Response to "Comment on 'Long-time tails in angular momentum correlations' " [J. Chem. Phys. 104, 7363 (1996)]

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In their comment on our paper, Cichocki and Felderhof point out that our conclusion about the shape independence of the long-time tail in the angular momentum correlation function must be incorrect because it contradicts a "rigorous" result that they have obtained previously. Moreover, they stress that we have incorrectly stated that their theoretical results are limited to the case that the orientation of the particle is fixed.

To start with the second point: It is obvious from Ref. 1, that Cichocki and Felderhof do *not* consider the case of large amplitude reorientational motion. In fact, they say as much in the preamble to the derivation in Ref. 1 where they state that they consider the case of "small amplitude motion." In our paper, we referred to this case as the limit that the orientation is "fixed." Admittedly, it would have been more accurate to speak about an orientation that is "essentially fixed," rather than literally fixed. We thought this to be unnecessarily verbose, because truly fixed orientations are unrealistic—in this limit the angular velocity autocorrelation function (AVACF) does not exist.

Let us then compare the two cases of real interest, namely the Cichocki–Felderhof limit of small amplitude angular motion and the limit of large amplitude reorientation that we considered in our paper.

Cichocki and Felderhof point out that for small amplitude motion, the AVACF does depend on the shape of the particle, and *we completely agree*. More interestingly, we were happy to notice that the theoretical predictions of Cichocki and Felderhof for this limit are *in excellent agreement* with the numerical results that for nonspherical objects *under conditions where the angular displacement is negligible.* For small amplitude motion, the shape and frequency dependent friction coefficient is proportional to the memory function of the AVACF and gives a shape dependent tail. Thus, so far as the calculation of the friction coefficient is concerned, the particle is fixed ("essentially fixed" in our present terminology). The calculation described in Ref. 1 neglects the effect of the particle's orientational motion on the frequency dependent rotational friction coefficient. Our simulations show quite convincingly, that for a particle that is "essentially fixed," a shape dependent decay of the AVACF is indeed observed. It is reassuring that our model, which uses a relatively crude lattice representation of a nonspherical object, is in essentially quantitative agreement with the relevant theoretical predictions. This gives us great encouragement that we can reliably extend our simulations to the case of concentrated suspensions of non-spherical objects.

Next, let us consider the case discussed in Ref. 2, viz. the effect of reorientation on the dynamics of the particle. Reorientation is important because, on a sufficiently long timescale, i.e., in the truly asymptotic regime, reorientation is always important. In Ref. 2 we stressed that the theory contained in Ref. 1 assumes that reorientation is negligible and is therefore not appropriate (let alone "rigorous") in this limit. However, the microscopic theory contained in Ref. 4 should apply in the limit of large-amplitude reorientation. It predicts that reorientation changes the decay of the AVACF in such a way that it becomes identical to the result for a spherical object with the same moment of inertia. A careful microscopic analysis produces kinetic equations that, in the Brownian limit, reduce to the Navier–Stokes equations (plus appropriate boundary conditions). Not all the mode-coupling or kinetic theory treatments, which have been proposed, give this limit correctly.[3] This is possibly the reason why Cichocki and Felderhof, in their comment, are somewhat skeptical about the reliability of such theories. However, the theory contained in Ref. 4 is not a normal mode coupling or kinetic theory. In macroscopic terms it amounts to obtaining the long time limit of the friction coefficient by solving the time dependent Navier–Stokes equations for fluid flow around a particle whose orientation at a given time is determined by the rotational diffusion equation. In these terms it is similar to the theory contained in Ref. 1–except that it does not impose the restriction that the orientation of the object remains unchanged. In any event, for our simulations, we did not have to assume that *any* of the theoretical treatments were correct. The purpose of the computer simulations was simply to test theory.

In essence, the simulations reported in Ref. 2 showed a shape dependent amplitude for the long-time tail for a particle undergoing small amplitude motion, as expected. In order to study the effects of reorientation we also performed simulations where the angular displacement was large. The extent of the angular displacement during the simulation was varied by varying the initial angular velocity of the particle. By arranging for a large reorientation (using a high initial velocity), we saw that the amplitude of the AVACF was

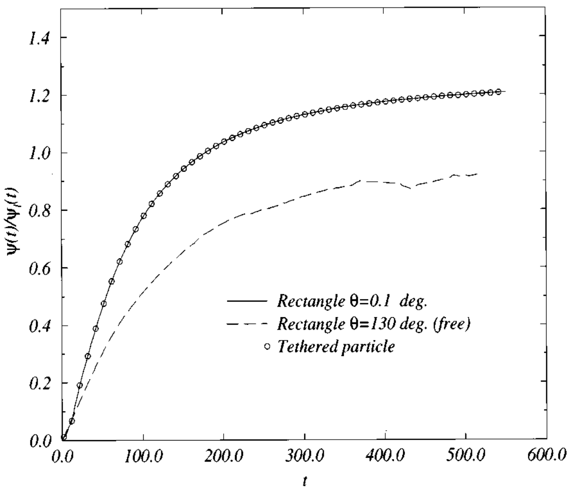

FIG. 1. The angular velocity autocorrelation function, $\psi(t)$, for a rectangle of width 3 and length 11 in two dimensions. $\psi(t)$ has been normalized by the theoretical long-time result, $\psi_l(t)$, for a disk with the same initial angular velocity and moment of inertia. The solid line is the result for a rectangle undergoing an angular displacement of 0.1 deg, the broad dashed line is the result for a rectangle undergoing an angular displacement of 130 deg. The circles are the results for a rectangle with the same initial angular momentum as for the run with a large angular displacement, but in this case tethered (i.e., we have not updated the orientation).

indeed affected, and that the decay approached the asymptotic decay for the equivalent spherical object. This is what the microscopic theory predicts.[4] In their comment, Cichocki and Felderhof seem to be concerned that what we saw was an effect of the high initial velocity and not of the reorientation. This would be surprising because the dynamics of the Lattice Boltzmann fluid in which we immersed the object are governed by the linearized Navier–Stokes equations. As described by Ladd,[5] this is easily arranged by using a suitable equilibrium distribution. The only possible nonlinearity comes from the way the boundary conditions are imposed. In our work we checked that the effect we saw was genuinely due to reorientation rather than the magnitude of the initial velocity. To do this we repeated the calculation with the high initial velocity, but considered the particle to be tethered. That is, we updated the angular velocity, so the dynamics were determined by the time-dependent velocity fields, but we did not change the orientation. The results of this procedure were omitted from Ref. 2 in the interests of brevity. However, in light of the comment we include the results here in Fig. 1. Clearly, the results for the tethered particle with high initial angular momentum are essentially identical to the results for the (untethered) particle undergoing negligible rotation. We can conclude that the effect we see when we actually allow the particle to reorient itself is an effect due entirely to the reorientation and not to the high initial velocity.

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[4] A. J. Masters and T. Keyes, J. Stat. Phys. **39**, 215 (1985).
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