

## Introduction



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# Advanced materials modelling via fractional calculus: challenges and perspectives

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Fractional calculus is now a well-established tool in engineering science, with very promising applications in materials modelling. Indeed, several studies have shown that fractional operators can successfully describe complex long-memory and multiscale phenomena in materials, which can hardly be captured by standard mathematical approaches as, for instance, classical differential calculus. Furthermore, fractional calculus has recently proved to be an excellent framework for modelling non-conventional fractal and non-local media, opening valuable prospects on future engineered materials. The theme issue gathers cutting-edge theoretical, computational and experimental studies on advanced materials modelling via fractional calculus, with a focus on complex phenomena and non-conventional media.

This article is part of the theme issue 'Advanced materials modelling via fractional calculus: challenges and perspectives'.

## 1. Overview

Fractional operators may be considered as integro-differential operators of the convolution type with hypersingular power-law kernels. Moving from the celebrated letter of De L'Hospital to Leibniz in 1695, discussing the concept of derivative of order  $\frac{1}{2}$ , the mathematical bases of fractional differentiation

and fractional integration were set by prominent mathematicians such as Liouville, Grünwald, Letnikov, Riesz, Caputo and many others up to recent times [1–5].

For a long time, however, fractional calculus was regarded as an elegant yet purely theoretical field of mathematics, with limited practical use. The relevance of fractional calculus in applied science has progressively grown in recent years and, now, a considerable number of studies have definitely unveiled its potential to address several problems, especially in materials science and engineering. Indeed, power-law dependence of fractional operators has proved ideally suitable to model non-local behaviour of materials in time or space, which plays a crucial role in several phenomena but cannot be captured by standard mathematical approaches as, for instance, classical differential calculus; in this context, fractional operators have been fruitfully applied to describe challenging phenomena such as viscoelasticity, heat conduction, diffusion in porous media and wave propagation. On the other hand, fractional calculus has provided a consistent framework to model non-conventional media, e.g. fractal and non-local ones, opening unexpected opportunities in the design of new materials. Understanding the complex behaviour of materials and modelling non-conventional media are, perhaps, the most fascinating and increasingly relevant applications of fractional calculus, whose potential impact on materials science and engineering has to be fully investigated.

Although an exhaustive description of all aspects of fractional calculus applications to materials modelling is almost prohibitive, this theme issue will attempt to provide a broad perspective on the state of the art and most recent developments, with 14 papers on viscoelasticity, heat conduction and diffusion in porous media, non-local continua and fractal media. The applications will involve a wide variety of fractional operators, with a constant order as well as variable one; the interest in fractional operators of variable order is growing in the recent literature, as valuable tools to model evolutionary phenomena without changing the governing equations.

## 2. Material hereditariness: viscoelasticity

Time non-locality of fractional operators is ideally suitable to model material hereditariness and, specifically, viscoelastic behaviour.

Early studies in this field are attributed to Boltzmann [6] and Volterra [7], who introduced the concept of the constitutive equation given by a convolution integral modelling memory of the past strain or stress histories. Pioneering work on fractional viscoelasticity was carried out by Nutting [8], who observed that the stress–strain datasets of many complex materials do exhibit a power-law relaxation, and by Gemant [9] and Bosworth [10], the first to propose a fractional-derivative model for the constitutive behaviour of viscoelastic media on introducing the power-law kernel in the Boltzmann–Volterra convolution integral. The use of fractional derivatives to fit experimental data was later pursued by Scott-Blair & Caffyn [11] and Caputo [12]. However, a first attempt to provide a theoretical basis for a fractional-derivative modelling of viscoelasticity was due, at the beginning of the 1980s, to Bagley & Torvik [13], who framed their model in the context of molecular theory. They also showed that, in order to capture the frequency dependence of damping properties in some viscoelastic materials, fractional derivatives are more appropriate than classical rheological models such as the Kelvin–Voigt model [14,15]; a further advantage over complex stiffness models of damping is that non-causal responses are avoided. In the last two decades, fractional viscoelastic models have been applied successfully in numerous studies, proving capable of describing complex material behaviours at a macroscopic level in the form of equations involving a small number of parameters and becoming, therefore, a well-established approach for viscoelastic media of various nature [16–20]. Studies in [13–20] deal with linear viscoelasticity and a comprehensive historical perspective on fractional calculus applications in this field has been outlined by Mainardi [21].

Experimental data fitting by linear fractional viscoelastic models has been reported in several studies, using various fractional operators. Caputo fractional-derivative viscoelastic models have

been proposed by Di Paola *et al.* for polymers [22] and epoxy resins [23], by Mahiuddin *et al.* [24] for fruit and vegetables; for example, experimental data obtained by Mahiuddin *et al.* [24] have demonstrated that the fractional order is closely related to the material's relaxation modulus and eventually to the degree of permanent deformation in the food tissue.

Along with successful experimental data fitting, considerable insights on the theoretical foundations of linear fractional viscoelastic models have been gained in recent years. For instance, a consistent mechanical description of the insurgence of power laws has been provided by Di Paola & Zingales [25], who have shown that a Couette problem involving heterogeneous fluid with prescribed features yields power laws with order related to the decay of fluid properties. A thermodynamic picture involving the notion of state of a material with power-law memory has been reported by Deseri *et al.* [26], proving that the state is determined by fractional-order integrals, or derivatives, of the stress or strain histories up to time  $t=0$ . A thermodynamic justification of power-law fading memory in terms of the Clausius–Duhem inequality has been also provided by Deseri *et al.* [27], showing that the mechanical energy dissipation corresponds to the Stavermann–Schwarzl dissipation [28]. Other interesting contributions on thermodynamics in fractional-order viscoelasticity may be found in some recent papers [29]. A multiaxial model of fractional-order hereditariness for isotropic materials has been recently provided [30] upon introducing thermodynamic restrictions on the orders of the power laws involved in the mathematical models of transverse and axial creep as well as relaxation functions.

An emerging issue in viscoelasticity theory is how to model mechanical properties of materials that may modify as a result of various phenomena. For this purpose, several authors have proposed variable-order fractional operators, i.e. operators whose order changes with time or depending on specific state variables. The interest traces back to the early work of Samko & Ross [31,32], and some other contributions to the development of variable-order operators have been provided at the beginning of the last decade [33,34]. Variable-order fractional operators with explicit dependence on a temperature field modelled as random noise have been proposed in [35]. Furthermore, the use of variable-order fractional calculus has been discussed by Beltempo *et al.* [36] to address ageing of pre-stressed concrete as an alternative to model B3 [37]. Variable-order fractional calculus has been applied to model ageing concrete, obtaining mathematically consistent relaxation functions to be coded into finite-element specific algorithms for a computer simulation of real-case structures [38,39].

Interesting applications of variable-order fractional differential calculus have been proposed to tackle nonlinear problems. In this regard, a position-depending variable-order fractional derivative, built as a generalization of the Caputo fractional derivative, has been used by Coimbra [40] to describe the mechanics of a mass oscillating on a guide covered with a non-uniform viscoelastic film, featuring a continuous variation of the order of the frictional force; the consistency of the proposed solution has been showed by comparison with an interpolative solution of a nonlinear fixed-order differential equation. Using the variable-order fractional derivative introduced by Coimbra [40], Ramirez & Coimbra [41] have developed a variable-order macroscopic constitutive relation for viscoelastic composite materials under compression at various constant strain rates; using a statistical mechanics approach for fitting experimental data on epoxy resin and a carbon/epoxy composite, the authors showed that the variable order of the operator is connected to the rate of change of the long-range order of the molecules at the mesoscale within the material. A further successful application of the variable-order operator proposed by Coimbra [40] can be found in the recent study of Meng *et al.* [42] to fit the stress responses of two polymers at various temperatures and across the glass transition; the connection between the variable order and the true strain was investigated at various temperatures. Ingman & Suzdalnitsky [43] have described the behaviour of a polymeric material using a fractional operator of time-dependent order, built as generalization of the Riemann–Liouville operator; in this case, the order varies with respect to the independent variable of the problem. Variable-order fractional operators have been used to capture nonlinear behaviour of metals and asphalt

mixtures [44,45]. The advantages of using variable-order operators to tackle nonlinear problems are described by Patnaik *et al.* [46], with focus on several application fields including nonlinear viscoelasticity.

Finally, a quite recent and promising application field for fractional viscoelastic models is the characterization of biological and biomedical systems. Fractional viscoelastic models have been proposed to fit experimental data on lung tissue [47], arterial blood flow [48], interfaces formed by bovine serum albumin and solution of acacia gum [49]. Indeed, many biological materials exhibit power-law dependence of stress relaxation, often with marked nonlinear dependence on the applied strain as it has been observed also for aortic valves.

The theme issue includes several contributions on linear and nonlinear fractional viscoelasticity, as applied to various types of materials [50–55]. Atanackovic *et al.* [50] investigate the thermo-dynamical restrictions on constitutive equations for viscoelastic fluids, as following from a weak form of entropy inequality under isothermal conditions. The restrictions are derived for fractional Burgers models and a more general class of linear constitutive equations with fractional derivatives. The authors show that the proposed restrictions are found to be weaker than classical existing ones, offering potentially more versatile approaches to capture creep behaviour in viscoelastic media. Ionescu *et al.* [51] focus on non-Newtonian fluids. The authors propose a minimal parameter (five) fractional-order impedance model to capture various degrees of viscoelasticity in non-Newtonian fluids. Using a frequency-identification method based on nonlinear least-squares, genetic and particle-swarm algorithms, the model proves to fit very well experimental data for oil, sugar, detergent and liquid soap over wide frequency ranges. A link between certain properties of the fluid and specific parameters of the fractional-order impedance model is also suggested. The work is complemented by a critical discussion on further potential applications of the model as well as on its limitations. Fang *et al.* [52] propose a three-branch fractional-derivative viscoelastic model for solid propellants. The model provides a good agreement with experimental data in terms of stress relaxation modulus and storage modulus, using a limited number of parameters compared with traditional models containing integer-order derivatives; the study is complemented by a simple and effective direct search method for data fitting. Tenreiro Machado *et al.* [53] address the mechanical characterization of epoxy resins via electrical impedance spectroscopy and fractional calculus tools. The authors compare integer and fractional models, proving that the latter is more effective than the former at low frequencies and require, in general, less parameters to achieve accurate fitting to experimental data. Finally, the electrical impedance spectroscopy data gathered from the epoxy samples are compared with those of different adhesives and sealants by means of a hierarchical clustering algorithm to detect the relationships between the distinct materials. Di Paola *et al.* [54] introduce a novel approach to time-dependent, variable-order fractional viscoelasticity. Moving from the observation that, in this case, the Boltzmann linear superposition principle does not apply in standard form because the fractional order is not constant with time, the authors propose a novel approach where the system response is derived by a consistent application of the Boltzmann principle to an equivalent system, built at every time instant based on the fractional order at that instant and the response at all the previous ones. The approach is readily implementable in numerical form to calculate either stress or strain responses of any fractional system where fractional order may change with time. Finally, Bologna *et al.* [55] propose a nonlinear extension of fractional calculus to tissue biomechanics, in order to handle the time-dependent mechanics of ligaments and tendons of the human knee. The authors point out that fibrous tissues exhibit a marked nonlinear behaviour in terms of relaxation and creep functions with coefficients and orders depending nonlinearly on applied strain and stress, respectively. On this basis, the authors show that, as nonlinearity is expressed by a power-law dependence on the applied stress, a modified version of fractional material hereditariness, namely the quasi-fractional material hereditariness, can be obtained with a nonlinear mapping of the state variables. As the mapping is established, specific relations among the parameters of creep and relaxation functions are obtained with a numerical assessment of the proposed formulation.

### 3. Heat conduction

Heat conduction is a typical phenomenon where time non-locality of fractional operators has been used to model memory effects. Various time-fractional heat conduction equations have been proposed, differing each other for the fractional operators involved [56–61]. In this framework, a thermodynamic model corresponding to power-law rise of temperature and heat flux has been proposed recently by Zingales [62].

Of particular interest in this context is the coupling between time-fractional heat transfer and material behaviour, which has led to the formulation of fractional thermoelasticity, fractional thermo-viscoelasticity and fractional electro-thermoelasticity [63–66]. For instance, a thermomechanical model with fractional-order heat equation has been developed, generalizing the Fourier diffusion with the introduction of a Caputo fractional operator [67,68].

A special interest exists also on thermal stresses which may arise at the vicinity of a crack as a result of thermal shocks. This is the problem investigated in the theme issue by Povstenko & Kyrylych [69], who solve the time-fractional heat conduction equation with the Caputo derivative for an infinite axisymmetric solid with a penny-shaped crack under a prescribed heat flux loading across its surfaces. Using Laplace, Hankel and cos-Fourier integral transforms, temperature field, thermal stress field and stress intensity factor are obtained in analytical form, namely in integral form involving the generalized two-parameter Mittag-Leffler function. The derived formulae allow a straightforward implementation of parametric analyses for different orders of the Caputo derivative. In the theme issue, a further contribution concerning heat conduction is given by Li & Cao [70], who show that fractional-order phonon Boltzmann transport equations, representing memory effects in phonon heat transport, lead to fractional heat conduction models capable of representing known anomalous heat diffusion, especially in low-dimensional systems. Additionally, the study highlights a non-trivial, fractional-order relationship between heat flux and entropy flux, as well as the contribution of initial effects to the entropy production rate.

### 4. Diffusion in porous media

Time and/or space non-locality of fractional operators is also suitable for addressing diffusion processes in various complex systems. In their seminal review, Metzler & Klafter [71] have discussed the relations between the fractional calculus approach and continuous time random walk theory and highlighted the advantages of fractional operators especially to describe diffusion phenomena involving boundary values or external velocity or force fields. Time-fractional operators are indeed ideally suitable to account for slow decay of initial conditions, slower dispersion and memory effects, which characterize anomalous sub-diffusion in complex systems as, for instance, proteins [72]. Time-fractional derivatives have been used to model reactive transport, since solutes may interact with the immobile porous medium in highly nonlinear ways; for instance, there is evidence that solutes may sorb for random amounts of time that have a power-law distribution, or move into irregularly sized blocks of relatively immobile water, producing similar behaviour [73]. Anomalous diffusion has been captured by a fractional-order generalization of the well-known Darcy equation for mass transport [74]. A further application of the fractional-order Darcy equation in the presence of flux across an elastic media has shown that an extended poro-elastic model is capable of capturing the faster, slower swelling/settlement of porous media in the presence of external applied load [75].

Beside the time-fractional models, non-local fractional operators in terms of spatial coordinates have been used to model diffusion phenomena [76] in porous media; for example, the effect of an infinite medium and its space interaction with the fluid has been represented by space-fractional derivatives, relating the flow to the pressure gradient experienced by the fluid in the path from the starting point to the measurement site [76]; furthermore, in the context of the flow in porous media, fractional-space derivatives have been used to model large motions through highly conductive layers or fractures [73].

Space–time-fractional equations have also been used to model anomalous diffusion, namely using a Riemann–Liouville time-fractional derivative and a Riesz–Feller space-fractional derivative [77]; space–time Caputo fractional derivatives have proved to fit experimental data on methanol transport through porous media, e.g. pelletized zeolite-based catalyst [78]; space–time-fractional differential equations have been investigated to model stochastic advection–diffusion problems in fractal media with long-range, correlated spatial fluctuations [79].

In the theme issue, Chugunov & Fomin [80] address the challenging issue of anomalous transport of contaminants within reservoirs. Focusing on the transport of radioactive materials in fractures surrounded by porous matrices of fractal structure, the authors propose a novel form of the fractional differential equation where fractional derivatives account for contaminant exchange between the fracture and the surrounding porous matrix; exact and approximate expressions for solute concentration in fracture and porous medium are obtained. Further, chloride diffusion in the reinforced concrete is the subject of the contribution by Chen *et al.* [81]. On observing that chloride ion penetration is generally slower than normal diffusion and exhibits anomalous characteristics as history dependence and long-range correlation, the authors propose a multi-term time-fractional model based on the Caputo fractional derivative and a pertinent numerical solution scheme, proving its convergence and stability. Next, the authors propose a modified grid approximation method to estimate the model parameters. Validation against real data from ordinary Portland cement and fly ash cement specimens, both exposed to chloride penetration over six different durations, prove that the proposed model is more accurate than alternative existing ones; in this context, a better fitting to experimental data is obtained using variable diffusion coefficients.

## 5. Non-local continua

Space-fractional operators have been used to formulate non-local continua, i.e. continua whose governing equations are endowed with appropriate non-local terms. There exist indeed several complex phenomena that cannot be addressed by classical local continua as, for instance, size effects in micro- and nanostructures resulting from non-local atomic or Van der Waals interactions or microstructure effects in elastic wave propagation through heterogeneous materials [82]; furthermore, space non-locality can also account for the effect of surrounding media that are not included in the model but produce a coupling between the responses of the model at different points. In this respect, space non-locality is definitely a relevant feature of fractional operators that, on the other hand, benefit of pertinent variational formulations ensuring consistency of the corresponding non-local governing equations.

Drapaca & Sivaloganatha [83] introduced a space-fractional continuum on introducing the concept of motion of order  $\alpha$  and deriving pertinent strain and stress tensors. Sumelka *et al.* [84] have proposed a space-fractional continuum formulation, using a Riesz–Caputo fractional derivative representing a non-local deformation gradient in small or finite strains, with applications to linear elasticity [84] and rate-independent plasticity [85]. Space-fractional operators have been used to model power-law long-range interactions between particles in  $n$ -dimensional lattice structures [86]; remarkably, on introducing a general form of lattice fractional derivatives and integrals, which revert to continuum Riesz fractional derivatives and integrals in the continuous limit, a bridge has been established by lattice structures and corresponding non-local continua [86,87]. A fractional generalization of the classical Eringen integral model of non-locality has been proposed by Carpinteri *et al.* [88] using a Riesz integral to represent a linearly elastic stress–strain integral relation. For the one-dimensional (1D) case, the authors derived a point-spring mechanical interpretation, with four sets of springs, one local and three non-local [88], which may be interpreted as describing long-range volume–volume and volume–surface interactions within the solid. A linearly elastic, mechanically based non-local fractional 1D continuum has been introduced by Di Paola and co-workers [89–91] based on the assumption that non-adjacent volumes mutually exert forces depending on relative displacements through distance-decaying power-law functions. Moving from this



assumption, Di Paola and co-workers obtained equilibrium equations involving the Marchaud fractional derivatives (their integral part only in bounded domains) and proved thermodynamic consistency [90]; furthermore, they revealed some inconsistencies in the fractional Eringen model for bounded domain, confirmed a previous non-local fractional 1D continuum introduced by Lazopoulos [92] under the assumption of vanishing boundary displacements and, finally, proposed pertinent numerical solution strategies of the governing equations [91]. The concept of power-law long-range interactions has been further developed in a fractional non-local Timoshenko beam theory by Alotta *et al.* [93], where non-adjacent beam elements mutually exchange moments and transverse forces decaying with power-law functions of distance along the beam axis; further applications have been found in two-dimensional (2D) foundation subgrade models and, in this case, non-local terms arise from the interaction between subgrade and foundation body [94]. A comprehensive fractional linear gradient elasticity theory has been proposed by Tarasov & Aifantis [95], which hinges on a three-dimensional Riesz fractional Laplacian modelling power-law, non-local constitutive behaviour; an alternative formulation of fractional linear gradient elasticity theory stemming from pertinent fractional variational principles has been presented by the same authors in a previous publication [96]. Applications of the formulations in [95,96] have been envisaged to address unusual phenomena in nanomaterials [97]. A fractional lattice approach has been proposed by Michelitsch *et al.* [98] for  $n$ -dimensional periodic and infinite lattices, introducing the concept of centred fractional-order difference operators as a generalization of the second-order centred difference operator appearing in the context of classical lattice models [98]; relations have been found between the fractional-order difference operators in the continuum limit and the classical Riesz fractional Laplacian derivative [98,99]. Non-local spatial fractional operators have been also used to model blood flow in capillary vessels, see [100,101], as well as long-range viscoelastic interactions [102,103].

In the theme issue, Patnaik & Semperlotti [104] propose a variable-order formulation to capture the evolution of edge dislocations through lattice structures under static shear stress. The authors simulate the microscopic structure of a material by a particle dynamic approach where constitutive atoms or molecules are represented via discrete masses and interparticle forces are represented by variable-order Riemann–Liouville operators depending on a quadratic potential field. Numerical results on 2D structure prove that such operators are intrinsically capable of capturing the complex linear-to-nonlinear dynamic transitions resulting from the translation of dislocations as well as the creation and annihilation of bonds between particles. Finally, the contribution to the theme issue of Zorica & Oparnica [105] focuses on time-fractional-wave equations modelling hereditary viscoelastic behaviour and space-fractional-wave equations associated with existing non-local elasticity models. For a number of fractional-wave equations, the authors provide rigorous mathematical evidence of energy dissipation and energy conservation. The authors provide also a comprehensive discussion on the physical meaning of the considered fractional-wave equations and their pertinence to real phenomena.

## 6. Fractal media

Certainly, an interesting and challenging topic is the application of fractional calculus concepts to describe the mechanics of fractal media. A space-fractional derivative approach to fractal media has been proposed in the early work of Carpinteri & Mainardi [106] and, on this basis, by Carpinteri *et al.* using the concept of local fractional derivative [107]. Later on, Tarasov [108] has proposed an alternative fractional-integral approach, where continuum models of fractal media are formulated using the fractional integration of non-integer order; Tarasov [109] also proved that fractional continuous models using fractional integrals can be used not only to describe a fractal medium with non-integer dimensions (e.g. mass dimension is found to be equal to the order of the fractional integral) but also to describe dynamical processes in the fractal medium and pertinent equations, as the equations of balance of mass density, momentum density, internal energy, Navier–Stokes and Euler equations, with applications to porous media. A gradient elasticity theory for fractal materials has been proposed by Tarasov & Aifantis [96]

starting from a fractional-integral formulation. A brief but illustrative review of continuum models of fractal media based on fractional calculus concepts may be found in [110]. A fractional-space approach to physical problems on fractals has been recently proposed by Balankin [111] based on the Stillinger's definition of space of fractional dimension [112]. Linear and nonlinear fractional viscoelastic models for fractal media have been derived from thermodynamic principles by Mashayekhi *et al.* [113] using fractal media representation as in [114]. Experimental validation was provided on a dielectric elastomer, whose a distinctive feature is a significant rate-dependent deformation during uniaxial stress measurements; using Bayesian statistics for calibration, the authors have shown that the fractional-order models are more accurate than integer-order ones for deformation rates spanning several orders of magnitude [113]. In a further recent work, Mashayekhi *et al.* [115] explored a physical connection between time-fractional derivative and fractal geometry of fractal media; on using thermodynamics law, the order of the fractional derivative in the linear fractional model of viscoelasticity was found to be a rate-dependent material property strongly correlated with fractal dimension and spectral dimension which characterizes diffusion in fractal media. The need for power-law functions to model phenomena in fractal media has been shown for the flux–time relations across fractal structures [116,117] as well as in the context of biomechanics of bone tissues [118].

In the theme issue, the mechanics of fractal media is addressed by Li & Ostoja-Starzewski [119] with focus on porous media. They propose a continuum model for anisotropic porous media of fractal type, which expresses the global balance laws in terms of fractal integrals based on proper product measures and converts them to integer-order integrals in conventional (Euclidean) space. Next, a continuum localization procedure leads to the local balance laws of the fractal medium as the conservation of mass, micro-inertia, linear and angular momenta, energy as well as to the pertinent second law of thermodynamics. Local balance laws involve the concept of fractal derivative or gradient. The relation between the proposed model and alternative fractional continua models of porous media is discussed throughout the paper. The proposed model has the potential of broadening the applicability of continuum mechanics/physics to the study of highly complex and fractal-type media (multiscale polycrystals, polymer clusters, gels, rocks, micro-cracked materials, percolating networks, nervous systems, and pulmonary systems); the authors present specific applications to model thermo-diffusion of liquid and gases in fractal poro-elastic media. Within the modelling of complex media, a challenging problem is the investigation of the dynamic response of media featuring random mass density with fractal and Hurst characteristics. This topic is addressed by Zhang & Ostoja-Starzewski in the theme issue [120], focusing on Lamb-type problems. The authors discuss the theoretical limitations in developing consistent space-fractional-derivative models of fractal media as well as in finding pertinent solutions and, in view of these outstanding challenges, propose an alternative approach simulating Cauchy and Dagum natural-like random fields by a Monte Carlo cellular automata approach; the latter has the advantage to assign cell-to-cell heterogeneous material properties and ensure equivalence to the continuum elasto-dynamics equations in the limit of infinitesimal cells. Wave propagation is investigated with focus on two Lamb-type problems on an elastic half-plane, specifically under a tangential point load and a concentrated point moment.

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