55**, 6896 (1997).