Numerical modeling of bubble dynamics in viscoelastic media with relaxation

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Cavitation occurs in a variety of non-Newtonian fluids and viscoelastic materials. The large-amplitude volumetric oscillations of cavitation bubbles give rise to high temperatures and pressures at collapse, as well as induce large and rapid deformation of the surroundings. In this work, we develop a comprehensive numerical framework for spherical bubble dynamics in isotropic media obeying a wide range of viscoelastic constitutive relationships. Our numerical approach solves the compressible Keller–Miksis equation with full thermal effects (inside and outside the bubble) when coupled to a highly generalized constitutive relationship (which allows Newtonian, Kelvin–Voigt, Zener, linear Maxwell, upper-convected Maxwell, Jeffreys, Oldroyd-B, Giesekus, and Phan-Thien-Tanner models). For the latter two models, partial differential equations (PDEs) must be solved in the surrounding medium; for the remaining models, we show that the PDEs can be reduced to ordinary differential equations. To solve the general constitutive PDEs, we present a Chebyshev spectral collocation method, which is robust even for violent collapse. Combining this numerical approach with theoretical analysis, we simulate bubble dynamics in various viscoelastic media to determine the impact of relaxation time, a constitutive parameter, on the associated physics. Relaxation time is found to increase bubble growth and permit rebounds driven purely by residual stresses in the surroundings. Different regimes of oscillations occur depending on the relaxation time. © 2015 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4922598>]

I. INTRODUCTION

The large-amplitude volumetric oscillations of cavitation bubbles lead to high temperatures and pressures at collapse, as well as induce large and rapid deformation of the surroundings. Considerable research has been dedicated to understanding cavitation dynamics in water, much of it based on the well-known Rayleigh–Plesset equation, a nonlinear second-order ordinary differential equation (ODE) which describes the response of a spherical bubble to a time-varying far-field pressure.¹ Extensions to include compressibility,² thermal effects,³ and non-spherical perturbations are well documented.⁴ Despite its importance to a variety of industrial, botanical, and biological settings, the response of bubbles in non-Newtonian media, particularly viscoelastic media such as polymeric fluids and soft tissue, remains poorly understood. In these materials, complicated rheology can have a significant impact on the cavitation dynamics.

In an industrial setting, cavitation in viscoelastic media is of increasing interest. Ultrasonic cavitation is used to modulate chemical and food processing in cavitation reactors,^{5,6} in which case the surrounding medium is non-Newtonian. Gas bubbles can be used to determine rheological properties of soft materials,⁷ where accurate cavitation models in such media would be valuable for calibration. In botany, water is transported under tension through the xylem of vascular plants, which exhibits viscoelastic properties;⁸ cavitation in trees may lead to embolism⁹ and cause

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branch dieback.¹⁰ In medical applications, ultrasound-induced cavitation can cause hemorrhages and microvasculature damage.¹¹ Shock waves, free radicals, and microjets produced by inertial collapse are potential mechanisms for bioeffects.¹² Techniques relying on high-amplitude and high-frequency ultrasound pulses have been developed to destroy tissue, thermally in high-intensity focused ultrasound¹³ or mechanically in histotripsy.^{14,15} Even in diagnostic ultrasound, the introduction of microbubble contrast agents can cause bleeding.¹⁶ Non-destructive bioeffects of cavitation may be produced with ultrasound, e.g., in sonoporation for gene therapy and drug delivery.¹⁷

Given challenges with experimental studies of cavitation in viscoelastic media, numerical modeling can provide insights into the associated physics and guide experiments. To account for the complex rheology of the aforementioned media, researchers have combined a number of non-Newtonian constitutive relationships, including viscoelastic, with Rayleigh–Plesset-like equations for bubble dynamics. A review of early developments in cavitation in viscoelastic media, with a focus on Maxwell-based models, has been published by Brujan.¹⁸ Motivated by cavitation in polymeric solutions, Fogler and Goddard¹⁹ and Tanasawa and Yang²⁰ laid the groundwork for bubble dynamics in viscoelastic media by coupling the Rayleigh–Plesset equation with linear Maxwell and three-parameter Oldroyd models. Other early studies focused on the three-parameter Oldroyd model,^{21,22} the Jeffreys model,²³ and empirical models.^{24–26} Since Kim’s direct simulations of bubble dynamics in an upper-convected Maxwell (UCM) fluid,²⁷ which highlighted the importance of relaxation effects, a large body of research has focused on Maxwell-based models. Allen and Roy^{28,29} coupled linear and upper-convected Maxwell models to the Rayleigh–Plesset equation and established that the effects of relaxation serve to enhance subharmonic response and increase the maximum bubble radius. Jiménez-Fernández³⁰ generalized this formulation to an Oldroyd-B model. By reducing the partial differential equations (PDEs) necessary to compute the stresses to a set of ODEs, they determined the dependence of inertial cavitation thresholds on the viscoelastic parameters. Naude and Méndez³¹ studied the UCM model for a nearly isothermal bubble to identify a critical Deborah number associated with chaotic response. Kafiabad and Sadeghy³² improved the numerical model of Allen and Roy²⁹ to investigate chaotic behavior in a Giesekus fluid. Direct simulations of non-spherical bubbles in Phan-Thien-Tanner (PTT) liquids³³ revealed that larger Reynolds and Deborah numbers increase a bubble’s tendency to maintain its original shape. Lind and Phillips³⁴ devised a boundary element method applicable for certain Maxwell-type models, and applied it to bubbles near rigid boundaries³⁵ and free surfaces,³⁶ and revisited the rigid boundary scenario using a computationally intensive, but physically less restrictive, spectral element method.³⁷

A variety of other constitutive relationships have recently been considered to model viscoelastic solids such as soft tissue. Yang and Church³⁸ coupled a Kelvin–Voigt model to a Keller–Miksis formulation for bubble dynamics in a compressible medium to investigate inertial cavitation thresholds; they later proposed an empirical modification to match with experimental data.³⁹ The theoretical approach by Yang and Church, combined with rectified diffusion effects, was analyzed by Zhang,⁴⁰ and Patterson *et al.*⁴¹ used their model to relate the inertial cavitation threshold to a bioeffects threshold. Hua and Johnsen⁴² performed a detailed analysis of bubble collapse in a Zener medium and Gaudron *et al.*⁴³ considered the role of nonlinear elasticity for a Kelvin–Voigt model.

Although the aforementioned studies have improved our understanding of cavitation in viscoelastic media, we are still limited by several outstanding issues. First, given the large number of increasingly more complex constitutive relationships, an overarching framework that can easily compare bubble dynamics in media obeying different constitutive models is missing. Second, problems involving violent bubble collapse can pose difficulties when constitutive models require numerical solutions to PDEs. Additionally, analytical relationships between relaxation time and fundamental quantities that describe linearized bubble response have remained largely undiscussed. Finally, an accurate treatment of compressible and thermal effects has yet to be incorporated into viscoelastic cavitation modeling.

The aim of the present study is to address these deficiencies and thereby accurately model spherical bubble dynamics in general isotropic viscoelastic media. The novelty of this contribution is twofold: (i) we develop a comprehensive, yet relatively simple, numerical framework for spherical bubble dynamics in media obeying a wide range of viscoelastic constitutive relationships

and (ii) we simulate bubble dynamics in various viscoelastic media and use theoretical analysis to determine the impact of relaxation time on the associated physics, specifically on growth and oscillations. Our numerical approach is based on the compressible Keller–Miksis equation with full thermal effects (inside and outside the bubble) and a highly generalized constitutive relationship (which allows Newtonian, Kelvin–Voigt, Zener, linear Maxwell, upper-convected Maxwell, Jeffreys, Oldroyd-B, Giesekus, and Phan-Thien-Tanner models). The numerical method can readily be adapted to incorporate more complex viscoelastic models such as the generalized Maxwell and the Burgers models. To solve the general constitutive PDEs, a Chebyshev spectral collocation method was developed, which is robust even for violent collapse; concise ODE formulations are also provided for several special cases. The relative simplicity of this latter formulation allows an exploration of a wide viscoelastic parameter space, with a focus on relaxation effects. Comparisons of various constitutive relationships are made, and the importance of compressible and thermal effects for viscoelastic bubble dynamics is shown. First, the physical model and its numerical solution are described, followed by representative bubble responses to different types of forcing. Theoretical analysis is then presented to explicate the associated physics, and the study ends with concluding remarks.

II. PHYSICAL MODEL

A. Mass conservation and momentum balance

Consider a spherical bubble of radius R in an infinite, isotropic, viscoelastic medium. Let the bubble be filled with non-condensable gas of pressure p . Assuming homobaricity inside the bubble and introducing a compressibility correction in the near-field yield²

$$\left(1 - \frac{\dot{R}}{c_\infty}\right) R \ddot{R} + \frac{3}{2} \left(1 - \frac{\dot{R}}{3c_\infty}\right) \dot{R}^2 = \frac{1}{\rho_\infty} \left(1 + \frac{\dot{R}}{c_\infty} + \frac{R}{c_\infty} \frac{d}{dt}\right) \left(p + J - p_A - \frac{2S}{R}\right), \quad (1)$$

where the surroundings have a constant density ρ_∞ and sound speed c_∞ , S is the constant surface tension between the gas and the surrounding medium, and dots denote time derivatives. The far-field absolute pressure p_A is a prescribed function of time t and is the sum of the constant ambient pressure p_∞ and the acoustic forcing pressure p_f . In Eq. (1), an integral involving the deviatoric stress tensor τ in the surrounding medium, given by Refs. 19 and 44

$$J = 2 \int_R^\infty \frac{\tau_{rr} - \tau_{\theta\theta}}{r} dr, \quad (2)$$

must be evaluated over the radial coordinate r . For a Newtonian fluid with viscosity μ , the deviatoric stresses are $\tau_{rr} = -2\tau_{\theta\theta} = -4\mu\dot{R}^2\dot{R}/r^3$, such that $J = -4\mu\dot{R}/R$. In a general nonlinear viscoelastic medium, the integration may be more complicated. Although spherical symmetry is assumed ($\tau_{\theta\theta} = \tau_{\phi\phi}$), the stress tensor need not be traceless. As in Yang and Church,³⁸ it is assumed that first-order compressibility corrections do not affect the calculation of the stress integral.

B. Constitutive relationships

To evaluate stress integral equation (2), a constitutive model relating the stress tensor τ to the deformation tensor γ is needed. Our interests lie in materials that exhibit the following viscoelastic properties: viscosity, relaxation, elasticity, and retardation. For the large variety of constitutive relationships under consideration, this work investigates those arising from the highly generalized spring-dashpot system in Fig. 1. This system is composed of two Kelvin–Voigt elements (spring and dashpot in parallel) placed in series. When each of the elastic (E_1 and E_2) and viscous (η_1 and η_2) constants is nonzero and finite, this material is henceforth referred to as a Kelvin–Voigt-series (KVS) model. Using dots to denote partial time derivatives, the KVS model can be mathematically expressed as

$$\tau + \lambda_1 \dot{\tau} = 2(G\gamma + \mu\dot{\gamma} + \mu\lambda_2 \ddot{\gamma}), \quad (3)$$

FIG. 1. Spring-dashpot representation of the generalized viscoelastic KVS model.

with relaxation time $\lambda_1 = (\eta_1 + \eta_2)/(E_1 + E_2)$, retardation time $\lambda_2 = \eta_1\eta_2/(E_1\eta_2 + E_2\eta_1)$, elastic modulus $G = E_1E_2/2(E_1 + E_2)$, and viscosity $\mu = (E_1\eta_2 + E_2\eta_1)/2(E_1 + E_2)$. Common viscoelastic models are recovered by setting the appropriate constants to zero. When $\lambda_1, \lambda_2 = 0$ and when G and μ are nonzero, Eq. (3) reduces to the standard Kelvin–Voigt model. The Kelvin–Voigt model further reduces to a Newtonian fluid when $G = 0$ and to a linear elastic solid when $\mu = 0$. For nonzero relaxation times ($\lambda_1 \neq 0$), the KVS model can be reduced to the Jeffreys ($G = 0$ and $\mu, \lambda_2 \neq 0$, with $\lambda_2 < \lambda_1$ to represent a physical fluid²⁸) and Zener⁴⁵ ($\lambda_2 = 0$ with $\mu, G \neq 0$, with $\lambda_1 < \mu/G$ to represent a physical solid⁴²) models. Both the Jeffreys and Zener models are generalizations of the linear Maxwell model, in which $G, \lambda_2 = 0$.

For nonlinear viscoelasticity in an Eulerian framework, we extend Eq. (3) to

$$\boldsymbol{\tau} \exp\left(\frac{\epsilon_2 \lambda_1}{\mu} \text{tr}(\boldsymbol{\tau})\right) + \lambda_1 \left(\overset{\nabla}{\boldsymbol{\tau}} + \epsilon_3 \frac{\boldsymbol{\tau} \cdot \boldsymbol{\tau}}{\mu} \right) = 2 \left(G \boldsymbol{\gamma} + \mu \dot{\boldsymbol{\gamma}} + \mu \lambda_2 \overset{\nabla}{\dot{\boldsymbol{\gamma}}} \right), \quad (4)$$

where the frame-indifferent Oldroyd corotational (upper-convected) derivative, defined as

$$\overset{\nabla}{\boldsymbol{\tau}} = \frac{\partial \boldsymbol{\tau}}{\partial t} + \epsilon_1 [\mathbf{u} \cdot \nabla \boldsymbol{\tau} - (\nabla \mathbf{u})^\top \cdot \boldsymbol{\tau} - \boldsymbol{\tau} \cdot (\nabla \mathbf{u})], \quad (5)$$

measures rates of change with respect to coordinates that translate and deform with the medium.⁴⁶ Here, τ is the transpose, $\text{tr}(\cdot)$ denotes the trace, $\mathbf{u}$ is the velocity field, and $\epsilon_1 = 1$ for all frame-indifferent models. Equation (4) is not meant to represent a specific material but rather reduces to a variety of viscoelastic models summarized in Table I. For the inelastic case ($G = 0$), Eq. (4) models an Oldroyd-B fluid when all of the viscoelastic parameters are nonzero, excluding ϵ_2 and ϵ_3 . When retardation time is neglected, the Oldroyd-B model reduces to the UCM model. The UCM and Oldroyd-B models differ from the linear Maxwell and Jeffreys models, respectively, in the use of frame-indifferent derivatives of the stress tensor $\boldsymbol{\tau}$ and the rate of deformation tensor $\dot{\boldsymbol{\gamma}}$. The UCM model is easily extended to the Giesekus and PTT models, which model complex polymers by introducing nonlinearities in the stress tensor to match experimentally observed behavior.⁴⁷ The Giesekus model corresponds to $\epsilon_2 = 0$ and $0 < \epsilon_3 \leq \frac{1}{2}$, with the quadratic term arising out of

TABLE I. The constitutive models accessible from the general stress-strain relationship Eq. (4) and their respective solution approaches.

Constitutive model	Assumptions	Constraints	Stress solution
Newtonian fluid	$\lambda_1, \lambda_2, G = 0$		Analytic integration (Sec. III C)
Linear elastic solid	$\lambda_1, \lambda_2, \mu = 0$		
Kelvin–Voigt solid	$\lambda_1, \lambda_2 = 0$		
Linear Maxwell fluid	$\epsilon_1, \epsilon_2, \epsilon_3, \lambda_2, G = 0$		Linear PDE to ODE reduction (Sec. III C)
Jeffreys fluid	$\epsilon_1, \epsilon_2, \epsilon_3, G = 0$	$\lambda_1 > \lambda_2$	
Zener solid	$\epsilon_1, \epsilon_2, \epsilon_3, \lambda_2 = 0$	$\lambda_1 < \mu/G$	
Upper-convected Maxwell fluid	$\epsilon_2, \epsilon_3, \lambda_2, G = 0$	$\epsilon_1 = 1$	Convected PDE to ODE reduction (Sec. III B)
Oldroyd-B fluid	$\epsilon_2, \epsilon_3, G = 0$	$\epsilon_1 = 1$	
Giesekus fluid	$\epsilon_2, \lambda_2, G = 0$	$\epsilon_1 = 1, \epsilon_3 \in (0, \frac{1}{2}]$	Spectral collocation of PDEs (Sec. III A)
Phan-Thien-Tanner fluid	$\epsilon_3, \lambda_2, G = 0$	$\epsilon_1, \epsilon_2 = 1$	

molecular theory.⁴⁸ The PTT model corresponds to $\epsilon_2 = 1$ and $\epsilon_3 = 0$, with the exponential factor a result of network theory;⁴⁹ other variations of the PTT model exist.⁵⁰

For the spherically symmetric problem under consideration, general constitutive relationship equation (4) can be written as PDEs for $\tau_{rr}(r, t)$ and $\tau_{\theta\theta}(r, t)$

$$\tau_{rr} \exp\left(\frac{\epsilon_2 \lambda_1}{\mu} (\tau_{rr} + 2\tau_{\theta\theta})\right) + \lambda_1 \left(\frac{\partial \tau_{rr}}{\partial t} + \epsilon_1 \frac{R^2 \dot{R}}{r^2} \frac{\partial \tau_{rr}}{\partial r} + 4\epsilon_1 \frac{R^2 \dot{R}}{r^3} \tau_{rr} + \frac{\epsilon_3}{\mu} \tau_{rr}^2 \right) = -\frac{4\Phi}{r^3} - 4\epsilon_1 \mu \lambda_2 \frac{R^4 \dot{R}^2}{r^6}, \quad (6)$$

$$\tau_{\theta\theta} \exp\left(\frac{\epsilon_2 \lambda_1}{\mu} (\tau_{rr} + 2\tau_{\theta\theta})\right) + \lambda_1 \left(\frac{\partial \tau_{\theta\theta}}{\partial t} + \epsilon_1 \frac{R^2 \dot{R}}{r^2} \frac{\partial \tau_{\theta\theta}}{\partial r} - 2\epsilon_1 \frac{R^2 \dot{R}}{r^3} \tau_{\theta\theta} + \frac{\epsilon_3}{\mu} \tau_{\theta\theta}^2 \right) = \frac{2\Phi}{r^3} - 10\epsilon_1 \mu \lambda_2 \frac{R^4 \dot{R}^2}{r^6}, \quad (7)$$

where

$$\Phi = \frac{G}{3} (R^3 - R_0^3) + \mu R^2 \dot{R} + \mu \lambda_2 (2R \dot{R}^2 + R^2 \ddot{R}). \quad (8)$$

Here, the model of Yang and Church³⁸ is used for the elastic term. This term could also be modeled using linear or nonlinear elasticity, e.g., through neo-Hookean or Mooney-Rivlin strain-energy functions.⁴³

C. Energy balance

To describe the pressure p of the bubble contents and account for heat transfer through the bubble wall, the thermal model of Prosperetti *et al.*^{3,51} is followed, in which the energy equation is solved inside and outside the bubble. Phase changes and mass transfer through the bubble interface lie outside the scope of this work. The key difference with our approach is that we include heat generation via deviatoric (viscoelastic) stresses. The internal bubble pressure is governed by

$$\dot{p} = \frac{3}{R} \left((\kappa - 1) K \frac{\partial T}{\partial r} \Big|_R - \kappa p \dot{R} \right). \quad (9)$$

The gas inside the bubble, with radially varying temperature T and constant ratio of specific heats κ , is assumed to have a thermal conductivity $K = K_A T + K_B$ for empirically determined constants K_A and K_B . The gas temperature is coupled to the temperature of the surrounding medium T_M by the interface conditions

$$T|_{r=R} = T_M|_{r=R} \quad \text{and} \quad K|_{r=R} \frac{\partial T}{\partial r} \Big|_{r=R} = K_M \frac{\partial T_M}{\partial r} \Big|_{r=R}. \quad (10)$$

The thermal conductivity K_M , the thermal diffusivity D_M , and the specific heat capacity C_p of the viscoelastic medium are taken to be constant. Inside the bubble, the energy equation is

$$\frac{\kappa}{\kappa - 1} \frac{p}{T} \left[\frac{\partial T}{\partial t} + \frac{1}{\kappa p} \left((\kappa - 1) K \frac{\partial T}{\partial r} - \frac{r \dot{p}}{3} \right) \frac{\partial T}{\partial r} \right] - \dot{p} = \nabla \cdot (K \nabla T), \quad (11)$$

while outside the bubble, the energy equation is

$$\frac{\partial T_M}{\partial t} + \frac{R^2 \dot{R}}{r^2} \frac{\partial T_M}{\partial r} = D_M \nabla^2 T_M + \frac{\boldsymbol{\tau} : \nabla \mathbf{u}}{\rho_\infty C_p}. \quad (12)$$

The heat generation due to deviatoric stresses in Eq. (12) expands as $\boldsymbol{\tau} : \nabla \mathbf{u} = 2R^2 \dot{R} (\tau_{\theta\theta} - \tau_{rr}) / r^3$. For the remaining boundary conditions, the external temperature decays to a constant temperature T_∞ far from the bubble, and, by symmetry, the internal temperature gradient at the bubble center is zero.

III. DISCRETIZATION OF THE GOVERNING EQUATIONS

The numerical treatment of the constitutive equations varies with the constitutive model itself. These approaches are independently detailed in Secs. III A–III C. For general nonlinear models (in this work, PTT and Giesekus), a spectral collocation method is used to solve the resulting PDEs (Sec. III A); the PDEs for the upper-convected KVS models can be solved via a PDE to

ODE reduction technique (Sec. III B); a similar technique can be used for the linear KVS case (Sec. III C). Meanwhile, the numerical treatment of energy equations (9)–(12) remains the same throughout this study and is described in Appendix A. Except for the inclusion of stress-induced heat generation and a computational improvement, this approach to the energy equations follows that of Kamath and Prosperetti.⁵²

The initial bubble radius R_0 , the density ρ_∞ , the characteristic speed $u_c = \sqrt{p_0/\rho_\infty}$ (where $p_0 = p_\infty + 2S/R_0$ is the equilibrium pressure of the bubble contents), and the ambient temperature T_∞ are used for nondimensionalization. Although the dimensionless-number parameter space is large, the nondimensional parameters of most relevance to this study are the Reynolds $\text{Re} = \rho_\infty u_c R_0/\mu$, Deborah $\text{De} = \lambda_1 u_c/R_0$, and Cauchy $\text{Ca} = p_0/G$ numbers, as well as the nondimensional retardation time $\text{Je} = \rho_\infty R_0^2/\mu\lambda_2$. Other dimensionless parameters appearing in forthcoming formulations are the dimensionless sound speed $C = c_\infty/u_c$ and the Weber number $\text{We} = p_0 R_0/2S$. Except when otherwise noted, variables will henceforth be written in their dimensionless form.

A. Spectral collocation method for general nonlinear viscoelastic fluids

The constitutive models for the Giesekus and PTT fluids contain a nonlinearity in τ that obstructs the construction of a simple ODE formulation, which is possible for other models (as in Secs. III B and III C). For the Giesekus fluid, a coordinate transformation as in Sec. III B reduces the constitutive relationship to a Riccati equation, but this is of little help for salvaging an ODE for J . Likewise, for the PTT fluid, the cross-coupled nonlinearity through the trace operator is prohibitively complex. Past authors have therefore resorted to numerical methods capable of solving the full set of equations but have often encountered difficulty resolving the stress fields during strong bubble collapse. Herein, we present a robust spectral approach and note that this method is effective for *any* constitutive relationship of this form. We present the fundamentals of spectral collocation necessary to treat the present problem and refer the reader to Boyd⁵³ for a rigorous discussion.

In dimensionless variables, the PDEs arising from the constitutive relationships for the Giesekus and PTT fluids, from Eqs. (6) and (7), can be written as the single equation

$$\tau_{qq} e^{\epsilon_2 \text{De}(\tau_{rr} + 2\tau_{\theta\theta})} + \text{De} \left(\frac{\partial \tau_{qq}}{\partial t} + \frac{R^2 \dot{R}}{r^2} \frac{\partial \tau_{qq}}{\partial r} + c_{qq} \frac{R^2 \dot{R}}{r^3} \tau_{qq} + \epsilon_3 \text{Re} \tau_{qq}^2 \right) = -\frac{c_{qq}}{\text{Re}} \frac{R^2 \dot{R}}{r^3}. \quad (13)$$

The subscript qq is not indicative of summation, but rather is a placeholder for both the normal radial and polar components, for which $c_{rr} = 4$ and $c_{\theta\theta} = -2$. We begin by projecting the semi-infinite, mobile domain $r \in [R(t), \infty)$ onto $\zeta \in [-1, 1)$ by means of the coordinate transformation

$$\zeta = 1 - \frac{2}{1 + (r/R - 1)/L_v}, \quad (14)$$

where L_v is a characteristic viscous length, in analogy to the approach of Kamath *et al.*⁵¹ for temperature fields in the surroundings. In the new coordinates, Eq. (13) yields

$$\frac{\partial \tau_{qq}}{\partial t} = - \left(\frac{e^{\epsilon_2 \text{Re} \text{De}(\tau_{rr} + 2\tau_{\theta\theta})}}{\text{De}} + c_{qq} \frac{\dot{R}}{y^3 R} + \epsilon_3 \text{Re} \tau_{qq} \right) \tau_{qq} + \frac{(1 - \zeta)^2 \dot{R}}{2L_v R} \frac{y^3 - 1}{y^2} \frac{\partial \tau_{qq}}{\partial \zeta} - \frac{c_{qq}}{\text{Re} \text{De}} \frac{\dot{R}}{y^3 R}, \quad (15)$$

where $y = r/R$ is a function of ζ by inverting Eq. (14).

The PDEs arising from the constitutive relationships can be written in the form $\partial f/\partial t = \mathcal{D}[f]$ where $\mathcal{D}$ is, in general, a nonlinear differential operator over some spatial coordinate x . The unknown function $f = f(x, t)$ is approximated by an expansion in terms of N basis functions $\phi_n(x)$ and N time-dependent spectral coefficients $k_n(t)$: $f(x, t) \approx f^N(x, t) = \sum_{n=1}^N k_n(t) \phi_n(x)$. After making this truncated spectral approximation, the general PDE becomes

$$\sum_{n=1}^N \frac{dk_n}{dt} \phi_n = \mathcal{D} \left[\sum_{n=1}^N k_n \phi_n \right] \quad (16)$$

and can be evaluated at N collocation points in the spatial domain, forming a system of N ordinary differential equations in time. For the nonperiodic problems herein, the basis functions are written in terms of the Chebyshev polynomials, whose n th function is given by $T_n(x) = \cos n\theta$, with $x = \cos \theta \in [-1, 1]$. By expanding the stresses in terms of the shifted Chebyshev polynomials $\phi_n(\zeta) = T_n(\zeta) - 1$, the boundary conditions at infinity are automatically satisfied since $T_n(1) = 1$ for all n . Therefore, we use the spectral truncation approximations

$$\tau_{rr}(\zeta, t) \approx \tau_{rr}^N(\zeta, t) = \sum_{n=1}^N c_n(t) (T_n(\zeta) - 1), \quad \tau_{\theta\theta}(\zeta, t) \approx \tau_{\theta\theta}^N(\zeta, t) = \sum_{n=1}^N d_n(t) (T_n(\zeta) - 1), \quad (17)$$

which are substituted into (15) and then evaluated at the Chebyshev-Gauss-Lobatto collocation points $\zeta_j = \cos \frac{\pi j}{N}$, with $j = 1, 2, \dots, N$. This forms a system of N equations for N unknown time-derivatives of the spectral coefficients, $\dot{c}_n$ and $\dot{d}_n$, which can be solved for and then advanced in time.

A benefit of the spectral method is that, after spectral approximations (17) are made, integrals over the stress field can be computed analytically. In particular, J is given by

$$J^N \equiv 2 \int_R^\infty \frac{\tau_{rr}^N - \tau_{\theta\theta}^N}{r} dr = 2 \sum_{n=1}^N e_n (c_n - d_n),$$

where

$$e_n = 2 \int_{-1}^1 \frac{T_n(\zeta) - 1}{(1 - \zeta)[2 + (1 - \zeta)(1/L_v - 1)]} d\zeta. \quad (18)$$

This result is a significant advantage over a finite-element discretization of the stress fields, in which a quadrature method is required to compute J . The time derivative of J is likewise simply given by $\dot{J}^N = 2 \sum_{n=1}^N e_n (\dot{c}_n - \dot{d}_n)$. The coefficients e_n can be precomputed to high precision.

In terms of dimensionless variables and spectral coefficients, Keller–Miksis equation (1) takes the form

$$\dot{R} = U, \quad (19)$$

$$\begin{aligned} \dot{U} = & \left[-\frac{3}{2} \left(1 - \frac{U}{3C} \right) U^2 + \left(1 + \frac{U}{C} \right) \left(p - \frac{1}{\text{We}R} + 2 \sum_{n=1}^N e_n (c_n - d_n) - 1 + \frac{1}{\text{We}} - p_f \right) \right. \\ & \left. + \frac{R}{C} \left(\dot{p} + \frac{U}{\text{We}R^2} + 2 \sum_{n=1}^N e_n (\dot{c}_n - \dot{d}_n) - \dot{p}_f \right) \right] \left/ \left(R - \frac{RU}{C} \right) \right. . \end{aligned} \quad (20)$$

Convergence rates are demonstrated in Fig. 2 for the growth and collapse of a bubble subjected to a Gaussian waveform in an UCM medium ($\epsilon_2, \epsilon_3 = 0$). The first two minima and maxima of the $R(t)$ curve are compared against the ODE solution in Sec. III B, which is exact (that is, not affected by spatial discretization errors). Exponential convergence rates appear even for the maxima and minima following a strong collapse. While comparison to an exact solution is not possible for the Giesekus and PTT models, the method remains convergent despite the terms added by nonzero ϵ_2 or ϵ_3 , as expected. We note that the incompressible near field cannot support shock waves produced at collapse.

B. Upper-convected models

For the upper-convected viscoelastic models, the method described in Sec. III A can be used directly. However, there exists a simpler and less computationally expensive approach by exactly reducing the constitutive PDEs to ODEs. For Oldroyd-B (and hence UCM) materials, the stress fields for the radial and polar components are described by

$$\tau_{qq} + \text{De} \left(\frac{\partial \tau_{qq}}{\partial t} + \frac{R^2 \dot{R}}{r^2} \frac{\partial \tau_{qq}}{\partial r} + c_{qq} \frac{R^2 \dot{R}}{r^3} \tau_{qq} \right) = -c_{qq} \frac{\Phi(t)}{r^3} + k_{qq} \frac{R^4 \dot{R}}{r^6}, \quad (21)$$

FIG. 2. Convergence tests using the spectral method for the UCM model. The bubble is forced using the Gaussian pulse in Eq. (44) with $P_A = 6p_\infty$. The ODE solution of Sec. III B for the UCM model was used as the reference, and the isothermal assumption was made for simplicity.

where $k_{qq} = c_{qq}(3 - c_{qq})/\text{Je}$, and, as before, $c_{rr} = 4$ and $c_{\theta\theta} = -2$. Due to the upper-convected derivatives, Eq. (21) does not imply $\tau_{\theta\theta} = -2\tau_{rr}$.

After the coordinate transformation $z = (r^3 - R^3)/3$, the spatial derivative of τ_{qq} can be absorbed, giving

$$\tau_{qq} + \text{De} \left(\frac{\partial \tau_{qq}}{\partial t} + c_{qq} \frac{R^2 \dot{R}}{3z + R^3} \tau_{qq} \right) = -c_{qq} \frac{\Phi(t)}{3z + R^3} + k_{qq} \frac{R^4 \dot{R}^2}{(3z + R^3)^2}. \quad (22)$$

Using an integrating factor, we obtain

$$\tau_{qq} = \frac{1}{\text{De}} \int_0^t e^{(\xi-t)/\text{De}} \left(-c_{qq} \frac{(3z + R^3(\xi))^{c_{qq}/3-1}}{(3z + R^3(t))^{c_{qq}/3}} \Phi(\xi) + k_{qq} \frac{(3z + R^3(\xi))^{c_{qq}/3-2}}{(3z + R^3(t))^{c_{qq}/3}} R^4(\xi) \dot{R}^2(\xi) \right) d\xi, \quad (23)$$

which allows the calculation of $J_{qq} \equiv 2 \int_R^\infty \frac{\tau_{qq}}{r} dr$,

$$J_{qq} = \frac{2}{\text{De}} \int_0^t e^{(\xi-t)/\text{De}} \int_0^\infty \left(-c_{qq} \frac{(3z + R^3(\xi))^{c_{qq}/3-1}}{(3z + R^3(t))^{c_{qq}/3+1}} \Phi(\xi) + k_{qq} \frac{(3z + R^3(\xi))^{c_{qq}/3-2}}{(3z + R^3(t))^{c_{qq}/3+1}} R^4(\xi) \dot{R}^2(\xi) \right) dz d\xi. \quad (24)$$

Noting that

$$\int_0^\infty \frac{(3z + R^3(\xi))^{c_{qq}/3-1}}{(3z + R^3(t))^{c_{qq}/3+1}} dz = \frac{R^{c_{qq}}(t) - R^{c_{qq}}(\xi)}{c_{qq} R^{c_{qq}}(t) [R^3(t) - R^3(\xi)]}, \quad (25)$$

$$\int_0^\infty \frac{(3z + R^3(\xi))^{c_{qq}/3-2}}{(3z + R^3(t))^{c_{qq}/3+1}} dz = \frac{c_{qq} R^3(t) R^{c_{qq}}(\xi) - 3 R^{c_{qq}}(t) R^3(\xi) + (3 - c_{qq}) R^{3+c_{qq}}(\xi)}{c_{qq} (3 - c_{qq}) R^{c_{qq}}(t) R^3(\xi) [R^3(t) - R^3(\xi)]^2}, \quad (26)$$

it can be shown after extensive manipulation that

$$J = J_{rr} - J_{\theta\theta} = \frac{-2}{\text{De} R^4(t)} \int_0^t e^{(\xi-t)/\text{De}} \left(\frac{R^3(\xi) + R^3(t)}{R^2(\xi)} \Phi(\xi) + \frac{R^3(\xi) - 2R^3(t)}{R(\xi)} \frac{\dot{R}^2(\xi)}{\text{Je}} \right) d\xi. \quad (27)$$

Rather than compute J by a quadrature rule at each time step, it is advantageous to express J in terms of ODEs that can be marched in time along with the differential equations for radial evolution.³¹ Defining

$$J_1 = \frac{-2e^{-t/\text{De}}}{\text{De}R(t)} \int_0^t \frac{e^{\xi/\text{De}}}{R(\xi)} \left(\frac{\Phi(\xi)}{R(\xi)} - \frac{2\dot{R}^2(\xi)}{\text{Je}} \right) d\xi, \quad (28)$$

$$J_2 = \frac{-2e^{-t/\text{De}}}{\text{De}R^4(t)} \int_0^t e^{\xi/\text{De}} \left(R(\xi)\Phi(\xi) + \frac{R^2(\xi)\dot{R}^2(\xi)}{\text{Je}} \right) d\xi, \quad (29)$$

such that $J_1 + J_2 = J$, differentiating each with respect to time gives

$$\dot{J}_1 = -\left(\frac{1}{\text{De}} + \frac{\dot{R}}{R} \right) J_1 - \frac{2}{\text{DeRe}} \frac{\dot{R}}{R} - \frac{2}{\text{DeJe}} \frac{\ddot{R}}{R}, \quad (30)$$

$$\dot{J}_2 = -\left(\frac{1}{\text{De}} + \frac{4\dot{R}}{R} \right) J_2 - \frac{2}{\text{DeRe}} \frac{\dot{R}}{R} - \frac{2}{\text{DeJe}} \left(\frac{3\dot{R}^2}{R^2} + \frac{\ddot{R}}{R} \right). \quad (31)$$

The $\ddot{R}$ term renders Eqs. (30) and (31) unwieldy since the ODE for radial dynamics is also second order in R . Past authors²⁰ have addressed a similar situation by differentiating the Rayleigh–Plesset equation and forming a third-order ODE. Differentiation of Keller–Miksis equation (1), however, is a prohibitively inconvenient route, especially since it introduces the term $\ddot{J}$. Such difficulties can be avoided by the transformations

$$J_1 = \frac{Z_1}{R^3} - \frac{2}{\text{DeJe}} \frac{\dot{R}}{R}, \quad J_2 = \frac{Z_2}{R^3} - \frac{2}{\text{DeJe}} \frac{\dot{R}}{R}, \quad (32)$$

where $Z_1(t)$ and $Z_2(t)$ are auxiliary variables. This allows the integro-differential equation for radial dynamics, Eq. (1), and the PDEs for Oldroyd-B viscoelasticity, Eq. (21), to be written as a system of four nonlinear ODEs,

$$\dot{R} = U, \quad (33)$$

$$\dot{Z}_1 = -\left(\frac{1}{\text{De}} - \frac{2U}{R} \right) Z_1 + \frac{2}{\text{De}} \left(\frac{1}{\text{DeJe}} - \frac{1}{\text{Re}} \right) R^2 U, \quad (34)$$

$$\dot{Z}_2 = -\left(\frac{1}{\text{De}} + \frac{U}{R} \right) Z_2 + \frac{2}{\text{De}} \left(\frac{1}{\text{DeJe}} - \frac{1}{\text{Re}} \right) R^2 U, \quad (35)$$

$$\begin{aligned} \dot{U} = & \left[-\frac{3}{2} \left(1 - \frac{U}{3C} \right) U^2 + \left(1 + \frac{U}{C} \right) \left(p - \frac{1}{\text{We}R} + \frac{Z_1 + Z_2}{R^3} - \frac{4}{\text{DeJe}} \frac{U}{R} - 1 + \frac{1}{\text{We}} - p_f \right) \right. \\ & \left. + \frac{R}{C} \left(\dot{p} + \frac{U}{\text{We}R^2} + \frac{\dot{Z}_1 + \dot{Z}_2}{R^3} - \frac{3U}{R^4} (Z_1 + Z_2) + \frac{4}{\text{DeJe}} \frac{U^2}{R^2} - \dot{p}_f \right) \right] \left/ \left(R - \frac{RU}{C} + \frac{4}{\text{DeJe}C} \right) \right. \end{aligned} \quad (36)$$

For $\text{Je} \rightarrow \infty$, these equations treat bubble dynamics in an UCM fluid.

C. Linear Kelvin–Voigt-series models

In the case of linear KVS media (that is, for linear Maxwell, Zener, and Jeffreys materials), the radial stresses are given by

$$\tau_{rr} + \text{De} \frac{\partial \tau_{rr}}{\partial t} = -\frac{4}{r^3} \left(\frac{R^3 - 1}{3\text{Ca}} + \frac{R^2 \dot{R}}{\text{Re}} + \frac{2R\dot{R}^2 + R^2 \ddot{R}}{\text{Je}} \right) \quad (37)$$

and $\tau_{\theta\theta} = -2\tau_{rr}$. This PDE can be reduced to an ODE by the same method as in Sec. III B. The stress integral becomes

$$J = 3 \int_R^\infty \frac{\tau_{rr}}{r} dr = -\frac{4}{\text{De}R^3} \int_0^t e^{(\xi-t)/\text{De}} \Phi(\xi) d\xi, \quad (38)$$

and after defining $Z = R^3 J + 4R^2 \dot{R}/\text{DeJe}$, the following ODE system results:

$$\dot{R} = U, \quad (39)$$

$$\dot{Z} = -\frac{Z}{\text{De}} + \frac{4}{\text{De}} \left(\frac{1}{\text{DeJe}} - \frac{1}{\text{Re}} \right) R^2 U - \frac{4}{3\text{CaDe}} (R^3 - 1), \quad (40)$$

$$\begin{aligned} \dot{U} = & \left[-\frac{3}{2} \left(1 - \frac{U}{3C} \right) U^2 + \left(1 + \frac{U}{C} \right) \left(p - \frac{1}{\text{We}R} + \frac{Z}{R^3} - \frac{4}{\text{DeJe}} \frac{U}{R} - 1 + \frac{1}{\text{We}} - p_f \right) \right. \\ & \left. + \frac{R}{C} \left(\dot{p} + \frac{U}{\text{We}R^2} + \frac{\dot{Z}}{R^3} - \frac{3U}{R^4} Z + \frac{4}{\text{DeJe}} \frac{U^2}{R^2} - \dot{p}_f \right) \right] \left/ \left(R - \frac{RU}{C} + \frac{4}{\text{DeJe}C} \right) \right. \end{aligned} \quad (41)$$

For zero relaxation (i.e., Kelvin–Voigt solids), Eq. (40) is replaced by

$$Z = -\frac{4}{\text{Re}} R^2 \dot{R} - \frac{4}{3\text{Ca}} (R^3 - 1). \quad (42)$$

Other elastic terms⁴³ can be substituted into this expression.

Past researchers^{28,42} have used ODEs different from those above to treat the stresses in linear Maxwell/Zener/Jeffreys models. These ODEs were derived by evaluating Eq. (37) at the bubble wall,²⁸

$$\tau_{rr}|_R + \lambda_1 \frac{\partial \tau_{rr}}{\partial t} \Big|_R = -\frac{4\Phi|_R}{R^3}, \quad (43)$$

and implicitly assuming that $\frac{\partial \tau_{rr}}{\partial t} \Big|_R$ is equivalent to $\frac{\partial \tau_{rr}|_R}{\partial t}$. Since $\frac{\partial \tau_{rr}}{\partial t} \Big|_R \neq \frac{\partial \tau_{rr}|_R}{\partial t}$ in general, this leads to an erroneous ODE system. This past ODE system, while not seriously misleading for small perturbations,⁴² tends to make unphysical large-amplitude predictions (e.g., unexpected explosive growth²⁸), and has led to the conclusion that the linear Maxwell model is inferior to the UCM model.²⁹ In our observations from the correct set of ODEs, Eqs. (39)–(41), obviously unphysical predictions are no longer present, and there is much more agreement between the linear models and their nonlinear counterparts in Eqs. (33)–(36).

IV. NUMERICAL RESULTS

We numerically investigate the response of bubbles in viscoelastic media when subjected to various canonical pressure forcing functions relevant to a wide range of applications: Gaussian pulse, Rayleigh growth, Rayleigh collapse, and harmonic forcing. Where applicable, except when otherwise stated, the following viscoelastic properties will be used for the surrounding medium, chosen to broadly estimate materials that might appear in biomedicine and industry: $\mu = 35$ cP, $\lambda_1 = 1.0$ μ s, $\lambda_2 = 0.2$ μ s, $G = 10$ kPa, and $\epsilon_3 = 0.5$. The surrounding medium is initially at standard ambient temperature $T_\infty = 293$ K and pressure $p_\infty = 101$ kPa, with density $\rho_\infty = 1060$ kg/m³ and sound speed $c_\infty = 1430$ m/s. The thermal properties of the surroundings are taken to be those of water ($K_M = 0.55$ W/m K, $D_M = 1.41(10^{-7})$ m²/s, and $C_p = 4.18$ kJ/kg K), which correspond closely to many bodily tissues. For the problem under consideration, temperature variations in the surroundings, and their effect on the bubble dynamics, are small.³ The gas inside the bubble is air with $\kappa = 1.40$, $K_A = 5.28(10^{-5})$ W/m K², and $K_B = 1.17(10^{-2})$ W/m K, and the surface tension is that between air and blood: $S = 0.056$ N/m. Blood is a biological medium possessing a well-established surface tension in air; the surface tension of many engineering substances is of this order. Unless otherwise stated, the initial radius is set at $R_0 = 3.0$ μ m, the initial wall velocity is zero, the surrounding medium is initially unstressed, and the bubble is initially in pressure equilibrium.

A Dormand-Prince fourth-order Runge-Kutta method⁵⁴ with adaptive stepsize control is used for time-marching. Although the spectral collocation method can be used to treat any of the constitutive relationships where a single time-derivative of τ appears, the ODE solutions provide a simpler alternative for constitutive relationships linear in τ . When the spectral collocation method is used, excellent resolution can be expected when $|c_N|, |d_N| < 10^{-4}$. This constraint dictates a large number of collocation points N when studying violent collapse problems, in which sharply peaked stresses appear at the bubble wall. A fast Fourier transform (FFT)-based algorithm for fast

computation of spectral operations is given in Appendix B and succeeds in speeding up computations for $N \gtrsim 650$. However, with the exception of Fig. 2, the relatively mild cases investigated herein achieve excellent resolution with $N = 50$. In this work, $L_v = 3$ was used throughout, but choices of L_v from $O(10^{-2})$ to $O(10^2)$ were not found to significantly influence convergence efficiency. Adequate resolution of the temperature fields was usually achieved with 10–20 basis functions, in agreement with the findings of past authors.⁵⁵

A. Gaussian pulse

In hydrodynamic cavitation applications, the local pressure may momentarily decrease in a smooth manner. To model this phenomenon, we use a Gaussian drop in pressure, given in dimensional units as

$$p_f(t) = P_A \exp \left[-\left(\frac{t - \delta t}{t_w} \right)^2 \right], \quad (44)$$

with amplitude $P_A = 2p_\infty$ and timing parameters $\delta t = 1.74 \mu\text{s}$ and $t_w = 0.498 \mu\text{s}$. The nondimensional characteristic width of the Gaussian pulse is $O(1)$. In Fig. 3(a), the dependence of the bubble response on different viscoelastic models is shown. For the given forcing and viscosity, the oscillations in a Newtonian fluid are overdamped. For the Kelvin–Voigt model, the growth is reduced due to elasticity, in accordance with past experimental and numerical observations.⁵⁶ However, for models in which relaxation is present, the growth is enhanced. For the present value of relaxation time ($\lambda_1 = 1.0 \mu\text{s}$), elasticity plays only a small role in the Zener model, but the oscillation frequency is increased over the Maxwell model, as expected.⁴² The largest radius and least amount of damping are produced in the Maxwell material. Retardation in the Jeffreys model reduces growth and enhances damping, as observed in the past work.²⁸

For nonlinear viscoelasticity, Fig. 3(b), similar observations can be made. Each medium allows a more dynamic response than the Newtonian fluid. Differences between the UCM and Oldroyd-B models are similar to differences between linear Maxwell and Jeffreys models. The models with

FIG. 3. Bubble response to the Gaussian pulse (44) in different (a) linear and (b) nonlinear viscoelastic materials. In (c), an UCM fluid is used for three different relaxation times. Relaxation time is varied in (d), where the maximum radius and rebound (second maximum) radius are measured for UCM and Oldroyd-B fluids.

FIG. 4. Maximum shear stress fields for the (a) Giesekus and (b) PTT responses to a Gaussian pulse shown in Fig. 3. The color axis measures the stress in Pascals with logarithmic scaling.

relaxation but no retardation exhibit similar growth, but differences at collapse: the PTT model produces the smallest minimum radius, followed by Giesekus and UCM, respectively. Conversely, the largest rebound radius (a measure of least damping during collapse) is observed with the UCM model, followed by Giesekus and PTT. This behavior may result from the larger amount of energy dissipated acoustically upon collapse for smaller radii;⁵⁷ Fig. 18 in Appendix A, which shows the bubble pressure and center temperature for the case study in Figs. 3(a) and 3(b), demonstrates that trends in bubble pressure are consistent with this acoustic dissipation theory.

As seen in Figs. 3(c) and 3(d), as the relaxation time is increased, the maximum radius increases as well. Relaxation strongly affects the character of the oscillations; this feature is clear when considering the dependence of growth on the relaxation time. As shown analytically in Sec. V B 1, higher relaxation time increases the effective pressure drop experienced by the bubble, and for a longer duration.

The numerical model is well-suited for determining the surrounding stress fields at high resolution, even in cases of violent collapse. Since τ_{rr} and $\tau_{\theta\theta}$ are principal stresses, the maximum shear stress is given by $|\tau_{r\theta}| = |\tau_{rr} - \tau_{\theta\theta}|/2$. In Fig. 4, $|\tau_{r\theta}|$ is graphed in the field surrounding bubbles in PTT and Giesekus media. Although the $R(t)$ curves are similar, the stress fields exhibit clear differences. The stresses at collapse are notably higher in the Giesekus medium. The position of zero shear stress, achieved where $\tau_{rr} = \tau_{\theta\theta}$, shows significant differences between the PTT and Giesekus models. Regions of low stresses illustrate clear memory effects due to relaxation.

B. Rayleigh growth

The results in Sec. IV A on Gaussian forcing provide useful qualitative information for a practically relevant problem. However, the results depend on the pulse width. To eliminate this parameter, we consider “Rayleigh growth” (by analogy to the well-known Rayleigh collapse problem), in which the bubble is subjected to a step change in pressure from p_∞ at $t = 0^-$ to $p_\infty - \Delta P$ at $t = 0^+$. The bubble grows explosively and tends toward an equilibrium radius larger than its initial radius. For Rayleigh growth, we use $\Delta P = 0.9p_\infty$. Fig. 5 shows the bubble response for different viscoelastic media and properties. Once again, conditions are such that the oscillations are overdamped in the Newtonian case. The final dimensionless equilibrium radius $\mathcal{R}$ is given implicitly from the following equation:⁴²

$$0 = \frac{1}{\mathcal{R}^{3\kappa}} - 1 + \frac{1 - 1/\mathcal{R}}{\text{We}} - \frac{4}{3\text{Ca}} \left(1 - \frac{1}{\mathcal{R}^3} \right) - \frac{\Delta P}{p_0}. \quad (45)$$

$\mathcal{R}$ is closer to unity in the Kelvin-Voigt and Zener cases due to the elastic restoring force. Although it affects the final state, elasticity does not greatly modify the initial growth ($t \lesssim 0.5 \mu\text{s}$), whereas at early times, relaxation substantially increases growth rate. Growth is enhanced and oscillations are less damped as the relaxation time is increased, but the initial growth rate remains unchanged.

FIG. 5. Bubble response to Rayleigh growth in different (a) linear and (b) nonlinear viscoelastic materials. In (c), an UCM fluid is used for three different relaxation times. Relaxation time is varied in (d), where the overshoot radius R_{over} is measured for UCM and Oldroyd-B fluids, and their linear counterparts.

For large relaxation times, significant overshoots (the difference between the first maximum radius and the equilibrium radius) can be produced as the relaxation time is increased, while retardation reduces this overshoot. As in Sec. IV A, oscillatory solutions are observed in media with relaxation, retardation damps the oscillations, and the behavior in the nonlinear viscoelastic media does not significantly deviate from their linear counterparts. The differences between the Giesekus and UCM models are less pronounced in this problem. As in Fig. 3, the PTT model achieves the largest growth. Fig. 5 also shows that, for certain relaxation times, the maximum radius after rebound can be less than that for the Newtonian case. This result does not contradict our forthcoming conclusion that relaxation causes an increased *initial* growth, although these numerical results indicate that, for large enough relaxation times, this faster initial growth dominates other effects and indeed causes a positive overshoot.

C. Rayleigh collapse

Given that the solutions deviate strongly following collapse in Fig. 3, we also investigate the Rayleigh collapse problem. Here, a bubble of radius $R_0 = 15 \mu\text{m}$ is subjected to a step increase in pressure of $35p_\infty$. The bubble, now out of equilibrium, collapses and rebounds before settling to a smaller equilibrium radius, given by Eq. (45) with $\Delta P = -35p_\infty$. Fig. 6 shows the response of a bubble to Rayleigh collapse for different viscoelastic media and properties. In this problem, the bubble response is essentially the same until the first collapse. The collapse time and minimum radius for the Newtonian and Kelvin-Voigt models are slightly longer and larger, respectively, than for the models with relaxation. Although the models with relaxation each predict a similar minimum radius, the stress distributions stored up to the instant of first collapse can vary widely and thereby strongly affect the subsequent oscillations.

Perhaps, the most interesting observation is that the models that include relaxation produce smaller minimum radii than models without relaxation, yet their rebound radii are larger. This phenomenon is counter-intuitive in that more energy is expected to be dissipated via viscous and compressible damping when smaller radii are achieved at collapse.⁵⁷ Furthermore, the rebound radius does not monotonically increase with increasing relaxation time: there is an intermediate

FIG. 6. Bubble response to Rayleigh collapse in different (a) linear and (b) nonlinear viscoelastic materials. In (c), an UCM fluid is used for three different relaxation times. Relaxation time is varied in (d), where the rebound radius R_{reb} is measured for UCM and Oldroyd-B fluids, and their linear counterparts. The minimum radii of the collapsing bubbles are less than the equilibrium radius, which is approximately $0.43R_0$ (or $0.44R_0$ for the Kelvin–Voigt and Zener models).

value of relaxation time for which the rebound is maximum, and this value does not correspond to the largest minimum radius. The presence of relaxation time in the constitutive model is certainly responsible for this behavior, similarly to the way growth was affected in Sec. IV B. However, between the Rayleigh growth and collapse problems, the driving mechanism for bubble growth is different. For Rayleigh growth, a low external pressure is solely responsible. For Rayleigh collapse, the rebound is motivated by the bubble wall pressure differential as well as deviatoric tensile stresses generated during the collapse phase. As will be discussed in the forthcoming analysis, there is less viscous dissipation for large relaxation times, while small relaxation times make the surroundings capable of storing large tensile stresses during collapse. At the relaxation time where these two effects most constructively interfere, the maximum rebound radius occurs. By graphing J as a function of λ_1 , it is easily confirmed that the largest deviatoric stress contribution is generated for this same relaxation time. Predicting the relaxation time where this transition between regimes occurs is the subject of Sec. V A 1.

D. Harmonic forcing

The response of bubbles in various viscoelastic media to harmonic forcing is of interest to ultrasound applications. We consider the following dimensional forcing: $p_f(t) = P_A \sin(\omega t)$, with $P_A = 0.4 \text{ MPa}$ and $\omega/2\pi = 1.0 \text{ MHz}$. We choose a relatively low amplitude for this forcing frequency representative of medical ultrasound in order to highlight the effects of relaxation. Figs. 7 and 8 show the response of bubbles in linear and nonlinear viscoelastic media with different relaxation times. At this forcing amplitude and frequency, the response in the Newtonian and Kelvin–Voigt media does not exhibit substantial nonlinearities, while, for the media with relaxation, oscillations are significantly larger and more nonlinear. For low relaxation time in the linear media, the oscillations are out-of-equilibrium, in that the entirety of the oscillation cycle takes place at radii larger than the equilibrium radius. This is discussed in Sec. V B 2. At higher relaxation time, the oscillations gain amplitude and lose periodicity. The behavior in the nonlinear media is similar to the corresponding linear case.

FIG. 7. Bubble response to harmonic forcing in linear viscoelastic media at two relaxation times: (a) $\lambda_1 = 0.10 \mu s$ and (b) $\lambda_1 = 1.00 \mu s$. In each case, $\lambda_2 = \lambda_1/5$ for the Jeffreys model.

FIG. 8. Bubble response to harmonic forcing in nonlinear viscoelastic media at two relaxation times: (a) $\lambda_1 = 0.10 \mu s$ and (b) $\lambda_1 = 1.00 \mu s$. In each case, $\lambda_2 = \lambda_1/5$ for the Oldroyd-B model.

V. DISCUSSION AND ANALYSIS

The presence of relaxation time implies that the governing equation for bubble radius, when it can be written in a closed form, is at least third order in time. For simplicity in the following analysis, the surrounding medium is assumed incompressible with linear KVS viscoelasticity, and the bubble gas contents are polytropic. Under these conditions substituting Eq. (38) into Eq. (1), multiplying by $\exp(t/De)$, differentiating with respect to time, and simplifying yield

$$\begin{aligned} De \left(R\ddot{R} + 7\dot{R}\ddot{R} + \frac{9}{2} \frac{\dot{R}^3}{R} \right) + R\ddot{R} + \frac{3}{2} \dot{R}^2 = & \left(3De \frac{\dot{R}}{R} + 1 \right) \left(p + \frac{1 - 1/R}{We} - 1 - p_f(t) \right) \\ & + De \left(\dot{p} + \frac{\dot{R}}{WeR^2} - \dot{p}_f(t) \right) - \frac{4}{3Ca} \left(1 - \frac{1}{R^3} \right) - \frac{4\dot{R}}{ReR} - \frac{4}{Je} \left(\frac{2\dot{R}^2}{R^2} + \frac{\ddot{R}}{R} \right). \end{aligned} \quad (46)$$

This third-order ODE, for a bubble in an incompressible and linear Maxwell/Zener/Jeffreys material, forms the basis of our analysis. As $De \rightarrow 0$, while $Ca, Je \rightarrow \infty$, the traditional Rayleigh–Plesset equation is recovered.

A. Linear oscillations

We first consider linear oscillations about the nondimensional equilibrium radius $\mathcal{R}$, which is the real root of Eq. (45). For small-amplitude perturbations $a = R - \mathcal{R}$, the corresponding linearized system is

$$De\mathcal{R}\ddot{a} + \left(\mathcal{R} + \frac{4}{Je\mathcal{R}} \right) \ddot{a} + \left[\frac{4}{Re\mathcal{R}} + De \left(\omega_0^2\mathcal{R} - \frac{4(1 - 1/\mathcal{R}^3)}{Ca\mathcal{R}} \right) \right] \dot{a} + \left(\omega_0^2\mathcal{R} + \frac{4}{Ca\mathcal{R}^4} \right) a = -p_f(t) - De\dot{p}_f(t), \quad (47)$$

where

$$\omega_0^2 = \frac{3\kappa}{\mathcal{R}^{3\kappa+2}} - \frac{1}{We\mathcal{R}^3} \quad (48)$$

is the square of the bubble natural frequency in a Newtonian medium. Equation (47) is described by the transfer function

$$H(s) = \frac{-Des - 1}{De\mathcal{R}s^3 + \left(\mathcal{R} + \frac{4}{Je\mathcal{R}} \right) s^2 + \left[De \left(\omega_0^2\mathcal{R} - \frac{4(1 - 1/\mathcal{R}^3)}{Ca\mathcal{R}} \right) + \frac{4}{Re\mathcal{R}} \right] s + \omega_0^2\mathcal{R} + \frac{4}{Ca\mathcal{R}^4}}, \quad (49)$$

where s is the Laplace variable. The three poles s_1, s_2 , and s_3 of $H(s)$ give the damped frequency of the system,

$$\omega_D = \max \{ \Im(s_1), \Im(s_2), \Im(s_3) \}, \quad (50)$$

where $\Im$ denotes the imaginary part. When $\omega_D = 0$, the system is overdamped. As a measure of damping, we define the time constant as $t_C = -1/\Re(s_m)$, where s_m is the pole with the smallest magnitude and $\Re$ denotes the real part. As a third-order system, there is not a simple expression for these constants.

In Fig. 9, the damped frequency and the time constant computed numerically over a wide range of relaxation times are shown. The dependence of ω_D and t_C on relaxation is generally complicated. The system appears to go through two bifurcations as the bubble radius is increased. Considering $R_0 = 0.3 \mu\text{m}$, no oscillations occur until a critical relaxation time is achieved, at which point the oscillations exhibit a maximum in frequency. This value decreases for increasing relaxation time until a certain λ_1 , beyond which it tends towards a constant value. As the initial radius is increased, the critical relaxation time increases and the maximum oscillation frequency decreases. At a certain initial radius, a bifurcation occurs, in that the behavior at low relaxation times is no longer overdamped up to a relaxation time less than the critical relaxation time. As the initial radius is further increased, the oscillation frequency no longer changes with λ_1 . The relationship between relaxation and the driven oscillation response is investigated in Sec. V A 2.

FIG. 9. The dimensional damped frequency (a) and time constant (b) of the linearized system as a function of relaxation time for several initial radii. Computed for a Maxwell fluid and with $\mathcal{R} = 1$.

1. Relaxation regimes

The limits of large and small Deborah number allow the dynamics to be described by second-order ODEs. For large relaxation times, Eq. (47) yields

$$\mathcal{R}\ddot{a} + \frac{1}{\text{De}} \left(\mathcal{R} + \frac{4}{\text{Je}\mathcal{R}} \right) \dot{a} + \left(\omega_0^2 \mathcal{R} - \frac{4(1 - 1/\mathcal{R}^3)}{\text{Ca}\mathcal{R}} + \frac{4}{\text{DeRe}\mathcal{R}} \right) a = -\frac{1}{\text{De}} \int_0^t p_f(\xi) d\xi - p_f(t), \quad (51)$$

with time constant and oscillation frequency,

$$t_{C,\Lambda} = \frac{2\text{De}}{1 + \frac{4}{\text{Je}\mathcal{R}^2}}, \quad \omega_{D,\Lambda} = \sqrt{\omega_0^2 - \frac{4(1 - 1/\mathcal{R}^3)}{\text{Ca}\mathcal{R}^2} + \frac{4}{\text{DeRe}\mathcal{R}^2} - \frac{1}{4\text{De}^2} \left(1 + \frac{4}{\text{Je}\mathcal{R}^2} \right)^2}. \quad (52)$$

For oscillations about the equilibrium radius ($\mathcal{R} = 1$), it is not surprising that as $\text{De} \rightarrow \infty$, the frequency of oscillations approaches that of a bubble in an inviscid medium.³⁴ This behavior is expected since the constitutive model approaches the form $\dot{\tau}_{rr} = 0$ as De becomes large.

For small relaxation times, Eq. (47) yields

$$\left(\mathcal{R} + \frac{4}{\text{Je}\mathcal{R}} \right) \ddot{a} + \left[\text{De} \left(\omega_0^2 \mathcal{R} - \frac{4(1 - 1/\mathcal{R}^3)}{\text{Ca}\mathcal{R}} \right) + \frac{4}{\text{Re}\mathcal{R}} \right] \dot{a} + \left(\omega_0^2 \mathcal{R} + \frac{4}{\text{Ca}\mathcal{R}^4} \right) a = -p_f(t) - \text{De}\dot{p}_f(t), \quad (53)$$

with time constant and oscillation frequency,

$$t_{C,\lambda} = \frac{2 \left(\mathcal{R} + \frac{4}{\text{Je}\mathcal{R}} \right)}{\text{De} \left(\omega_0^2 \mathcal{R} - \frac{4(1 - 1/\mathcal{R}^3)}{\text{Ca}\mathcal{R}} \right) + \frac{4}{\text{Re}\mathcal{R}}}, \quad \omega_{D,\lambda} = \sqrt{\frac{\omega_0^2 \mathcal{R} + \frac{4}{\text{Ca}\mathcal{R}^4}}{\mathcal{R} + \frac{4}{\text{Je}\mathcal{R}}} - \frac{1}{4} \left[\frac{\text{De} \left(\omega_0^2 \mathcal{R} - \frac{4(1 - 1/\mathcal{R}^3)}{\text{Ca}\mathcal{R}} \right) + \frac{4}{\text{Re}\mathcal{R}}}{\mathcal{R} + \frac{4}{\text{Je}\mathcal{R}}} \right]^2}. \quad (54)$$

Fig. 10 shows that the small and large relaxation time behaviors are well captured by these limits. As in Fig. 9(b), for small relaxation times, the time constant is relatively small and does not strongly depend on λ_1 ; after a certain critical relaxation time, it increases linearly. The analysis clearly explains the behavior of ω_D in Fig. 9(a): the “left” branch of the solution (small relaxation time), which is only present for bubbles larger than a certain size, is given by Eq. (53); on the other hand, the “right” branch of the solution (large relaxation time) is given by Eq. (51). Comparing the two

FIG. 10. The dimensional damped frequency (a) and time constant (b) computed from the poles of (49), for $R_0 = 3 \mu\text{m}$, are overlaid by large relaxation time (52) and small relaxation time (54) approximations. When $3.69 \text{ ns} \lesssim \lambda_1 \lesssim 18.3 \text{ ns}$, the system is overdamped.

second-order systems, it is interesting to note that the restoring parameter in Eq. (51) becomes the damping parameter in Eq. (53) as relaxation time is decreased.

To predict the values of λ_1 at which these transitions occur, we first consider small λ_1 , where overdamping is observed for certain R_0 in Fig. 9. To simplify the analysis, we consider the Maxwell case ($\text{Ca}, \text{Je} \rightarrow \infty$). Equation (54) gives an approximation of the frequency of free oscillations. When $\omega_{D,\lambda} = 0$, the system is critically damped; the Deborah number at which this occurs is

$$\text{De}_{C1} = \frac{2}{\omega_0} \left(1 - \frac{2}{\omega_0 \text{Re} \mathcal{R}^2} \right). \quad (55)$$

If λ_1 is small enough that Eq. (53) holds and if $\text{De} > \text{De}_{C1}$, the system is overdamped. However, as Fig. 9 shows, in certain cases the system is underdamped for all λ_1 . Although $\omega_{D,\lambda} = 0$ for some λ_1 (critical damping for the small- λ_1 case), there are only certain parameters for which $\omega_D = 0$ (critical damping for the general case) is possible. In general, for any λ_1 , third-order ODE (47) is critically damped,⁵⁸ if $(9c_3 - c_1c_2)^2 = 4(c_1^2 - 3c_2)(c_2^2 - 3c_1c_3)$. Here, c_1, c_2 , and c_3 are the constants that appear by writing the homogeneous form of Eq. (47) as $\ddot{a} + c_1\dot{a} + c_2\ddot{a} + c_3a = 0$. This equation can be solved for a second critical Deborah number, De_{C2} , although the solution does not have a simple closed form. A reasonable approximation of the solution for physical parameters, however, is

$$\text{De}_{C2} \approx \frac{\text{Re} \mathcal{R}^2}{16}, \quad (56)$$

which arises by letting $\omega_0 \rightarrow 0$. The smallest possible equilibrium radius for which the system is underdamped ($R_{C,eq}$, the dimensional critical equilibrium radius) is given by $\text{De}_{C1}(R_{C,eq}, R_{C,0}) = \text{De}_{C2}(R_{C,0})$. Note that the critical initial radius $R_{C,0}$ has not been fixed, and that De_{C1} and De_{C2} depend on $R_{C,0}$ through the dimensionless parameters. Using the approximate form of De_{C2} in Eq. (56), the condition $\text{De}_{C1} = \text{De}_{C2}$ gives $\mathcal{R}_C \equiv R_{C,eq}/R_{C,0}$ implicitly by

$$\frac{2}{\omega_0} \left(1 - \frac{2}{\omega_0 \text{Re} \mathcal{R}_C^2} \right) = \frac{\text{Re} \mathcal{R}_C^2}{16}. \quad (57)$$

This equation can be solved numerically, simultaneously with equilibrium condition (45), to find $R_{C,eq}$ and $R_{C,0}$. The critical initial radius and critical Deborah number have the following interpretation: if the initial radius R_0 is greater than $R_{C,0}$ or the Deborah number De is greater than De_C , then the linearized system is underdamped. This result is for a Maxwell fluid, but the same is expected to

FIG. 11. $R(t)$ curves in a Maxwell fluid for choices of R_0 near $R_{C,0}$ and λ_1 near λ_C . Surface tension was neglected so that (58) accurately approximates $R_{C,0}$. The high and low values of R_0 were $2R_{C,0}$ and $R_{C,0}/2$, respectively, while the high and low values of λ_1 were $5\lambda_C$ and $\lambda_C/5$. The isothermal approximation ($\kappa = 1$) was made to focus on viscoelastic effects, so that $\mu = 1.0$ Pa s implies $R_{C,0} = 120$ μm and $\lambda_C = 950$ ns. For both the full solution and the linearized form in Eq. (47), the solution is only overdamped in (a), where both $\lambda_1 < \lambda_C$ and $R_0 < R_{C,0}$.

roughly hold for Kelvin–Voigt and Jeffreys materials if the elastic modulus and retardation time are not overly large. A numerical demonstration of this result is shown in Fig. 11.

$R_{C,0}$ can be approximated from (57). We let $\Delta P = 0$ (that is, $R_{C,eq} = R_{C,0}$) and $S = 0$. With zero surface tension $\omega_0 = \sqrt{3}\kappa$, and Eq. (57) becomes a quadratic equation in $R_{C,0}$ after dimensionalizing the Reynolds number. The smallest of the two roots gives an approximation of the critical initial radius

$$R_{C,0} \approx \frac{8(2\sqrt{3} - 3)}{3} \frac{\mu}{\sqrt{\kappa p_\infty \rho_\infty}}. \quad (58)$$

As shown in Fig. 12, Eq. (58) provides an upper bound on $R_{C,0}$ for nonzero surface tension. The associated critical relaxation time, from either Eq. (55) or Eq. (56), is

$$\lambda_C \approx \frac{4(7 - 4\sqrt{3})}{3} \frac{\mu}{\kappa p_\infty}. \quad (59)$$

FIG. 12. The approximate critical radius in a Maxwell fluid as a function of viscosity by solving Eq. (57) with $\Delta P = 0$. The line with $S = 0$ corresponds to Eq. (58); this approximation is best in cases of high viscosity.

2. Forced oscillations: Harmonic response

For a bubble forced harmonically at dimensionless angular frequency ω and amplitude ϵ , the linear oscillations following an initial transient are of the form

$$a(t) = \epsilon |H(i\omega)| \cos(\omega t + \phi), \quad (60)$$

where i is the imaginary unit, and where the gain $|H(i\omega)|$ and the phase angle ϕ are given, respectively, by the magnitude and argument of $H(i\omega)$ from Eq. (49). A simple expression for the frequency that maximizes the gain (peak frequency) can be found for a Zener solid in the limit $\text{Ca} \rightarrow \text{ReDe}$. In this case, the gain is

$$\lim_{\text{Ca} \rightarrow \text{ReDe}} |H(i\omega)| = \left| \frac{\text{DeRe}}{4 + \text{DeRe}(\omega_0^2 - \omega^2)} \right|. \quad (61)$$

Thus, the resonance frequency is

$$\omega_P = \sqrt{\frac{4}{\text{DeRe}} + \omega_0^2}. \quad (62)$$

Although this peak frequency ω_P arises from a special case, it provides an excellent approximation of the true peak frequency for small Reynolds number, whether in Zener, Maxwell, or Jeffreys materials. For large Reynolds or Deborah numbers, the peak frequency approaches ω_0 .

In Fig. 13, the gain of the linear system is compared to that of the full nonlinear model when subjected to harmonic forcing of amplitudes $\epsilon = p_\infty/10p_0$ and $p_\infty/2p_0$. A 200×200 logarithmically spaced set of relaxation times and frequencies is used in the simulations, with the isothermal approximation ($\kappa = 1$) for simplicity. The surroundings are taken to be a linear Maxwell fluid. At low amplitudes, the linear frequency response agrees relatively well with the full nonlinear solution, except for the appearance of a harmonic. When high-amplitude resonance occurs, the resonance frequency is largely independent of relaxation time, which, for these parameters, is approximately 1 MHz. As the relaxation time is decreased, this resonance peak shifts toward higher frequencies, up to a certain value of λ_1 (λ_C from Sec. V A 1) beyond which the oscillation amplitude is not very large ($\lesssim 0.2$). The effectiveness of Eq. (62) for approximating the peak frequency is demonstrated for several models and parameters in Figs. 14 and 15. In Fig. 14, significant resonance only occurs when $\lambda_1 > \lambda_C$, except in (c), where $R_0 > R_{C,0}$, as expected. In Fig. 15, the relaxation time is set very low ($\lambda_1 = 10$ ns), such that $\lambda_1 < \lambda_C$.

3. Forced oscillations: Step and impulse responses

From Figs. 5(c) and 6(c), the instantaneous response of a bubble does not depend on relaxation time, although at later times, the damping and restoration forces exhibit a strong dependence on λ_1 . The early time behavior can be explained by considering the linearized third-order system for a step change in the far-field pressure amplitude $\Delta P/p_0$. For such forcing, the solution $a(t)$ in Laplace space is given by

$$A(s) = \frac{\Delta P}{p_0} \frac{H(s)}{s}. \quad (63)$$

The initial value theorem gives the initial acceleration of the bubble wall as

$$\ddot{a}(0^+) = \lim_{s \rightarrow \infty} s^3 A(s) = -\frac{\Delta P}{p_0 \mathcal{R}}, \quad (64)$$

despite the initial condition $\ddot{a}(0) = 0$. Thus, the initial acceleration is independent of relaxation time, though it may depend on the elastic modulus through $\mathcal{R}$. The same analysis can be used for impulse response, with a delta function of amplitude $\Delta P/p_0$ as the forcing pressure. In this case, $\dot{a}(0^+) = -\frac{\Delta P}{p_0 \mathcal{R}}$, $\ddot{a}(0^+) \rightarrow \infty$, and the instantaneous response remains independent of relaxation time. At later times, however, relaxation time significantly impacts bubble response. This matter is revisited in Sec. V B 1 in the context of growth.

FIG. 13. The gain of the linear system (a) computed from $H(i\omega)$ is compared to the actual gain computed from the full model, defined as $(R_{\max}/R_0 - 1)/\epsilon$, for two different forcing amplitudes in (b) and (c), where $R_{\max}$ is the maximum dimensional radius achieved during steady oscillations. Several lineouts are taken in (d) for relaxation times in the vicinity of λ_C .

B. Analysis of the nonlinear response

1. Enhanced growth

An important effect of relaxation is increased bubble wall acceleration during growth (e.g., Fig. 5(a)) compared to models without relaxation. This behavior can be explained as follows. Suppose that, at time $t = 0$, a bubble starts from rest in a Zener medium and grows due to a decrease in surrounding pressure until some time t_f when $\dot{R}$ is first zero. Strain function (8) and its time derivative are given by

$$\Phi = \frac{R^3 - 1}{3Ca} + \frac{R^2 \dot{R}}{Re}, \quad \dot{\Phi} = \frac{R^2 \dot{R}}{Ca} + \frac{2R\dot{R}^2 + R\ddot{R}}{Re}. \quad (65)$$

Since $\Phi(0) = 0$ and $\dot{\Phi}(t) > 0$ for $0 < t \leq t_f$, the strain function Φ is positive and monotonically increasing in the interval $(0, t_f)$, i.e., during the initial growth phase. As a result, during this interval,

FIG. 14. The gain of the linear system, $\log_{10}|H(i\omega)|$, as a function of frequency and relaxation time. Each case uses different constitutive properties: (a) Maxwell fluid ($\mu = 30$ cP), (b) Jeffreys fluid ($\mu = 300$ cP, $\lambda_2 = 10$ ns), and (c) low-viscosity Maxwell fluid ($\mu = 1.0$ cP). The solid curve follows the peak frequency given by Eq. (62), derived for a Zener solid with $\text{Ca} \rightarrow \text{DeRe}$. The dashed curve denotes the critical relaxation time as a function of frequency approximated by Eq. (59).

$$\int_0^t e^{(\xi-t)/\text{De}} \Phi(\xi) d\xi < \Phi(t) \int_0^t e^{(\xi-t)/\text{De}} d\xi = \text{De}\Phi(t) (1 - e^{-t/\text{De}}) < \text{De}\Phi(t). \quad (66)$$

When relaxation is neglected, the stress integral is given by $J_{\text{De}=0} = -4\Phi/R^3$, as opposed to $J_{\text{De}\neq 0}$ given in Eq. (38). Therefore, during the initial growth phase, $J_{\text{De}\neq 0} > J_{\text{De}=0}$. From the radial dynamics equation (1), a positive J decreases the effective external pressure. Or, in this case, when J is less negative, the effective amplitude of the negative acoustic pressure increases. Therefore, since $J_{\text{De}\neq 0} > J_{\text{De}=0}$, the presence of relaxation accelerates the initial growth of the bubble. Furthermore, the factor $(1 - e^{-t/\text{De}})$ in Eq. (66) shows that *as relaxation time increases, it takes more time for this growth-enhancing phenomenon to decay*. This explains the trends in Fig. 5(d). This analysis can also be applied to a Jeffreys fluid as long as the bubble wall jerk $\ddot{R}$ does not cause $\dot{\Phi}$ to fall below zero ($\ddot{R}$ is not positive for all $t < t_f$). This again agrees with the results in Fig. 5, where, during growth, the bubble radius in the Jeffreys fluid is bounded above by that in the Maxwell fluid.

2. Out-of-equilibrium oscillations

Under certain conditions, the presence of relaxation gives rise to steady, out-of-equilibrium bubble oscillations, as shown in Fig. 16. After about $10 \mu\text{s}$, the harmonically forced bubble enters steady oscillations in which its minimum radius (≈ 2.5) is greater than its initial (equilibrium) radius. Throughout this process, the bubble pressure remains low and contributes little to the rebound. This phenomenon, only observed for nonzero relaxation time, lies in contrast with archetypal inertial

FIG. 15. The same study as in Fig. 14, but for varying initial radii with λ_1 fixed at 10 ns. The dashed curve now denotes the critical initial (equilibrium) radius.

FIG. 16. Out-of-equilibrium oscillations due to periodic forcing in an upper-convected Maxwell fluid. The acoustic pressure $p_f(t)$ is overlaid above. Parameters are as usual but with $R_0 = 1.0 \mu\text{m}$, $\lambda_1 = 0.20 \mu\text{s}$, $\omega/2\pi = 1.3 \text{ MHz}$, and $P_A = 300 \text{ kPa}$.

cavitation, where a bubble, having reached a large maximum radius, collapses to a small radius. Rather, in these out-of-equilibrium oscillations, the rebound is driven by tensile stresses in the surrounding medium.

Consider a bubble in a Maxwell medium, under no forcing, starting from rest at a radius much larger than the equilibrium radius $\mathcal{R}$. As the bubble collapses, approaching $\mathcal{R}$, suppose it rebounds at some time t_R , at a radius $R(t_R)$ still much larger than the equilibrium radius. This behavior is readily observed in simulations. The bubble radius at t_R is a local minimum; hence, $\dot{R}(t_R) = 0$ and $\ddot{R}(t_R) > 0$. Since $R(t_R) \gg 1$, we can let $p(t_R) \rightarrow 0$ and ignore surface tension. Evaluating Eq. (46) at this instant yields the minimum radius

$$R(t_R) = \frac{-1}{\text{De} \ddot{R}(t_R) + \ddot{R}(t_R)}. \quad (67)$$

Since $\ddot{R}(t_R) > 0$, $R(t_R)$ is only positive and large if $\ddot{R}(t_R) < 0$ and if De is nonzero. In the absence of relaxation, Eq. (67) predicts a negative rebound radius, meaning that large, unforced, out-of-equilibrium oscillations are impossible. In such media (Newtonian, Hookean, or Kelvin–Voigt), rebound can only occur when $R < \mathcal{R}$ and will be governed by internal pressure and/or elastic effects. This analysis indicates that, independent of forcing, media that exhibit stress relaxation have an intrinsic ability to support out-of-equilibrium oscillations. Of course, an additional mechanism is granted by nonzero forcing, which, when present, sets the numerator in Eq. (67) to $-(1 + p_f + \text{De} \dot{p}_f)$. This shows that when relaxation is present, the time rate of change of acoustic pressure can influence out-of-equilibrium rebound, in addition to acoustic pressure itself. For example, for harmonic forcing given dimensionally by $p_f(t) = P_A \sin(\omega t)$, the numerator in Eq. (67) can be as large as $\frac{P_A}{p_0} \sqrt{1 + \lambda_1^2 \omega^2} - 1$. Overall, however, the amalgamation of these different analytical factors confirms the computational result that, for a given forcing, out-of-equilibrium oscillations are much more likely to occur in media with relaxation.

VI. CONCLUSIONS

A numerical model for the simulation of spherical gas bubble dynamics in various isotropic viscoelastic media has been presented. Our approach is based on the compressible Keller–Miksis equation with the addition of thermal effects inside and outside the bubble. A Chebyshev spectral collocation method was developed to treat general nonlinear constitutive models. Simple ODE formulations were derived for common upper-convected and linear viscoelastic models. A series of canonical problems were investigated numerically and theoretically, with results leading to the following conclusions:

- The computational strategy presented here effectively and robustly treats spherical bubble dynamics in viscoelastic media.
 - The spectral method accurately computes the viscoelastic stress fields, even in cases of violent bubble collapse. This method can easily be adapted for nonlinear constitutive models other than those presented and can be replaced by ODE formulations for several common constitutive models.
 - Although compressibility and heat transfer increase the computational complexity of the model, they strongly influence viscoelastic bubble dynamics for the problems under consideration (see Appendix A).
- Bubble response to various types of forcing heavily depends on relaxation time.
 - Relaxation causes faster growth due to the manner in which it modifies the integral over the stress field. Increasing the relaxation time increases the bubble growth, but not necessarily the maximum growth rate.
 - For low relaxation times, the surroundings behave as a Newtonian fluid (when elasticity is absent), while for large relaxation times, the surroundings tend toward an inviscid medium. We can approximate the critical radius and relaxation time that separate these two regimes and determine the state of damping of the linearized system. Although the behavior at either end of the spectrum is clear, real viscoelastic materials are expected to exist between these extremes.
 - Differences between the various nonlinear viscoelastic models are most prominent for large relaxation times, which lead to less damping and thus more violent, less periodic responses. Retardation generally damps the bubble response.
 - Relaxation permits bubble rebound driven purely by residual stresses in the surroundings—a phenomenon which Newtonian media cannot support. As a result, relaxation makes steady, out-of-equilibrium oscillations readily possible, as well as unforced, larger-than-equilibrium rebound.

The tool developed in this work has a broad range of applications throughout biomedicine and industry. The model provides radius-time data for a bubble response, as well as the complete temperature, velocity, pressure, and deviatoric stress fields in the surroundings. In therapeutic ultrasound, for example, given an incoming pressure waveform, this model could be used to calculate stresses produced on cells near a cavitation bubble. The efficiency of the method makes this model ideal for cavitation rheology, where the results must be fitted to an experimental radius-time curve by varying the viscoelastic parameters. Incorporating additional viscoelastic models (such as the generalized Maxwell and Burgers materials), treating compressibility in the stress calculation, and accounting for mass transfer would increase the model's scope. For simulating bubbles in anisotropic materials or non-symmetric geometries, more sophisticated computational approaches are necessary.

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APPENDIX A: THERMAL EFFECTS

The PDEs for the temperature fields are effectively treated by spectral collocation, as done by others.^{51,52,55} This method is summarized below, and a modification to this technique for the external temperature field is proposed. By means of the coordinate transformation $y = r/R$, the domain $r \in [0, R(t)]$ of gas energy equation (11) is immobilized and projected onto $y \in [0, 1]$. It is further convenient to introduce the change of dependent variables from the dimensionless

temperature T to the integrated quantity

$$\sigma(y, t) = \frac{1}{K(T_\infty)} \int_1^T K(T_\infty \theta) d\theta, \quad (\text{A1})$$

where K is in dimensional units and is given by $K(T_\infty \theta) = K_A T_\infty \theta + K_B$. In terms of the new variables, the gas energy equation becomes

$$\frac{\partial \sigma}{\partial t} = D \dot{p} + \frac{\chi}{R^2} \left[\frac{2D}{y} - \frac{\kappa - 1}{\kappa p} \left(\frac{\partial \sigma}{\partial y} - \frac{\partial \sigma}{\partial y} \Big|_{y=1} y \right) \right] \frac{\partial \sigma}{\partial y} + \frac{\chi D}{R^2} \frac{\partial^2 \sigma}{\partial y^2}. \quad (\text{A2})$$

The dimensionless gas thermal diffusivity $D = (1 - 1/\kappa)(\alpha T^2 + (1 - \alpha)T)/p$ can be evaluated by inverting Eq. (A1), giving $T = (\alpha - 1 + \sqrt{1 + 2\alpha\sigma})/\alpha$. Here, the dimensionless parameters are $\chi = T_\infty K(T_\infty)/p_0 R_0 u_c$ and $\alpha = K_A T_\infty / K(T_\infty)$. Using the new dependent variable and coordinate system, we make the spectral truncation approximation $\sigma(y, t) \approx \sigma^P(y, t) = \sum_{n=0}^P a_n(t) T_{2n}(y)$. By using only the even Chebyshev polynomials T_{2n} as the basis functions, we automatically satisfy the symmetry condition at $r = 0$. Replacing σ by σ^P in (A2) and evaluating at the Chebyshev-Gauss-Lobatto collocation points $y_j = \cos(\pi j/2P)$, $j = 1, 2, \dots, P$, we form a linear system of P equations for $P + 1$ unknown spectral coefficient time derivatives, $\dot{a}_n$. For an additional equation, attention must be paid to the interface conditions; hence, we turn to the temperature distribution in the surrounding medium.

Previous authors^{51,55} have used the coordinate transformation $1/x = 1 + (y - 1)/L_t$ to project the energy equation of surrounding medium (12) from the semi-infinite domain $r \in [R(t), \infty)$ to the bounded stationary domain $x \in [1, 0)$, where L_t is a thermal diffusion characteristic length. Doing so allows a basis of even Chebyshev polynomials to satisfy the boundary condition of zero temperature gradient at infinity (where $x = 0$). However, *all* Chebyshev polynomials that are a function of x under this coordinate transformation have a zero gradient as $r \rightarrow \infty$. Therefore, to ensure a complete representation of the temperature distribution, we use both even and odd Chebyshev polynomials in the spectral truncation approximation $T_M(x, t) \approx T_M^Q(x, t) = \sum_{n=0}^Q b_n(t) T_n(x)$, along with the new coordinate transformation

$$x = \frac{2}{1 + (y - 1)/L_t} - 1 \quad (\text{A3})$$

on the domain $[1, -1)$. Under this transformation, the energy equation in the surroundings takes the form

$$\frac{\partial T_M}{\partial t} = \frac{(1+x)^2}{L_t R} \left[\text{Fo} \left(\frac{1+x}{2L_t} - \frac{1}{y} \right) + \frac{\dot{R}}{2} \frac{1-y^3}{y^2} \right] \frac{\partial T_M}{\partial x} + \frac{\text{Fo} (1+x)^4}{4} \frac{\partial^2 T_M}{L_t^2 R^2 \partial x^2} - 2\text{Br} \frac{\dot{R}}{R y^3} (\tau_{rr} - \tau_{\theta\theta}). \quad (\text{A4})$$

FIG. 17. Thermal and compressible effects for (a) Rayleigh growth and (b) Rayleigh collapse in an upper-convected Maxwell material, using the standard parameters of Figs. 5 and 6.

FIG. 18. The internal bubble pressure and the temperature at the bubble's center for the Gaussian collapse case in Fig. 3.

Here, the Fourier number is $Fo = D_M/u_c R_0$, the Brinkman number is $Br = u_c^2/C_p T_\infty$, and Eq. (A3) gives y as a function of x . If stress-induced heat generation is significant, the spectral method is required to obtain the complete stress distributions in the medium, except when Newtonian and Kelvin–Voigt models are used. While τ_{rr} and $\tau_{\theta\theta}$ are treated as functions of ζ , Eqs. (14) and (A3) together give ζ as a function of x . Evaluating Eq. (A4) at the Chebyshev–Gauss–Lobatto collocation points $x_j = \cos(\pi j/Q)$, $j = 1, 2, \dots, Q$, we form a system of Q unknowns for the $Q + 1$ time-derivatives of the spectral coefficients b_n . Substituting the spectral expansions into the interface conditions, Eq. (10), gives a total of $P + Q + 2$ equations. At $x = -1$, Eq. (A4) reduces to $\partial T_M / \partial t|_{x=-1} = 0$, which automatically satisfies the boundary condition at infinity.

Fig. 17 highlights the role of thermal as well as compressible effects in the present model. For Rayleigh growth, the compressible correction is not important, but there is a large discrepancy between the full thermal model and the commonly used adiabatic approximation. For strong collapse, the compressible corrections are responsible for significant damping. Following collapse, neither the adiabatic nor isothermal models accurately approximate the full model, although at very late times, the isothermal model predicts the correct final radius. Fig. 18 shows an example of internal bubble pressure and temperature at the center of the bubble, predicted by the full thermal model for the Gaussian collapse case in Fig. 3.

APPENDIX B: FAST CHEBYSHEV DIFFERENTIATION

The following technique can be used to write the discrete Chebyshev transform in terms of the discrete Fourier transform, whereby the FFT accelerates computations for large N . The Chebyshev spectral coefficients $\{c_n\}_{n=0}^N$ and the physical data $\{p(x_j)\}_{j=0}^N$ located at the Chebyshev–Gauss–Lobatto points $x_j = \cos(\pi j/N)$ are related by the forward and inverse discrete Chebyshev transforms

$$c_n = \frac{2}{f_n N} \sum_{j=0}^N \frac{p(x_j)}{f_j} T_n(x_j) \quad \text{and} \quad p(x_j) = \sum_{n=0}^N c_n T_n(x_j), \quad (\text{B1})$$

respectively, where $f_n = 1$ for all n , except $f_0 = f_N = 2$. Letting $c_{2N-n} = c_n$ and using the Chebyshev polynomial property $T_n(x_j) = \frac{1}{2}(e^{i\pi j n/N} - e^{-i\pi j n/N})$, it follows that

$$\sum_{n=0}^N c_n T_n(x_j) = \frac{1}{2} \sum_{n=0}^{2N-1} f_n c_n e^{i\pi j n/N}, \quad j = 0, 1, \dots, N. \quad (\text{B2})$$

Thus, the inverse Chebyshev transform of $\{c_n\}_{n=0}^N$ is proportional to the inverse Fourier transform of $\{f_n c_n\}_{n=0}^{2N-1}$. Via FFT, the computational cost of evaluating Eq. (B2) is reduced from $O(N^2)$ to $O(N \log N)$. The derivative of the interpolated function $p(x)$, evaluated at the collocation points,

$$\left. \frac{dp}{dx} \right|_{x=x_j} = \frac{\csc(\pi j/N)}{2i} \sum_{n=0}^{2N-1} n f_n c_n e^{i\pi j n/N}, \quad j = 0, 1, \dots, N, \quad (\text{B3})$$

can again be computed via FFT. While Eq. (B3) is undefined at the endpoints $x = 1, -1$, l'Hôpital's rule can be used to determine the limiting expressions. Expressions for higher derivatives can be procured in an analogous manner, as can the forward Chebyshev transform.

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