Comment on "Short-Wavelength Sound Modes in Liquid Argon"

From an analysis of neutron-scattering data for the density response function $S(k,\omega)$ of liquid argon in the wave-vector range $0.4 < k < 3.8$ Å^{-1} de Schepper *et al.*[1] argue that it is meaningful to employ the concept of extended sound modes even though the measured $S(k,\omega)$ is a monotonically decreasing function of $\omega > 0$, and shows no structure that can be attributed to a collective density oscillation. They attach physical significance to features of the derived dispersion function ω_s which include positive dispersion for $0.4 < k < 1.1$ Å^{-1}, and a gap around the position of the main peak in the structure factor $S(k)$, at $k = 2.0$ Å^{-1}. I believe that there is minimal physical value in these observations since the data are obtained for relatively large k and ω that lie well outside the domain in which linear hydrodynamics is valid. For the k and ω values accessed in the experiment a viscoelastic theory is more appropriate.[2] A viscoelastic response is manifest in the fourth frequency moment of $S(k,\omega)$ which is proportional to the elastic moduli in the limit of long wavelengths. Of the various satisfactory fits to data reported in Ref. 1, case (3), for which the maximum number of sum rules are satisfied, is identical with a fit to a viscoelastic model for $S(k,\omega)$ given in Ref. 2. We can therefore identify the parameters of the fit, such as ω_s, with physically meaningful quantities.

In order to quantify my assertions I display in Fig. 1 various quantities derived from $S(k)$ and the transport coefficients obtained in a computer simulation of a Lennard-Jones fluid[3] which models liquid argon at $T = 86.5$ °K, and density $= 1.418$ g/cm^3. A viscous response is obtained for frequencies that satisfy $\omega\tau \ll 1$, where the Maxwell relaxation time $\tau = 0.36$ ps is the ratio of the viscosity and rigidity modulus. Linear hydrodynamics shows that sound dispersion is $ck + ik^2\Gamma$, and this theory is valid provided the damping does not greatly exceed the sound frequency. The wave vector $c/\Gamma = 0.3$ Å^{-1} is indicated in Fig. 1, together with the frequency $1/\tau$. For small wave vectors and frequencies bounded by these values, the liquid displays a viscous response described by linear hydrodynamics, and the corresponding $S(k,\omega)$ is the sum of three Lorentzian functions.[2].

The dashed curve in Fig. 1 is $\omega_0 = [k^2T/MS(k)]^{1/2}$; ω_0^2 is the normalized second-frequency moment of $S(k,\omega)$. The deep minimum in ω_0 at $k = 2.0$ Å^{-1} means that $S(k,\omega)$ is very narrow, as pointed out by de Gennes.[2] When $\omega\tau \gg 1$, the liquid responds elastically, just as if it were a solid

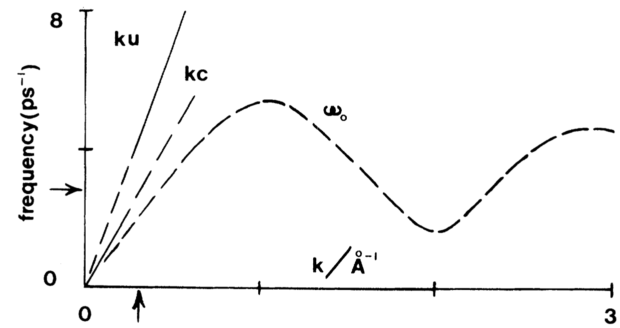

FIG. 1. Quantities defined in the text and computed in Ref. 3 for liquid argon.

body. The velocity of the elastic collective oscillation u ($\geqslant c = 9.1$ Å/ps) is determined by the rigidity and bulk modulii. Hence in liquids that support a short wavelength collective-density oscillation, as the frequency is increased the velocity increases. The observed positive dispersion is likely to be smaller than that obtained from the difference $u - c = 5.3$ Å/ps because of the significant damping and dispersion at the relatively short wavelengths.

By comparing case (3) of Ref. 1 with the model given by Eq. (9.131) of Ref. 2 we identify the various parameters in the former, viz. z_s' and ω_s;

	$k \lesssim 1.7$ Å^{-1} and $k \gtrsim 2.1$ Å^{-1}	1.7 Å$^{-1} \lesssim k \lesssim 2.1$ Å^{-1}
$1/\tau$	$2z_s + z_0$	$z_0 + z_+ + z_-$
ω_0^2	$z_0(\omega_s^2 + z_s'^2)\tau$	$z_0 z_+ z_- \tau$
ω_l^2	$\omega_s^2 + z_s'^2 + 2z_0 z_s$	$z_0 z_+ + z_0 z_- + z_+ z_-$

$\omega_0^2\omega_l^2$ is the normalized fourth frequency moment of $S(k,\omega)$. The fits reported in Ref. 1 therefore provide experimental estimates for the wave-vector dependence of low-order frequency moments and the relaxation time. Values for the latter and the second moment are in accord with previous results[4] and Fig. 1, respectively.

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