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On the correspondence between creeping flows of viscous and viscoelastic fluids

Ke Xu [a,*], M. Gregory Forest [a], Isaac Klapper [b]

[a] Departments of Mathematics & Biomedical Engineering & Institute for Advanced Materials, University of North Carolina at Chapel Hill, Chapel Hill, NC 27599-3250, United States

[b] Department of Mathematical Sciences, Montana State University, Bozeman, MT 59717, United States

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Abstract

From the wealth of exact solutions for Stokes flow of simple viscous fluids [C. Pozrikidis, Introduction to Theoretical and Computational Fluid Dynamics, Oxford University Press, Oxford, 1997, pp. 222–311], the classical "viscous–viscoelastic correspondence" between creeping flows of viscous and linear viscoelastic materials yields exact viscoelastic creeping flow solutions. The correspondence is valid for an arbitrary prescribed source: of force, flow, displacement or stress; local or nonlocal; steady or oscillatory. Two special Stokes singularities, extended to viscoelasticity in this way, form the basis of modern microrheology [T.G. Mason, D.A. Weitz, Optical measurements of the linear viscoelastic moduli of complex fluids, Phys. Rev. Lett. 74 (1995) 1250–1253]: the Stokeslet (for a stationary point source of force) and the solution for a driven sphere. We amplify these viscoelastic creeping flow solutions with a detailed focus on experimentally measurable signatures: of elastic and viscous responses to steady and time-periodic driving forces; and of unsteady (inertial) effects. We also assess the point force approximation for micron-size driven beads. Finally, we illustrate the generality in source geometry by analyzing the linear response for a nonlocal, planar source of unsteady stress.

Keywords: Creeping flow; Generalized Stokes–Einstein relation; Microrheology; Viscoelasticity; Correspondence

1. Introduction

Linear response theory, of thermal fluctuations and their associated power spectra and of driven motion from an imposed source, provides a basis for exploring viscous, elastic and compressible properties of condensed matter. For the case of a moving sphere, the viscous–viscoelastic correspondence was developed in 1970 by Zwanzig and Bixon [34], motivated by numerical experiments of Alder and Wainwright [1] on atomic fluctuation spectra. Zwanzig and Bixon developed a quite general theory, allowing for linear viscoelasticity (assuming a single mode Maxwell law), compressibility of the surrounding medium, arbitrary degree of slip of the sphere, and inertial (unsteady) effects. They derived the generalized Stokes–Einstein drag law for viscoelastic fluids, and then the velocity correlation function for thermal fluctuations. We note an even earlier application of linear response theory was carried out by Thomas and Walters [30] in 1965 to model a sedimenting sphere in a viscoelastic fluid. Their focus was on the transient motion and passage to terminal velocity (which they showed depends only on the zero strain rate viscosity of the fluid). Oscillatory forcing of magnetic beads in viscoelastic materials was carried out by Ziemann et al. [33] and then modeled with force balance arguments and spring-dashpot mechanical models to give an alternative method for storage and loss modulus characterization.

Mason and Weitz [19] and Mason et al. [20] had the seminal idea to apply the generalized Stokes–Einstein drag law and associated power spectra of thermally fluctuating beads to rheology. The field of microrheology is now established as a viscoelastic characterization technique, with many variants of the original Mason–Weitz protocol. Gittes et al. [10] used laser-based microscopy techniques to measure trajectories of individual spheres, together with linear response theory. Crocker et al. [8] and Levine and Lubensky [16] developed the elastic–viscoelastic correspondence to relate two-point tracer statistics with viscoelastic (loss and storage) moduli, in some sense a mirror-equivalent approach to the viscous–viscoelastic correspondence emphasized in the present paper. More recently, Liverpool and MacKintosh [17] and Atakhorrami et al. [2] high-

* Corresponding author. Tel.: +1 919 962 5752; fax: +1 919 962 9345.
E-mail address: xuke@email.unc.edu (K. Xu).

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lighted inertial (unsteady but still linear) features of the flow generated by colloidal particles, using the exact solution of linear response theory for a stationary point source of force in a viscoelastic material.

We refer the reader to various review articles in the past few years cf. [11,28,5,18]); the present article has a review component as well. Our paper aims to place these results in a unified context in which results and perspectives from viscous hydrodynamics are transferred to linear viscoelasticity with relative ease, consistent with [34,19], by a straightforward prescription—*the viscous–viscoelastic correspondence*. The analogous elastic–viscoelastic correspondence is addressed in detail by Christensen [6].

The present paper derives from the Virtual Lung Project at UNC and specifically our collaborations with R. Superfine, D. Hill and J. Cribb, in order to model their driven microbead experiments for viscoelastic characterization of lung airway surface liquids. The two special Stokes singularities that have been applied in microrheology thus far, the Stokeslet and the flow generated by a driven sphere, are analyzed here in greater detail than in the microrheology literature. These results are necessary to model and interpret a range of active microrheology experiments, including driven magnetic beads as well as bead tracers for propagating shear waves.

Why revisit these two examples at all? After all, Mason and Weitz already applied the results for a single localized source or a spherical source in their seminal papers [19,20], and Levine and Lubensky [16] and Crocker et al. [8] already analyzed and applied special features of the displacement of one bead due to the thermal motion of another bead, in their analysis of two-point passive microrheology. The two-point focus is in the special regime where the beads are separated by several bead diameters, where bead-fluid interactions are suppressed. In our colleagues' experiments, the beads do not all satisfy this criterion, and it is of interest to know the response function in the immediate neighborhood of a driven bead. The microscope takes data in a focal plane, so it is also relevant to know whether or on what timescale beads will stray out of the focal plane for a given experiment. The standard model for driven magnetic beads [33] relies on a force balance argument with an *ad hoc* geometric factor, and analogies with the Voigt mechanical model are often used to interpret creep-recovery data. More recent models [32] have incorporated polymer network deformations in the immediate neighborhood of the driven bead. It is clearly of interest to derive an explicit expression for a bead driven by a magnetic field in an arbitrary linear viscoelastic material, which yields the bead motion in time as well as the displacement and stress fields in the neighborhood of the bead. This information follows from our analysis of a forced sphere. In shear wave experiments with embedded bead tracers [21], normal stress generation is capable of generating bead motion along the direction of wave propagation. Can one quantify this effect?

For these and related applications, we analyze the viscoelastic creeping flow induced by a time-varying point source and a driven sphere to contrast responses from near-field to far-field, and in different focal planes. Can we distinguish quasi-steady versus unsteady (inertial) effects in the field surrounding a harmonically driven sphere, and if so, where? An investigation into this question is approximated with the viscoelastic analog of a Stokeslet for an imposed time-varying point force by Liverpool and MacKintosh [17] and Atakhorrami et al. [2]. Here, we analyze inertia-induced vortices in both viscous and viscoelastic fluids, and in particular, we show where vortices are spawned in bead diameter dimensions and analyze the vortex strength relative to the applied force, for time-varying point forces and driven spheres. In each illustration, our goal is to inform experimental protocols as to whether and where signatures of elastic versus viscous properties are most accessible.

To formulate the viscous–viscoelastic correspondence, the first step is to cast linear response theory in parallel with the classical hydrodynamic analyses of viscous creeping (Stokes) flow [12,24]. Because of linearity, generalizations to richer sources relevant for modern experiments are immediate, e.g., point or spherical sources with oscillating strength; we provide these results, which are straightforward, but which have not previously appeared in this detail in the literature. Again, our emphasis is on illuminating experimentally measurable features. The more challenging analysis of initial-value problems, as in Thomas and Walters [30], has not been introduced into microrheology thus far, and we do not take up the challenge here.

In the viscoelastic formulation of linear response theory, the viscosity of simple viscous liquids is replaced by the complex viscoelastic modulus of linear viscoelastic materials, after the equations have been transformed from the time domain to frequency space. This identification is possible because linear viscoelasticity presumes a convolution integral for the stress tensor, whose Fourier transform yields the Stokes relation with a complex (frequency-dependent) viscosity. The creeping flow equations (steady or unsteady) can then be posed consistent with point, finite or extended sources, of force, velocity, strain or stress, and the correspondence remains intact for the associated geometry and boundary-initial value problems. Whenever the creeping viscous flow problem can be solved, the analogous solution of the viscoelastic flow problem follows.

Thus far, the field of microrheology has exploited two such creeping viscoelastic flow solutions, for a stationary point source of force and a driven sphere. In fact, only partial features of these solutions are typically used; we illustrate additional information of experimental relevance in the analysis and figures below. These are two from a large family of special solutions arising from "Stokes singularity theory" of viscous fluids [24]. By varying the geometry of the problem and the source (local or nonlocal, steady or unsteady, of force, stress, displacement or velocity), the essential calculation is that of a Green's function, called in this context a viscous Stokes singularity. We present illustrations for point, spherical, and planar sources.

All Stokes singularities (and appropriate sums of them) carry over to viscoelastic media via this simple prescription. The inferences that can be drawn from each creeping flow solution require some analysis and work. Detailed relationships between force, displacement, stress and flow fields are available, which can then be applied to experimental data, or even to design experiments. First, we provide a straightforward exten-