Supplemental Material for:

“Overestimation of viscosity by the Green-Kubo method in a dusty plasma experiment”

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I. Experimental Setup

The experimental apparatus is sketched in Fig. SM1. The plasma was generated in a vacuum chamber by applying 13.56 MHz power to a horizontal electrode, while the outer chamber walls were grounded. There was negligible gas flow.

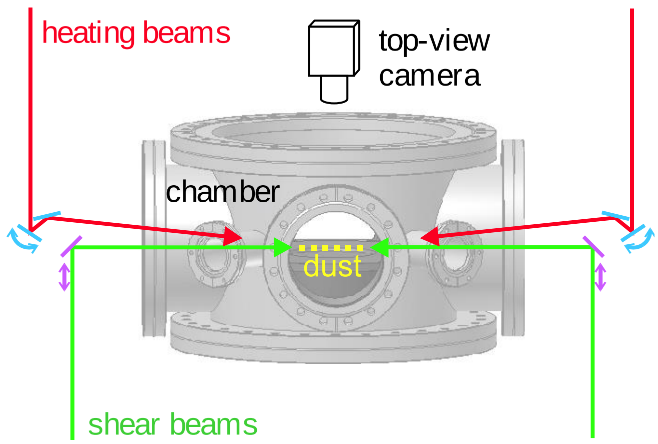

FIG. SM1. Sketch of experimental apparatus. Our polymer microspheres are labeled “dust” here.

After the microspheres were introduced and they settled into a horizontal monolayer, they had so little vertical motion that it was undetectable by our side-view camera, not shown in Fig. SM1. Based on the precise microscopic measurements Samsonov *et al.* [1] in a dusty plasma similar to ours, we estimate that the vertical motion of microspheres in our monolayer was less than 10 μm. This low level of vertical motion, which was on the order of a particle diameter, would not permit particle transport by buckling of the monolayer (when a particle moves past another by jumping over in the out-of-plane direction).

The microspheres were illuminated by a horizontal sheet of laser light, not shown in Fig. SM1, and imaged from above by the top-view camera, which was a Phantom Miro M120. Each experimental run corresponded to recording one video, lasting 62.6 s as limited by the camera’s memory, while keeping conditions steady. While the 70 frames/s framerate we used was lower than that used by Feng *et al*. [2] in their experimental demonstration of the Green-Kubo method in a 2D dusty plasma, this framerate is higher than was used in the other viscosity-related dusty plasma experiments of Nosenko *et al.* [3] and Hartmann *et al.* [4].

Two types of laser manipulation were used in the experiment, heating and shear, provided by two separate pairs of laser beams, as sketched in Fig. SM1. The heating beams moved back and forth over the entire layer of microspheres, following a pattern of randomized circular arcs that was optimized to cause the microsphere motion to mimic that of particles in thermal equilibrium at elevated temperature [5]. This manipulation effectively melted the ground state crystal of the microsphere layer, as evidenced by the fact that the kinetic temperature of the microspheres was above the expected melting point [6, 7]. Moreover, we inspected the paircorrelation functions *g(r)* for our heated microspheres and found none of the long-range order indicative of a crystalline phase.

The shear beams were shaped like horizontal ribbons and directed in the plane of the microsphere layer with a gap between them to create a shear flow for the hydrodynamic viscosity measurement [7]. The shear laser was operated at a power of 118 mW, and the ribbon shaped beams had a width of 3.5 mm and thickness of 1.2 mm, with a gap of 4.0 mm between them. The beams were linearly polarized, and two rotating polarizers were used to attenuate the beams individually, so that the power in each beam could be matched [7]. The polarization of the shear beams has no effect on the microspheres because the radiation pressure force depends only on the laser beam's intensity, and it has no effect on the electrons or ions because the beam's intensity is many orders of magnitude too small. Inside the chamber, these beams were weak enough that they did not affect the plasma, and only provided a weak force on the particles. The two beams drove a shear flow that had a peak velocity of only half the thermal velocity of the particles [7]. We chose to drive such a weak flow in order to avoid shear thinning and the generation of excess viscous heat that would cause a nonuniform temperature across the flow.

These flow velocities in the runs with shear were 0.5 mm/s or less, which means that the current due to flowing dust particles was less than 1 pA. The magnetic field generated by this current is more than ten orders of magnitude less than that of Earth.

The viscous transport of the microsphere component of the dusty plasma was unaffected by the other components (electrons, ions, and neutral gas atoms). The main effect of the electrons and ions was to screen the microspheres' repulsive potential [2]. The gas atoms played the role of a background that applied a weak friction to the particles. Because the gas was rarefied, it contributed nothing to the momentum exchange between two particles, so it had no effect on viscous transport of momentum within the particle component [2, 7].

II. Time Series of P_{xy}

After obtaining microsphere positions and velocities from video data, we calculated a time series of shear stress $P_{xy}(t)$, using Eqs. (3-4). As is necessary for a long-range repulsion such as the Debye-Hückel (DH) potential, we cut off the potential at maximum distance, which we chose as 6λ. Also, as we did in calculating $P_{xy}(t)$ for the hydrodynamic method in [7], we interpolated particle positions between consecutive frames so that positions were recorded at the same time as velocities. The velocities were obtained at times between each frame simply as the change in particle position divided by the time between frames [8].

In Fig. SM2, an example of the time series $P_{xy}(t)$ is shown, for an entire run (left panel) under shear-free conditions, and for only a 4.0 s portion (right panel) of the same run. We inspected the power spectra of these time series to verify that they are not dominated by a few particular frequencies.

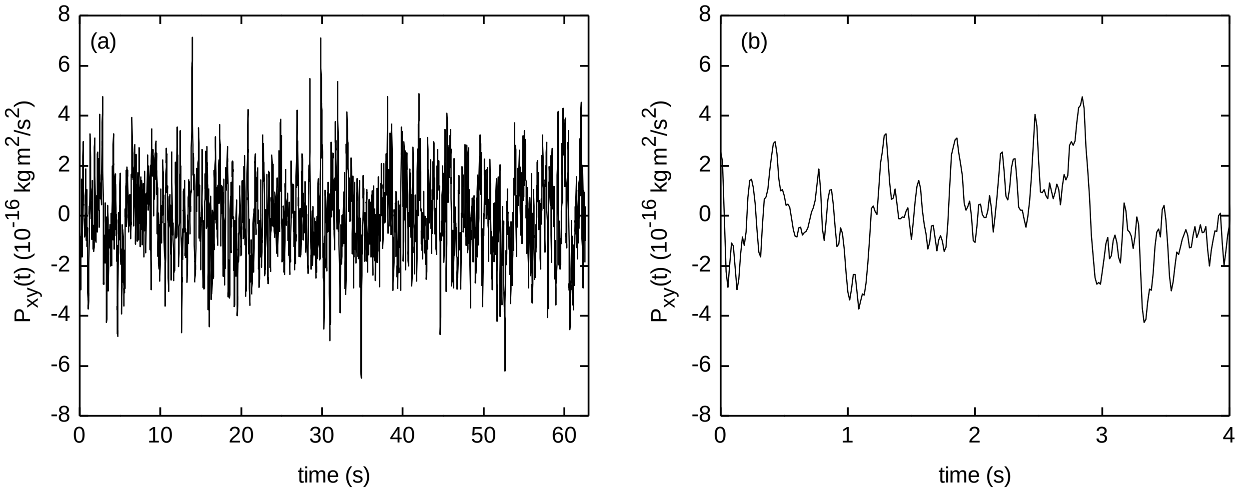

FIG. SM2. Time series of P_{xy} over (a) an entire shear-free run and (b) the first 4.0 s of the same run.

The Green-Kubo (GK) theory, since it was intended for systems under equilibrium conditions, assumes that $P_{xy}(t)$ is a quantity that fluctuates around zero. A nonzero average value of $P_{xy}(t)$ would give an unphysical extra contribution to the integral of the stress autocorrelation function C_η, and therefore an unphysical increase to the GK viscosity values [2]. Such a nonzero average of $P_{xy}(t)$ could be an artifact of our heating method or of the fact that our camera's field of view included only a portion of the microsphere layer. To eliminate this small unphysical artifact we subtracted the time-averaged value of $P_{xy}(t)$, which was always about an order of magnitude less than the rms fluctuations of the time series.

III. Random and systematic errors

We have performed several tests to check whether our finding that the GK method overestimates viscosity could be affected by various random or systematic errors, or by traits of our experiment.

Particle position measurements are made directly from our recorded videos, and we have verified that small errors in them do not affect our results for viscosity with the GK and hydrodynamic methods [9]. Our verification consisted of repeating our viscosity calculations after adding Gaussian random errors, with an rms value of 0.5 pixel, to each microsphere position. We found that this additional error had negligible effect on our final viscosity values. Since our velocities are also obtained from the positions, this test convincingly demonstrates that errors in microsphere positions and velocities cannot account for the 60% overestimation.

We also checked the effects of errors in our values of Q and λ. These two parameters were obtained from the minimum of a χ^2 plot, so that they have uncertainties; these uncertainties are essentially random errors. Additionally, these parameters have small systematic errors resulting from a slow drift of experimental conditions. We now discuss the effects of these random errors and drift separately.

We have determined that the random errors in Q and λ contribute an error of less than 10% to our final GK viscosity results. We quantified the random errors in Q and λ by mapping the value of χ^2 from the fitting process and finding a contour corresponding to a one-sigma uncertainty [7]. We then repeated our GK analysis using various pairs of values of Q and λ from the extremes of their uncertainty ranges, and we found that variation in these parameters resulted in viscosity variations of about 10% or less. The GK and hydrodynamic viscosities varied similarly when varying Q and λ, so that the discrepancy between the two types of results remained nearly the same. This finding about the discrepancy supports our argument that errors due to the DH interparticle potential model enter our viscosity results from both methods in the same way and cannot be responsible for the overestimation we reported for the GK method.

There were also small systematic drifts in Q and λ during the experiment, as discussed in detail in [7]. The value of Q drifted slightly from -15500 e to -15900 e, and λ drifted slightly from 0.38 mm to 0.42 mm. The corresponding drift in the nominal 2D dusty plasma frequency ω_{pd} was from 86 s^{-1} to 92 s^{-1}. To assure that small drifts did not affect our results, we used interpolated values of these parameters for each run in all our analysis to obtain viscosity.

The final systematic error, as mentioned in the Letter, is due to the GK integration limit. We estimated this error by varying the integration limit and observing the change in viscosity. In [2], this limit was chosen to be the time at which C_η first crossed zero, so we varied our limit from the first zero-crossing down to our choice of the time when the C_η decayed to 7% of its initial peak. We found that our viscosity values diminished by only 1% to 30%.

IV. Debate about validity of Green-Kubo in low-dimensional systems

As we mention in the manuscript, there are arguments against the applicability of GK methods in low-dimensional systems. These arguments began in 1970 with a report that GK integrals appeared not to converge in 2D simulations with small numbers of hard disks [10]. Later, mode-coupling theory suggested that GK methods generally fail in low-dimensional systems [11-13]. This debate remains unsettled, however, because larger system sizes and longer time series are required to test convergence in simulations, as reported recently [14]. In fact, these recent simulations [14] have suggested that the GK integral for viscosity does converge for 2D particles interacting with the DH potential at the temperatures achieved in our experiment.

V. Dismissing instrumental effects as reasons for viscosity overestimation

We mention in the manuscript that we can dismiss as the reason for the overestimation three instrumental effects: neutral gas, anisotropy, and erroneous inputs. We address each of these aspects below.

First, there is neutral gas that collides constantly with the microspheres, and one might ask whether this could affect the transport of momentum carried by the microspheres. For example, one could imagine a situation where a microsphere could push a gas molecule forward so that it then collides with another gas molecule before colliding again with another microsphere and thereby impart momentum indirectly from one microsphere to another. This possibility was already considered [16] and shown to have no effect on the results of the GK method, even for gas densities eightfold higher than ours.

Second, anisotropy is present in the motion of our microspheres, as in previous experiments [5, 17, 18]. The anisotropy is a trait seen in the kinetic temperature in our experiment. There is a 40% difference between T_x (obtained using only v_{ix}) and T_y (using only v_{iy}), due to our use of two heating beams [7]. The GK theory does not account for such anisotropy. While we cannot entirely rule out anisotropy as a candidate, we have performed a test that casts doubt on this possibility: we repeated our GK method calculations using T_x or T_y instead of T, and we found that the viscosity overestimation was unaffected. Therefore, it is unlikely that this temperature anisotropy can account for the observed viscosity overestimation by the GK method.

Third, as always we must be wary of erroneous inputs, which in our experiment include the microsphere positions and velocities as well as the model we assume for the interparticle potential. We have verified that errors in positions and velocities cannot cause a significant effect, as described above in Section III. We also performed a test allowing us to dismiss the potential as a significant source of error; in this test, also described above in Section III, we repeated our calculations with varying values of the input parameters (Q and λ) in the potential, and we found virtually no effect on the overestimation.

VI. Other schemes for a test of the Green-Kubo method

One might ask whether, instead of comparing two experimental results as we do, a test of the GK method's applicability could be performed by comparing an experiment to a simulation. In fact, such comparisons are made often by theorists [19-21], who obtain GK viscosities from simulations and compare them to hydrodynamic viscosities from experiments. These comparisons do not, however, serve as tests of the GK method because simulations cannot distinguish whether a viscosity differs from an experimental value due to a bad model for Φ_{ij} or a failure of the GK method itself. In practice, when making such comparisons, theorists generally question the potential model and not the GK method [22, 23].

Another approach to testing would be a comparison of two theoretical results: GK equilibrium simulation vs nonequilibrium simulation or theory. We expected that the literature should have many such comparisons, but in our search we found only a few comparisons for viscosity [24, 25], which are not presented as tests of the GK method [26]. Analogous comparisons are somewhat more common for the self-diffusion coefficient, such as [27], but again these are generally not presented as tests of the GK method.

References:

[1] D. Samsonov, J. Goree, H. M. Thomas, and G. E. Morfill, Phys. Rev. E **61**, 5557 (2000).
[2] Y. Feng, J. Goree, B. Liu and E. G. D. Cohen, Phys. Rev. E **84**, 046412 (2011).
[3] V. Nosenko and J. Goree, Phys. Rev. Lett. **93**, 155004 (2004).
[4] P. Hartmann, M. C. Sándor, A. Kovács, and Z. Donkó, Phys. Rev. E **84**, 016404 (2011).
[5] Z. Haralson and J. Goree, IEEE Trans. Plasma Sci. **44**, 549 (2016).
[6] P. Hartmann, G. J. Kalman, Z. Donkó, and K. Kutasi, Phys. Rev. E **72**, 026409 (2005).
[7] Z. Haralson and J. Goree, Phys. Plasmas **23**, 093703 (2016).
[8] Y. Feng, J. Goree, and B. Liu, Rev. Sci. Instrum. **82**, 053707 (2011).
[9] We performed this verification with the data from our newly reported shear-free runs, as we did previously in [2] for the shear runs, which were used with the hydrodynamic method.
[10] B. J. Alder and T. E. Wainwright, Phys. Rev. A **1**, 18 (1970).
[11] M. H. Ernst, E. H. Hauge, and J. M. J. van Leeuwen, Phys. Rev. Lett. **25**, 1254 (1970).
[12] J. R. Dorfman and E. G. D. Cohen, Phys. Rev. A **6**, 776 (1972).
[13] Y. Pomeau and P. Résibois, Phys. Rep. **19**, 63 (1975).
[14] Z. Donkó, J. Goree, P. Hartmann, and B. Liu, Phys. Rev. E **79**, 026401 (2009).
[15] H. Stassen and W. A. Steele, J. Chem. Phys. **102**, 8533 (1995).
[16] Y. Feng, J. Goree, and B. Liu, Phys. Plasmas **18**, 057301 (2011).
[17] V. Nosenko, J. Goree, and A. Piel, Phys. Plasmas **13**, 032106 (2006).
[18] J. Schablinski, D. Block, A. Piel, A. Melzer, H. Thomsen, H. Kählert, and M. Bonitz, Phys. Plasmas **19**, 013705 (2012).
[19] R. Eggenberger, S. Gerber, H. Huber, D. Searles, and M. Welker, Chem. Phys. **164**, 321 (1992).
[20] S. H. Lee, Bull. Korean Chem. Soc. **34**, 2931 (2013).

[21] K. Nieszporek, J. Nieszporek, and M. Trojak, Comput. Theor. Chem. **1090**, 52, (2016).
[22] Y. Sakiyama, S. Takagi, and Y. Matsumoto, J. Chem. Phys. **122**, 234501 (2005).
[23] N. Meyer, H. Xu, and J.-F. Wax, Phys. Rev. B **93**, 214203 (2016).
[24] S. H. Lee, G. K. Moon, and S. G. Choi, Bull. Korean Chem. Soc. **12**, 315, (1991).
[25] Z. Donkó, J. Goree, P. Hartmann, and K. Kutasi, Phys. Rev. Lett. **96**, 145003 (2006).
[26] One limitation of a test using a comparison of two simulations would be that the nonequilibrium simulation requires boundary conditions and a thermostat that only artificially mimic an experimental flow.
[27] T. Haxhimali, R. E. Rudd, W. H. Cabot, and F. R. Graziani, Phys. Rev. E **90**, 023104 (2014).