

# Reply to Angelani et al.: The $G' \sim L^{-3}$ law for the elasticity of confined liquids can be proved exactly

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Angelani et al. [1] suggest an alternative derivation of the relation between low-frequency shear modulus  $G'$  and confinement length  $L$  derived in [2]. They seem to imply that an integral over plane waves in 3D isotropic media can be decoupled into a separate integral over  $k_z$  ( $z$ -component of wavevector  $\mathbf{k}$ , the confined dimension), times an independent 2D integral in the unconfined ( $k_x, k_y$ ) plane. This would then change the final integration in Eq. 9 of [2] from  $G' = G_\infty - \alpha \int_{1/L}^{k_D} k^2 dk$  to  $G' = G_\infty - \gamma \int_{1/L}^{k_D} dk$ , where  $\alpha$  and  $\gamma$  constants, and the scaling of  $G'$  with  $L$  would change from  $G' \sim L^{-3}$  to  $G' \sim L^{-1}$ .

Angelani et al.’s reasoning [1] is erroneous because they confuse the geometry of real space with the geometry of reciprocal  $k$ -space.

The plane waves in a 3D isotropic system of *any* shape in real space, must satisfy, in  $k$ -space, the relation:

$$\frac{1}{k^2}(k_x^2 + k_y^2 + k_z^2) = 1, \quad (1)$$

with  $|\mathbf{k}| = k = 2\pi/\lambda$ , where  $\lambda$  is the wavelength: see, for example, Eq. 24 on p. 493 in Ref. [3] or Eq. 12 on p. 38 in Ref. [4]. Then, considering the infrared cutoff  $k_{min} \equiv |\mathbf{k}_{min}| = 2\pi\sqrt{(1/L_x)^2 + (1/L_y)^2 + (1/L)^2}$  and confinement along the  $z$  direction where  $L \equiv L_z \ll L_x, L_y$ , gives the lower limit in the integral over  $k$ -space in Eq. 9 of [2] as  $1/L$ . This gives  $G' \sim L^{-3}$  as predicted [2].

The spherical constraint Eq. (1) implies that the 3D integral of plane waves in the isotropic  $k$ -space cannot be decoupled into a separate integral over  $k_z$  times an

independent 2D integral in  $dk_x dk_y$ , contrary to what Angelani et al. are proposing. Integration can be done in Cartesian (or any other) coordinates, but this introduces couplings of coordinates in the integration limits due to Eq. (1) above (Angelani et al.[1] erroneously missed this). As a result, the Debye metric factor  $k^2 dk$  emerges and applies to any shape of 3D isotropic media - the standard textbook result [3–7]. The same spherical constraint applies to the Fermi *sphere* in 3D  $k$ -space for plane waves in a box and holds for parallelepipeds, cubic, cylindrical boxes or any other geometry [3, 4]. Implying otherwise gives the incorrect result that differently-shaped systems give rise to different density of states and thermodynamic properties. In particular, Angelani et al.[1] imply that the phonon density of states is  $\sim \omega$  for a macroscopic 3D cylindrical sample, instead of Debye’s law  $\sim \omega^2$ . This is incorrect.

The detailed and rigorous calculation of the size effect is presented in Ref. [8] and shows that the  $k$ -space integral over a cylinder confined along the  $z$  direction gives:

$$\int d^3\mathbf{k} = \frac{4}{3} \pi k_D^3 - \frac{8}{3} \pi^4 L^{-3}. \quad (2)$$

in agreement with our result in Ref. [2] and the law  $G' \sim L^{-3}$  derived therein.

Angelani et al.[1] mention a “non-universal” exponent between 2 and 3 but do not provide references. The scaling  $G' \sim L^{-3}$  with exponent 3 is clearly seen in several other experiments as well, see Fig. 3 of Ref. [8].

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[4] C. Kittel, Introduction to Solid State Physics (Wiley and Sons, Hoboken NJ, 2005), pp. 111-112, and p. 138.

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