Scopigno et al. Reply: The inelastic x-ray scattering data on liquid Ga presented in our Letter showed that the characteristic lifetime of the extended heat mode is well described by an expression that is based on the structural properties of the liquid augmented by a correction based on the collisional dynamics of hard spheres [1]. In their Comment, Bermejo et al. have now confirmed our x-ray scattering results by an independent technique [2]. However, while we interpret the characteristic length scale associated with the response function as a manifestation of the collisional dynamics not captured in the static properties, Bermejo et al. claim that the new length scale is none other than the hard-sphere diameter itself as determined from the pair correlation function; they assert that the lifetime of the heat mode is predicted by standard single-particle diffusion plus a correction based on the static properties alone, with no need to invoke the presence of structural units exceeding the size of a Gallium ion.

We disagree with this claim. First, the claim that the Ga data can be described without using adjustable parameters requires the usage of the very entity Bermejo et al. object to: a hard-sphere correction function that has an “unphysical” [2] hard-sphere diameter as its argument. Second, there are multiple ways of identifying an equivalent hard-sphere diameter for a liquid based on its static properties. In order to identify the distance of closest approach, one can use the position of the main peak in the static structure factor \( r_{ca} = 0.246 \text{ nm} \) for Ga, an approach used by Cohen et al. [3] to successfully describe the momentum dependence of the dynamics for a range of fluids near the main maximum of the static structure factor, one can use the height of the main peak \( (\sigma = 0.226 \text{ nm}) \), or one can use a comparison between the density at the melting point and the density at which a hard-sphere fluid melts \( (\sigma = 0.262 \text{ nm}) \). This approach has been advocated by van Loef [4] and has been shown to reproduce the oscillations in the half-width of the neutron scattering spectra for liquid mercury [5]. Restricting to real space methods, one could alternatively choose the closest approach distance \( r_{ca} \) [where \( g(r_{ca}) = 0 \), \( r_{ca} = 0.238 \text{ nm} \) in Ga] or even look at the radial probability \( P(r) = 4\pi r^2 g(r) \). As pointed out in our Letter, we find that all of these approaches fail to reproduce the oscillations observed in the Ga spectra, and that instead a larger length scale \( (\sigma = 0.279 \text{ nm}) \) is manifest.

Contrary to the above methods, Bermejo et al. do not equate the distance of closest approach with the hard-sphere diameter; instead they use the most probable interatomic distance (=10% larger). We are not familiar with this approach, and Bermejo et al. do not give any rationale behind it. Other than that, this particular identification would not reproduce the agreement found between the hard-sphere dynamics and the dynamics of range of fluids [3-5]. Moreover, the intrinsic merits of a method that determines a hard-sphere diameter (exceeding the theoretical limit) from static properties while being unable to reproduce these self-same static properties are not immediately clear. We have interpreted this abject failure as evidence that a dynamical length scale associated with the interaction potential is required to understand the response of liquid Ga. Finally, we point out that no commitments can be issued for the validity of Enskog theory in non-hard-sphere systems on the basis of Refs. [2] (only dealing with hard-sphere systems) and [3] (given the very limited explored \( Q \) range) of the Comment.

As such, we cannot support the claim by Bermejo et al. that the dynamics can be predicted based on static properties. In conclusion, the dynamics of liquid Ga exhibit a length scale which is different from the ionic diameter and which is characteristic of the interaction mechanism between the ions. Based on the results for other liquids that are known to show hard-sphere behavior [3] and where, in particular, standard hard-sphere diameters yield a satisfactory description, we do not believe that this new length scale can simply be equated with the average distance between ions; instead, it should be interpreted as a precursor to the structural units present in the solid phase. Whether the dynamics associated with these structural units truly represent or merely mimic hard-sphere behavior remains a topic for further investigation.


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