Overestimation of Viscosity by the Green-Kubo Method in a Dusty Plasma Experiment

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The Green-Kubo (GK) method is widely used in simulations of strongly coupled plasmas to obtain the viscosity coefficient. However, the method's applicability, which is often taken for granted, has not been tested experimentally. We report an experimental test using a two-dimensional strongly coupled dusty plasma. We find that the GK viscosity is ≈60% larger than the result of a benchmark hydrodynamic method, obtained in the same experiment with the same conditions except for the presence of shear.

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Viscosity, which describes momentum transfer in a flowing fluid, is a transport coefficient used widely in fluid mechanics [1–3], materials science [4,5], nanoscience [6], particle physics [7], biophysics [8], and other fields. At a microscopic level, viscosity arises from collisions, but at a macroscopic or hydrodynamic level it is defined by a constitutive relation [9],

$$\bar{P}_{xy} = -\eta\gamma. \tag{1}$$

Here, the steady applied shear stress is $\bar{P}_{xy}$, which corresponds to a transverse momentum flux. The shear flow is characterized by γ, which is the transverse gradient in the steady flow velocity.

As it does in fluids, viscosity in plasmas affects instabilities [10], waves [11–13], vortices [14], and heating [15]. Despite these similarities, plasmas have unusual viscosity properties because the underlying Coulomb forces have a long range, unlike the short-range interactions typical of liquids and gases.

Viscosity is an especially important parameter in plasmas that are strongly coupled [16–18]. Strong coupling, which means that the average interparticle potential energy exceeds the thermal kinetic energy, occurs in white dwarf and giant planet interiors, inertial confinement fusion, and electrons on the surface of liquid helium [19]. Other examples of strong coupling include ultracold plasmas [20], pure ion plasmas [21], and dusty plasmas [22]. Instead of behaving like a gas, charged particles in a strongly coupled plasma behave like a liquid if the density is high enough, the temperature is low enough, or the particle charge is high. In these liquidlike strongly coupled plasmas, the particles can flow collectively, and in such a flow viscous dissipation plays a large role. For these reasons, there have been many theoretical studies of the viscosity of strongly coupled plasmas [18]. While these studies are important due to their influence on other theories that use the viscosity, they are usually done with simulations that bear little resemblance to the standard experimental methods of measuring the viscosity.

One standard experimental method, for simple liquids, is what we call the "hydrodynamic method." Experimenters apply a steady shear stress at a boundary, measure the resulting steady velocity gradient, and divide the two to obtain the viscosity using Eq. (1). Theorists, however, are less likely to use this hydrodynamic method when obtaining the viscosity from molecular dynamics simulations. Such a nonequilibrium simulation would impose the complications of how to sustain a macroscopic gradient and a steady temperature, while eliminating the viscous heat that is produced. These complications lead to a need for subtle boundary conditions and a carefully chosen thermostat [23,24].

For strongly coupled plasmas, it is common for theorists to use an equilibrium simulation [25,26], avoiding the complications of simulations based on the hydrodynamic method. A transport coefficient is obtained by the Green-Kubo (GK) method [9,27–29], with an input of the particle motion recorded in the simulation. The GK method has been used to obtain plasma transport coefficients since at least 1971 [30].

In this Letter, we use experimental data to test whether the widely used GK method of obtaining the viscosity is applicable to a strongly coupled plasma. Our literature search revealed widespread theoretical use of the GK method [17,18,25,26,31–37], but no previous experimental tests. A well-designed test requires comparing experiments done two ways, under nearly identical conditions: one way with a shear flow (for the hydrodynamic method), and the other with no flows (for the GK method). We use a strongly coupled dusty plasma, which allows a controlled shear flow. It also allows particle tracking, which yields the particle position and velocity data needed as inputs for the GK method.

A dusty plasma consists of ionized gas containing small particles of solid matter. Dusty plasmas are found in interstellar clouds, comet tails, planetary rings [38,39], and manufacturing plasmas [40,41]. The heaviest component of a dusty plasma, the solid particles, gain a negative electric charge Q, which is typically many thousands of elementary charges [42]. This large charge causes interparticle interactions to be very strong, so that laboratory dusty plasmas are often strongly coupled [22]. These solid particles can easily be made to flow, due to various forces such as the radiation pressure force from an externally applied laser beam [43]. The electron and ion components of a dusty plasma provide screening of the repulsive potential Φ between the solid particles.

Theorists widely use the GK method to predict the viscosity of strongly coupled dusty plasmas, for example Refs. [17,31,33,34] and as reviewed in Ref. [18]. In performing their simulations, theorists usually do not question whether the GK method is applicable. Like most theoretical descriptions of plasmas, the GK method assumes such fundamental concepts as conservation of momentum and mass, but nevertheless it is not applicable for every substance. The substance of interest here, a strongly coupled plasma, has several special characteristics that lead us to question whether the GK method is accurate. One such characteristic of strongly coupled plasmas is non-Newtonian behavior, for example, shear thinning [44] and the memory effect of viscoelasticity [45], which have both been observed experimentally [46–48]. Non-Newtonian fluids, in general, can lack the proportionality between a current and force that is required for applicability of the GK method [49]. Besides non-Newtonian behavior, we can identify two characteristics of strongly coupled plasmas that are uncommon in simple liquids: a long-range interaction, and a minimum in the viscosity's temperature dependence [44]. All the characteristics we have listed here give reason to question the accuracy of the GK method for strongly coupled plasmas, motivating our experimental test.

The Green-Kubo relation.—The GK relation for viscosity is

$$\eta = \frac{A}{k_B T} \int_0^\infty C_\eta(t)dt, \tag{2}$$

for a temperature T. The stress autocorrelation function is $C_\eta(t) = \langle P_{xy}(t_0) P_{xy}(t_0 + t) \rangle$, where the ensemble average indicated by the brackets $\langle\rangle$ is typically computed as an average over starting times t_0. Equation (2) is written for a two-dimensional (2D) liquid; for a 3D liquid, the area A would be replaced with a volume.

The instantaneous shear stress is calculated from microscopic-level data as [9]

$$P_{xy} = \frac{1}{A} \sum_i \left(m v_{i,x} v_{i,y} - \frac{1}{2} \sum_{j \neq i} |x_j - x_i| \nabla \Phi_{ij} \cdot \hat{y} \right), \tag{3}$$

which fluctuates in time. Here, the inputs are the mass m, velocity v_i, and position of each particle i, and the x, y subscripts indicate vector components. Equation (3) invokes a binary-interaction approximation in the potential Φ_{ij} between particles i and j, neglecting many-body interactions.

When we ask whether the GK method of obtaining the viscosity is applicable, we are in essence asking whether the fluctuating microscopic shear stresses in Eq. (3) build up through Eq. (2) to drive a steady macroscopic momentum flux that matches that of Eq. (1).

Requirements for an experimental test.—Our test is designed as a comparison of two experimental values, obtained by the GK and hydrodynamic methods. The benchmark in our comparison is the hydrodynamic method because it uses the constitutive relation, which defines the viscosity. We obtain all the data for the two methods from the same experiment, which had separate runs with and without shear flow. The data we use for the GK method, obtained from the runs without shear flow, have not been previously reported. The resulting GK viscosity will be compared to our hydrodynamic result, from the runs with shear flow, which we reported in Ref. [50].

To ensure that we compared results that were truly comparable, we designed the test to satisfy two requirements. The first requirement was identical conditions for the GK and hydrodynamic methods, except for the presence of shear flow. We satisfied this requirement by using data obtained in a single experiment where we alternated runs with and without shear flow. Second, both methods should rely on the same key quantity, P_{xy}. Moreover, P_{xy} should be obtained the same way for both methods, so that any systematic errors will offset in the final comparison. (Systematic errors could arise from measurements of Q or the screening length λ, or from assumptions in the calculation of P_{xy}.) We satisfied this second requirement by using the same expression for P_{xy}, Eq. (3), with the same inputs and the same approximations for the interparticle potential Φ_{ij}, in both methods.

Here, we focus our attention on the transport of the solid-particle component of the dusty plasma, which is governed by the collisional interactions of the particles among themselves. The other components of the dusty plasma (electrons, ions, and neutral gas atoms) each have their own transport relations [50] and do not contribute to viscous transport of momentum within the solid-particle component [51,52].

To the best of our knowledge, all previous dusty plasma experimenters who have reported values for the viscosity used a hydrodynamic method [47,64–72], except for Feng *et al.* [48,51] who used the GK method with an input of experimental data. While Feng *et al.* [51] compared their experimental GK result to an earlier hydrodynamic result [65], that comparison was not definitive because the conditions in the two experiments were different. Moreover, the earlier hydrodynamic results [65] were hindered by two effects, nonuniform temperature and shear thinning, that were not controlled in the experiment. We avoided these two effects in our hydrodynamic measurement [50], and we ensured that conditions were the same for our hydrodynamic and GK methods by using only one experiment.

Experiment.—A radio-frequency plasma was formed above a horizontal electrode, where ≈6000 polymer microspheres were levitated by the electrode sheath. They were levitated in a stable monolayer, which was perpendicular to the downward ion flow. The primary diagnostic was video microscopy [52], with a top-view camera that recorded microsphere motion at 70 frames/s, allowing a direct measurement of microsphere positions and velocities [73,74] with sufficient time resolution to quantify the stress autocorrelation function $C_\eta(t)$. We also used a side-view camera to confirm that out-of-plane motion was too small to affect particle transport [52], allowing us to analyze microsphere motion as if it were 2D.

The collection of microspheres in the monolayer had a stable ground state, which was a triangular crystalline lattice [75–77]. In two "crystal runs," we recorded the motion in this lattice to analyze the phonon spectra [78–80], which reflect the interparticle potential energies with great sensitivity. This analysis yielded the charge $Q = -15700e$ and screening length $\lambda = 0.40$ mm [52], which have uncertainties that are correlated such that they largely cancel one another when we use Q and λ to calculate a potential. The microspheres of diameter 8.7 ± 0.3 μm and mass $m = 5.2 \pm 0.5 \times 10^{-13}$ kg experienced gas drag, with a coefficient [43] of 1.1 s^{-1}, for argon at 6 mtorr. The areal number density of microspheres was $n = 3.5 \times 10^6$ m^{-2}, corresponding to a 2D Wigner-Seitz radius $a = (n\pi)^{-1/2} = 0.30$ mm, an areal mass density $\rho = nm = 1.8 \times 10^{-6}$ kg/m^2, a nominal 2D dusty plasma frequency $\omega_{pd} = (Q^2/2\pi\epsilon_0 m a^3)^{1/2} = 89$ s^{-1}, and a screening parameter $\kappa = a/\lambda = 0.75$. Further details of the experimental setup, as well as discussion of the parameters and their uncertainties, are reported in [50,52].

In all our runs, other than the two crystal runs, we applied laser manipulation with an optical setup that allowed separate application of heating and shear [50,52]. The laser beams were all steady state, with power much too low to affect electrons or ions. For heating manipulation, we used a moving pair of laser beams [81] to augment the kinetic energy of microspheres in the entire monolayer, so that the crystal was melted and behaved like a liquid. We varied the kinetic temperature T by adjusting the laser power, but T was always kept above the melting temperature [82] of the crystalline ground state [52]. For shear manipulation, we used a different pair of laser beams to yield a straight shear flow, which had a transverse gradient in the flow velocity, especially in the gap between the two shear beams [50]. The power of the shear laser beams, which was constant throughout all shear runs [52], was adjusted to be below the threshold for shear thinning and small enough that temperature nonuniformity was negligible [50]. Additionally, we saw no change in particle spacing between adjacent runs with and without shear, so that we are confident Q and λ did not change due to the addition of the shear beams.

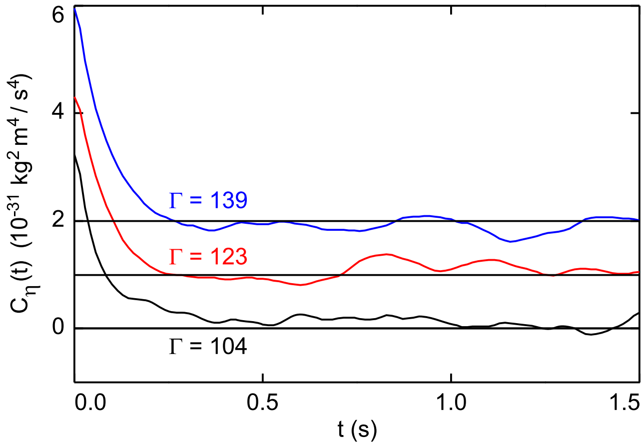

FIG. 1. Stress autocorrelation function $C_\eta(t)$ obtained from the shear-free runs in our experiment. Representative curves are shown, with displaced zeros. The GK viscosity is obtained by integrating these curves, with a choice for the integration limit that takes into account the noise level. The 1.5 s shown here correspond to 105 video frames.

Obtaining the viscosity.—For both the GK and hydrodynamic methods, our analysis starts with the time series [52] of the shear stress P_{xy}, calculated using Eq. (3). This calculation requires inputs of the microsphere positions and velocities as well as the interparticle potentials Φ_{ij}. We obtain Φ_{ij} from the experimentally measured microsphere positions using the Debye-Hückel (DH) model, which expresses the potential between two particles, separated by r_{ij}, as

$$\Phi_{ij}(t) = \frac{Q^2}{4\pi\epsilon_0} \frac{e^{-r_{ij}(t)/\lambda}}{r_{ij}(t)}. \tag{4}$$

The DH potential model in Eq. (4) is suitable for microsphere motion in a 2D plane perpendicular to ion streaming. For such a 2D plane, the DH potential model agrees with binary-collision experiments [83], kinetic simulations [84,85], and comparisons of experimental wave spectra to theory [79,86]. Nevertheless, our analysis is designed to be insensitive to any systematic error in the potential model. In particular, our analysis centers on detecting any difference in the viscosities obtained by the two methods, which both use Eq. (4) the same way. As a validation test [52], we verified that the difference was unaffected when we purposefully introduced errors into the potential.

After using Eqs. (3) and (4) to obtain the time series of P_{xy}, we then calculate its autocorrelation function $C_\eta(t)$, Fig. 1. After an initial decay, $C_\eta(t)$ exhibits noise with a rms level of about 6% of the initial peak.

We take this noise into account when choosing the upper integration limit in Eq. (2) for the GK viscosity. This limit is not chosen as a fixed time, but as the time at which $C_\eta(t)$ decays to 7% of its peak. This choice is a tradeoff between random and systematic errors: a high limit would include

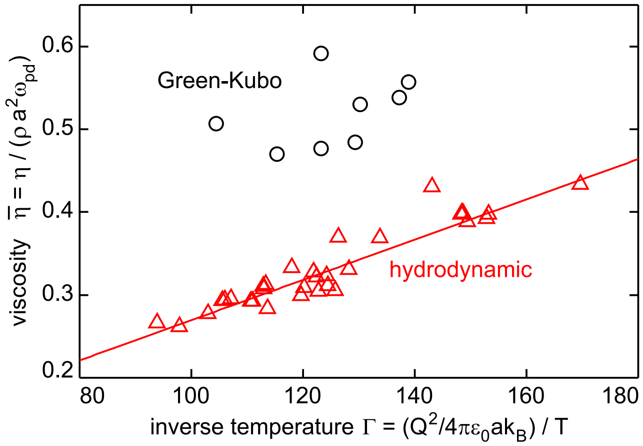

FIG. 2. Comparison of the GK and hydrodynamic results for viscosity. We find that the GK method overestimates the viscosity by about 60%, as compared to measurements by the hydrodynamic method. The GK and hydrodynamic results were obtained from runs without and with shear, respectively, in the same experiment. The overestimation is significantly larger than the scatter. Here, the axes have suppressed zeroes, each data point represents one run, and the line is a linear fit to guide the eye. The hydrodynamic data are from Ref. [50].

more random noise, while a low limit would exclude meaningful data in the initial decay, causing the GK viscosity value to be systematically too low. It will be important that this systematic error is one sided: it can only decrease the GK viscosity. Based on our tests [52], we have estimated this systematic error to range from −1% to −30%, for our choice of the integration limit.

We will report the viscosity in units normalized by $\rho a^2 \omega_{pd}$. Likewise, we will report a normalized inverse temperature as $\Gamma = (Q^2/4\pi\epsilon_0 a k_B)/T$, where the kinetic temperature was obtained from the velocity measurements as $T = m\langle v_x^2 + v_y^2\rangle/2k_B$.

Result of the test.—We find that the GK method overestimates the viscosity. Compared to our benchmark hydrodynamic result, the GK result is about 60% higher, over the entire temperature range of our experiment. This difference is seen clearly in Fig. 2. If systematic errors are taken into account, the difference must actually be larger than 60%, since the systematic errors arising from the integration limit decrease the GK result.

Possible sources for the overestimation.—We now consider candidate explanations for the overestimation of the viscosity by the GK method. Most interestingly, the GK method might simply be inaccurate for liquidlike strongly coupled plasmas, and we identify three reasons this could be so: (1) non-Newtonian behavior, (2) nonequilibrium, and (3) low dimensionality. Additionally, there are several candidates that we can dismiss: the role of the binary-interaction approximation and artifacts of the experimental procedure. We elaborate on all these candidate explanations next, starting with the first three, which we cannot dismiss.

First, non-Newtonian behavior is a property of liquidlike 2D dusty plasmas that may preclude the use of the GK method [49], as we have discussed above. Non-Newtonian behavior mentioned in the literature includes shear thinning [47] and memory effects due to viscoelasticity [46]. We made a particular effort in our experiment to avoid shear thinning, but the memory effects are surely still present, at least at short length and time scales [46,48].

Second, low dimensionality is a characteristic of our experiment since our microspheres were localized in a 2D monolayer. This two-dimensionality could cause the GK method to be inaccurate, according to theoretical arguments that GK methods are generally not applicable in any low-dimensional systems [87–90]. These arguments date back to the 1970s, although the debate remains unsettled [52,91].

Third, nonequilibrium conditions are typical for almost all laboratory plasmas, as they require energy input and have dissipation; this is the case in our experiment as well [51,81]. The GK method was intended for equilibrium [92], so we designed our experiment to mimic a steady equilibrium, with a nearly Maxwellian velocity distribution and a temperature variance close to that of a thermal equilibrium [50,93]. Nevertheless our laboratory plasma may have other nonequilibrium qualities that invalidate the assumptions of the GK method.

The other candidate explanations for the viscosity overestimation, which we can dismiss, are the binary-interaction approximation and three instrumental effects in our experiment. The binary-interaction approximation is made in our calculation of P_{xy}, neglecting three-particle correlations that can exist in a liquidlike 2D dusty plasma [94] as in other liquids. This binary-approximation candidate can be dismissed because any errors introduced would offset in our comparison, since both methods used the same expression for P_{xy}. The three instrumental effects that could affect the GK viscosity are transport associated with neutral gas, anisotropy effects, and erroneous inputs. Feng *et al.* [33] already showed that, for 2D dusty plasmas like ours, viscous transport is unaffected by the rarefied neutral gas. The other two instrumental effects are unlikely to explain the observed overestimation by the GK method, according to tests we performed. Further details of why we dismiss these other instrumental effects are given in the Supplemental Material [52].

Conclusion.—For strongly coupled plasmas, and dusty plasmas in particular, the Green-Kubo method is widely used to obtain the viscosity coefficient from simulations. Despite its wide use, the applicability of this method has until now not been tested experimentally for strongly coupled plasmas. We performed such a test, in a 2D dusty plasma, by comparing to our previously reported value [50] from a hydrodynamic method. Results with and without a flow-velocity gradient yielded the hydrodynamic and GK viscosities, respectively.

Our main result is that the GK method overestimates the viscosity by about 60%, over a wide range of temperature, as shown in Fig. 2. This large overestimation is not attributable to random or systematic errors.

We considered numerous candidate explanations for the overestimation, and the ones we cannot exclude are all consistent with a conclusion that the GK method is not accurate for our strongly coupled dusty plasma. This finding, for dusty plasmas, raises the question of whether the GK method is applicable to other kinds of strongly coupled plasmas.

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