Comment on "Model for Heat Conduction in Nanofluids"

In a recent Letter, Kumar *et al.* [1] introduced a model for heat conduction in nanofluids (liquid suspensions of nanosized particles) that was capable of describing experimental results on thermal conductivity of nanofluids. The model was built in two steps. In the first step, a static problem (immobile particles) was considered in which the total heat flux was a sum of heat conduction by the liquid and particles. In the second step, the effective thermal conductivity of nanoparticles was calculated in terms of their Brownian motion and the kinetic theory of heat flow. The authors claimed that the resulting formula for the thermal conductivity, using parameters consistent with reasonable physical assumptions, quantitatively described the experimental data.

In this Comment we point out that the treatment of the Brownian motion by the authors of the Letter requires an unphysical assumption about the nanoparticle mean free path and thus overestimates the contribution of Brownian motion to heat flow by several orders of magnitude, thus invalidating the physical justification for the proposed model.

According to the kinetic theory of heat flow, in agreement with Eq. (9) and the last paragraph on page 3 of the Letter [1], the contribution of Brownian motion of nanoparticles to the thermal conductivity, κ_p, is given by [2]

$$\kappa_p = \tfrac{1}{3} n l \nu c_p, \tag{1}$$

where n is the number particle density, l is the particle mean free path, ν is the average velocity, and c_p is the heat capacity per particle. In the estimate of κ_p the authors of the Letter [1] use the velocity that is the ratio of the particle size to time of the diffusive motion over which the particle moves by its size. However, to reach high values of κ_p the authors estimate the mean free path l to be of the order of 1 cm. This assumption is not only unphysical but also inconsistent with the authors reasoning, since the elementary treatment of the diffusive motion leads to the diffusion constant D being proportional to $l\nu$. For internal consistency, with the authors' definition of the velocity, the mean free path should be equal to the particle size. Considering that the particle size is $\sim$10 nm, the effective κ_p would be 6 orders of magnitude smaller than estimated by the authors, thus effectively irrelevant for the heat transport characteristics.

A similar conclusion is reached with a more rigorous treatment of the problem not involving a scale-dependent definition of velocity, and consequently the mean free path. We start with the Langevin equation describing nanoparticle motion [3]:

$$m\frac{d\nu}{dt} = -6\pi\eta R\nu + \xi(t). \tag{2}$$

Equation (2) describes a particle of radius R and mass m moving in a fluid of viscosity η under the influence of the Stokes drag force, $-6\pi\eta R\nu$, and a stochastic force, $\xi(t)$. A simple analysis of Eq. (2) leads to a characteristic relaxation time, $\tau = m/6\pi\eta R$, over which the particle moves in a deterministic manner. The mean free path is then

$$l = \tau\nu = \frac{m\nu}{6\pi\eta R}, \tag{3}$$

where the equipartition of energy gives thermal velocity $\nu = \sqrt{3kT/m}$, where k is Boltzmann's constant. We note this velocity is scale independent; this is in contrast to the scale-dependent velocity used by Kumar *et al.* [1]. By combining Eqs. (1) and (3) and the thermal velocity formula, the thermal conductivity due to particle motion can be expressed as

$$\kappa_p = \frac{nkTc_p}{6\pi\eta R} = \frac{\varepsilon ckT}{6\pi\eta R}. \tag{4}$$

The last equality in Eq. (4) originates from the fact that the heat capacity per particle c_p is equal to the product of the particle volume and the volumetric heat capacity, c; thus, nc_p is simply equal to εc, where ε is the particle volume fraction in the nanofluid.

We can estimate a typical value of κ_p, assuming a 1% volume fraction of $R = 5 \times 10^{-9}$ m gold nanoparticles in water suspension at $T = 300$ K, and the volumetric heat capacity of gold, $c = 130\ \mathrm{J/kg\,K} \times 19\,300\ \mathrm{kg/m^3} = 2.5 \times 10^6\ \mathrm{J/m^3\,K}$. With these values, Eq. (4) gives $\kappa_p \approx 1.1 \times 10^{-6}\ \mathrm{W/m\,K}$. A similar estimate can be obtained within the treatment by Kumar *et al.*, but with consistent values of the mean free path. This value is approximately *6 orders of magnitude* smaller than the thermal conductivity of pure water ($\sim$0.6 W/m K) and therefore has no observable effects on thermal transport.

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[1] D. Hemanth Kumar *et al.*, Phys. Rev. Lett. **93**, 144301 (2004).
[2] See, for example, F. Reif, *Fundamentals of Statistical and Thermal Physics* (McGraw-Hill, New York, 1965), pp. 478–482.
[3] See, for example, S. Chandrasekhar, Rev. Mod. Phys. **15**, 1 (1943).

