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Fluid–particle dynamics for passive tracers advected by a thermally fluctuating viscoelastic medium

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Many biological fluids, like mucus and cytoplasm, have prominent viscoelastic properties. As a consequence, immersed particles exhibit *subdiffusive* behavior, which is to say, the variance of the particle displacement grows sublinearly with time. In this work, we propose a viscoelastic generalization of the Landau–Lifschitz Navier–Stokes fluid model and investigate the properties of particles that are passively advected by such a medium. We exploit certain exact formulations that arise from the Gaussian nature of the fluid model and introduce analysis of memory in the fluid statistics, marking an important step toward capturing fluctuating hydrodynamics among subdiffusive particles. The proposed method is spectral, meshless and is based on the numerical evaluation of the covariance matrix associated with individual fluid modes. With this method, we probe a central hypothesis of *passive microrheology*, a field premised on the idea that the statistics of particle trajectories can reveal fundamental information about their surrounding fluid environment.

1. Introduction

Many fundamental biological processes concern the movement of microparticles in complex fluids – that is, liquids that contain suspended polymers, microbes, or other microstructures. For example, mucus is a suspension of oligomeric mucin proteins in a water-like fluid and whole blood is a suspension of red blood cells in plasma. Complex fluids are noted for the wide variety of mechanical responses they exhibit as a function of applied strain/stress. In response to certain stimuli, a complex fluid will act like a solid, but in response to other stimuli, the same fluid will act like a liquid. These viscoelastic rheological properties are often caused by interactions between suspended microstructures and a viscous background fluid [1–4].

Particles diffusing in viscous fluids act like classical Brownian motion, but particles moving in viscoelastic fluids tend to be statistically distinct, exhibiting the effects of memory and long-range particle–particle correlation. Indeed the motion of foreign microparticles in complex fluids is very rarely well-described by classical Brownian motion. The primary statistical tool used in particle tracking analysis is mean-squared displacement (MSD), the empirical second moment of change in particle position. For particles moving in a purely viscous fluid, the MSD grows linearly with time; while, in viscoelastic media, the MSD scales nonlinearly. Recent interest in material properties of biological fluids has led to the cataloguing of numerous natural examples of this so-called *anomalous diffusion*. For a few examples, see [5–9].

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The widespread observation of anomalous diffusion in complex fluids has led to the development of several mathematical models that have been variously successful in describing the paths of individual particles. For example, it was recently demonstrated that the motion of 500 nm radius latex beads in human mucus is well-described by fractional Brownian motion (FBM) [10]. Subsequently it was shown that a force-balance approach to modeling, called the generalized Langevin equation (GLE), can provide an even more faithful model for the data [11].

However the extension of single-particle theory to that of multiple interacting particles remains elusive. FBM and GLE are characterized by anti-persistent memory effects (meaning that movement in one direction during one time step is positively correlated with movement in the opposite direction in the next) that result from the storage of energy and response by the surrounding viscoelastic medium. There is no doubt that the medium also communicates disturbances nonlocally across distances as well, and it stands to reason that two proximate particles should have nontrivial interactions with each other. While there exist mathematical frameworks that might have the potential to do so (see complex fluid models like [12] and [13] for example), we know of no work that shows immersed particles behaving as FBM or GLE when alone, but when nearby each other, interacting through fluctuating hydrodynamics. In this article, we take a step in this direction by developing a modeling and simulation framework for particles that are passively advected by a linear viscoelastic fluid.

Because our fluid–particle model will require the interaction of forces, we adopt a GLE approach. The GLE is a stochastic integro-differential equation that was introduced by Kubo [14] and then revived for the purpose of modeling viscoelastic diffusion by Mason and Weitz [15]. Here we adopt the following definition for the GLE, which describes the velocity $V(t)$ of a particle:

$$m\mathrm{d}V(t) = \Big(-\gamma_s V(t) - \frac{\gamma_p}{\tau}\int_{-\infty}^{t} K(t-s)V(s)\mathrm{d}s + \sqrt{\frac{k_B T\gamma_p}{\tau}}F(t)\Big)\mathrm{d}t + \sqrt{2k_B T\gamma_s}\,\mathrm{d}W(t). \tag{1}$$

When referring to the particle position, $X(t) = \int_0^t V(s)\mathrm{d}s$, we say that X satisfies the integrated GLE (iGLE). The GLE (1) is a balance of forces equation relating the particle's acceleration to its velocity history and to thermal fluctuations in the fluid environment. Here, m is the particle's mass, k_B is Boltzmann constant and T is the temperature. The force of friction is decomposed into two components: the drag due to solvent viscosity γ_s and drag due to the polymeric component of the fluid environment, comprised of a memory kernel $K(t)$, a leading drag coefficient γ_p, and a normalizing constant $\tau = \int_0^\infty K(s)\mathrm{d}s$ that has units of [$\mathtt{time}$]. The memory kernel K summarizes the fluid environment's capacity to store energy and act back on a particle after a given increment of time. For discussion on the inclusion of instantaneous viscous response term see [16]. The force due to thermal fluctuations is also decomposed into solvent and polymeric component contributions. The term $W(t)$ is a standard Brownian motion, while $F(t)$ is a stationary Gaussian process with mean 0 and autocovariance given by

$$\mathbb{E}[F(t)F(s)] = K(t-s). \tag{2}$$

This form for the noise is posited in accordance with the *fluctuation–dissipation* relationship [14]. In Section 2, we establish some basic facts about the GLE, including a discussion of our choice for the memory kernel K and our method of simulation.

Initial efforts to model the interaction of multiple particles used a system of GLEs with an interaction term meant to summarize the hydrodynamic influence of one particle on the other [17–20]. However, when implementing the noise terms, these methods tended to ignore correlations that exist with past position and velocity. In particular, there is disagreement about how to enforce the fluctuation–dissipation relationship among multiple beads and through the background fluid without directly modeling the fluid velocity field as well. For this reason it is appealing to note recent developments in numerical fluid–particle coupling methods [12,21–23]. When considering neutrally buoyant particles in an incompressible fluid, the stochastic Immersed Boundary Method has been particularly successful in capturing statistical properties of particle trajectories in viscous fluids [24–28]. In this work, we present an important step toward implementing the stochastic IBM for *viscoelastic* fluids: generalizing the fluctuating Landau–Lifschitz Navier–Stokes equations for viscoelastic fluids and simulating trajectories of passively advected immersed particles.

In Section 3 we develop our model for the fluid velocity field $\mathbf{u}$ and explore the challenges that exist in generating efficient and accurate simulations. The main difficulty arises from two key issues. The first is that, unlike the stochastic IBM for viscous fluids, which can employ step-by-step simulation techniques exploiting the Markov property, the primary tool available for our model is the theory of stationary Gaussian processes. While conceptually straightforward, the implementation presents many practical problems, mostly stemming from our second challenge: that the physical regime of interest corresponds to a situation where the memory kernel has a very slow (power law) decay. As explained in the next section, we use a memory kernel whose form is a sum of exponential functions. The "more viscoelastic" a fluid is, the more terms are necessary in the sum, in turn complicating calculations involving the temporal Fourier transform.

For a neutrally buoyant particle, we consider the equation of motion to be given by averaging over a small region of the fluid velocity field $\mathbf{u}(\mathbf{x},t)$ centered at the particle location:

$$\frac{\mathrm{d}\mathbf{X}(t)}{\mathrm{d}t} = \int \delta_a(\mathbf{x}-\mathbf{X}(t))\mathbf{u}(\mathbf{x},t)\mathrm{d}\mathbf{x}. \tag{3}$$