ANOMALOUS DYNAMICAL NARROWING IN LIQUID Se

We have observed strong narrowing in the inelastic X-ray scattering (IXS) spectra of liquid (l-) Se at the momentum transfer *Q* between 12 and 15 nm^{-1} [1]. Typically, in simple monatomic liquids, narrowing is observed at the first structure factor maximum, in this case, about 19 nm^{-1}. However, we see it at a somewhat lower *Q*. This is probably related to the covalent nature of the liquid. Se forms two-fold coordinated linear chain molecules, where atoms are covalently bonded. Crystalline Se with a trigonal form is stable at ambient conditions and it consists of helical chains, while metastable monoclinic forms consist of Se_8 ring molecules. When Se is melted, its two-fold coordinated chain structure is largely preserved and l-Se consists of disordered long chains where segments with a helical chain-like configuration (a) and ring-like one (b) are randomly distributed (see Fig. 1).

We measured the dynamic structure factor *S(Q,E)*, where *E* is the energy transfer, using a high-resolution IXS spectrometer at beamline **BL35XU**. The energy of the incident beam was 21.747 keV and the spectrometer resolution depending on the analyzer crystals was 1.5 - 1.8 meV in the present experimental setup. The Se sample of 99.999 % purity and 0.04 mm in thickness was mounted in a single-crystal sapphire cell. IXS spectra of l-Se at 523 K were measured from 1.8 to 42 nm^{-1} over 40 meV.

Figure 2 shows the overall features of the *S(Q,E)* of l-Se at 523 K. The energy integrals of *S(Q,E)* become the static structure factor *S(Q)*, which agrees well with that obtained from neutron scattering (NS) [2] as shown in Fig. 3(a). *S(Q,0)*, shown in Fig. 2, has a sharp first peak at 15 nm^{-1}, which is a little smaller than the first *S(Q)* maximum. That is, the *S(Q,E)* observed is very narrow at around 15 nm^{-1}.

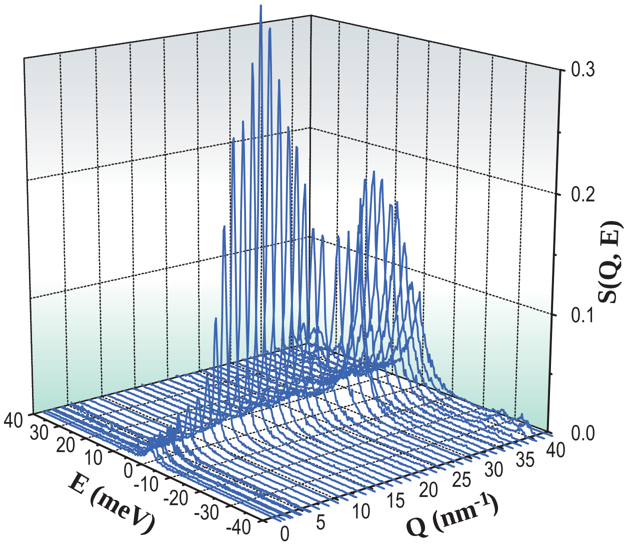

Fig. 2. Three-dimensional plots of *S(Q,E)* of liquid Se at 523 K.

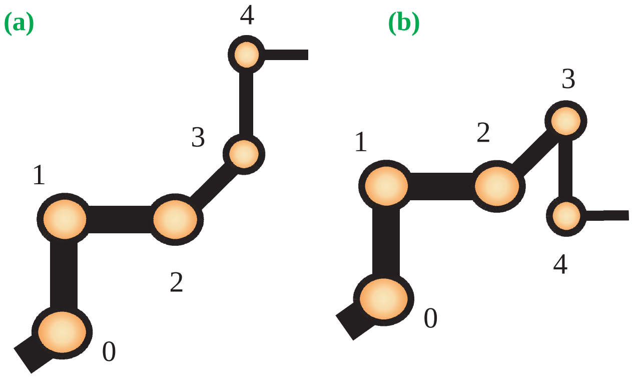

Fig. 1. Schematic illustration of helical chain-like **(a)** and ring-like **(b)** segments along a disordered chain.

The spectra were analyzed using a model function consisting of several Gaussians to deconvolute *S(Q,E)* from the spectrometer resolution. Then we calculated the normalized second frequency moment of *S(Q,E)*, ω_0 *(Q)*, from the deconvoluted model function. Figure 3(b) shows the *E*-*Q* dispersion relation of ω_0 *(Q)* deduced from *S(Q,E)* (triangles). The triangles reasonably follow the solid line in Fig. 3(b) that is calculated using the sum rule, $\omega_0^2 (Q) = k_B T Q^2 / (mS(Q))$, where *m* is a particle mass and *S(Q)* is obtained from NS [2]. More exactly, however, the triangles deviate below the solid line at the *Q* between 12 and 15 nm^{-1}. This discrepancy can be explained if we assume the *Q* dependence of *m*. Figure 3(c) shows the effective mass deduced from ω_0 *(Q)*. The mass as large as 2 - 3 atoms just pinpoints the *Q* where the strong narrowing occurs. The distance corresponding to the *Q* is close to the fourth-nearest-neighbor distance, and it is crucial to distinguish between helical chain-like and ring-like segments in the disordered chain as shown in Fig. 1. Thus, the large effective mass hints that the segments cooperatively move like a rigid molecule under the propagation of longitudinal waves with the corresponding Q. This must be the origin of the present narrowing.

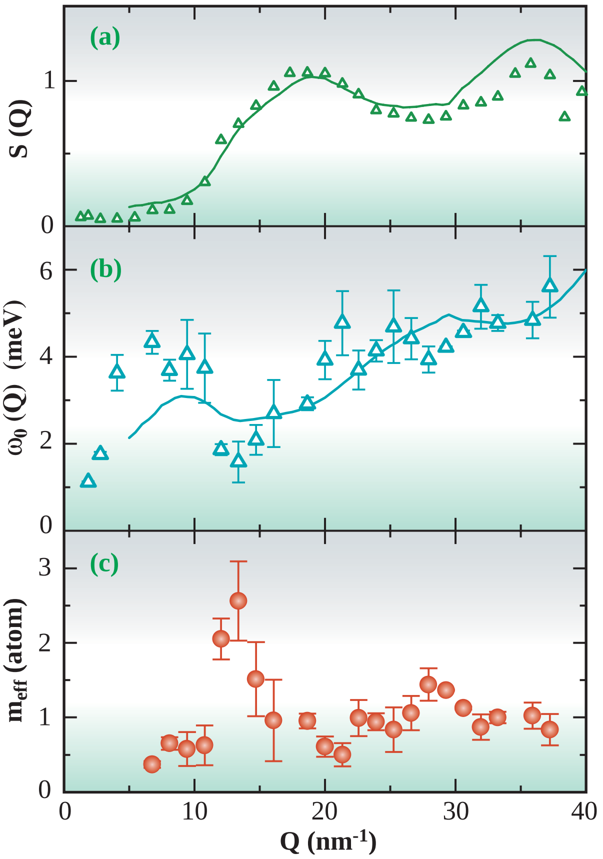

Fig. 3. **(a)** The energy integral of *S(Q,E)* (open triangles) is plotted after being properly normalized. Also shown is the *S(Q)* obtained from neutron scattering (solid line). **(b)** *E-Q* dispersion relation of ω_0 *(Q)* obtained from *S(Q,E)* (open triangles) and sum rule (solid line). **(c)** *Q* dependence of effective mass.

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References

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[2] K. Maruyama *et al.*: J. Phys. Soc. Jpn. **74** (2005) 3213.