








ModeCouplingTheory.jl: A solver for mode-coupling-theory-like integro-differential equations

Ilian Pihlajamaa ¹, Corentin C. L. Laudicina ¹, Thomas Voigtmann ^{2,3}, and Liesbeth M. C. Janssen ¹✉

¹ Soft Matter and Biological Physics, Department of Applied Physics, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, Netherlands ² Institut für Materialphysik im Weltraum, Deutsches Zentrum für Luft- und Raumfahrt (DLR), Köln, 51170, Germany ³ Heinrich Heine University, Universitätsstraße 1, Düsseldorf, 40225, Germany ✉ Corresponding author

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Summary

The mode-coupling theory of the glass transition is one of the most successful theories for predicting the dynamics of dense liquids to date. In the regime where it is applicable, the theory gives a set of detailed numerical and analytical predictions for the structural relaxation dynamics upon supercooling. In particular, the theory provides closed equations for dynamic correlation functions of the microscopic density field, including observables such as the coherent and incoherent intermediate scattering functions and the mean-squared displacement. These mode-coupling theory equations take the form of an integro-differential equation, i.e., a generalized Langevin equation, the kernel of which represents the coupling between different “relaxation modes”. Because these equations are difficult to solve numerically due to their non-linearity and the long-livedness of the solutions, specialized algorithms have been developed to tackle this issue. ModeCouplingTheory.jl is a package that implements such an algorithm, including a number of convenient features that make it simple to solve the complex equations involved even for those not well-versed in the theoretical and numerical background traditionally required.

Statement of need

The mode-coupling theory of the glass transition (MCT) is at the forefront of the study of the glass transition (Ulf Bengtzelius et al., 1984; Das, 2004; Götze, 2009; Janssen, 2018; Leutheusser, 1984; Reichman & Charbonneau, 2005) and has been applied to an abundance of different systems and scenarios in the past forty years. See for example versions of MCT including multiple particle species (Franosch & Voigtmann, 2002; Götze & Voigtmann, 2003; Luo et al., 2022; Weysser et al., 2010), confinement (Krakoviack, 2007; Lang et al., 2010), self-propelling particles (Berthier & Kurchan, 2013; Debets & Janssen, 2023; Feng & Hou, 2017; Liluashvili et al., 2017; Reichert et al., 2021; Szamel, 2016), high dimensionalities (Ikeda & Miyazaki, 2010; Schmid & Schilling, 2010), and molecular particles (Chong et al., 2000; Chong & Hirata, 1998; Schilling & Scheidsteger, 1997; Theis et al., 2000; Winkler et al., 2000) to name a few. Before the conception of the software this paper is based on, there was no open-source integrator for MCT-like equations available. This meant that each time an improvement, extension or application to the theory was employed, a new in-house integrator needed to be developed or adapted. ModeCouplingTheory.jl aims at providing an open-source and tested implementation of the scheme introduced by Fuchs et al. (1991), that is performant and easy to extend to new systems, yet simple to call interactively from dynamic languages

such as Python. As of writing, it has been used for several scientific works ([Kerr Winter et al., 2023](#); [Laudicina et al., 2022, 2023](#); [Pihlajamaa et al., 2023](#)).

The main equation that this package aims to solve is of the form

$$\alpha\ddot{F}(t) + \beta\dot{F}(t) + \gamma F(t) + \delta + \int_0^t d\tau K(t - \tau)\dot{F}(\tau) = 0$$

in which α , β , γ , and δ are coefficients (possibly dependent on time), $K(t) = K(F(t), t)$ is the memory kernel, and $F(t)$ is the function that is being solved for. Memory kernels for solving the most commonly encountered models, such as standard (multi-component) MCT, including tagged-particle correlators, and mean squared displacements, are implemented, and it is straightforward to implement custom memory kernels. Internally, the software completely separates the memory kernel, the type of equation, and the solver, each of which can be independently extended by a user.

The documentation details the features of this software, which among others include

1. **Generality:** the code was developed with generality in mind. For example, the code works for types between which product operation is defined among α , β , γ , K on the left and F on the right, returning something of the same type as F and δ . This implies that the code works for functions F that are scalar valued (schematic models), as well as those that are vectors of floating point numbers (standard MCT), and vectors with elements of different types. The latter could include, for example, numbers with measurement errors, dual numbers, and immutable matrices.
2. **Extensibility:** the solvers are easily extended to deal with coupled sets of equations that arise, e.g., in extensions of MCT to describe tagged-particle dynamics, or in certain asymptotic models.
3. **Speed:** the code is developed for performance. The solver allocates little memory and uses BLAS implementations for linear algebra where applicable ([Lawson et al., 1979](#)). The memory kernels of the single component and multi-component MCT as well as their tagged variants are implemented using Bengtzelius' trick, yielding algorithmic speed-up compared to more naive implementations ([U. Bengtzelius, 1986](#)).
4. **Ease of use:** solving the equations of standard MCT takes very few lines of code, see the example below. While written in Julia, the code can straightforwardly be called from Python and other languages.
5. **Measurement errors:** by leveraging the generality of the software it is straightforward to do standard error propagation through the solver.
6. **Non-ergodicity parameters:** there is built-in functionality for finding the long-time limits of the solution of the MCT equations.

Example Use

To solve the standard MCT equations in three dimensions for hard spheres using the Percus-Yevick structure factor ([Wertheim, 1963](#)), one may run the following code. See the documentation for a more in-depth explanation.

```
using ModeCouplingTheory
# the wave vector grid
Nk = 100; kmax = 40.0; dk = kmax/Nk; k = range(dk/2, kmax-dk/2, length=Nk)

# physical parameters
kBT = 1.0; m = 1.0; ρ = 0.983
```

```

# Hard-Sphere Percus-Yevick structure factor for this density
A = 5688.95; B = 2183.01; C = 661.463; D = 66.0759; E = 314.311;
Sk = @. k^6 /
    (A + B*k^2 + k^6 - (A - C*k^2 + D*k^4)*cos(k) - (A + E*k^2)*k*sin(k))

# initial conditions and coefficients
F0 = Sk; ∂F0 = zeros(Nk)
α = 0.0; β = 1.0; γ = @. k^2*kBT/(m*Sk); δ = 0.0

# construct the equation and solve it
kernel = ModeCouplingKernel(ρ, kBT, m, k, Sk)
equation = MemoryEquation(α, β, γ, δ, F0, ∂F0, kernel)
sol = solve(equation)

# plot the solution for several values of k
using Plots
p = plot(xlabel="log10(t)", ylabel="F(k,t)/S(k)",
        ylims=(0,1), xlims=(-6, 6))
for ik = [7, 18, 25, 39]
    t = get_t(sol)
    Fk = get_F(sol, :, ik)
    plot!(p, log10.(t), Fk/Sk[ik], label="k = $(k[ik])", lw=3)
end
display(p)

```

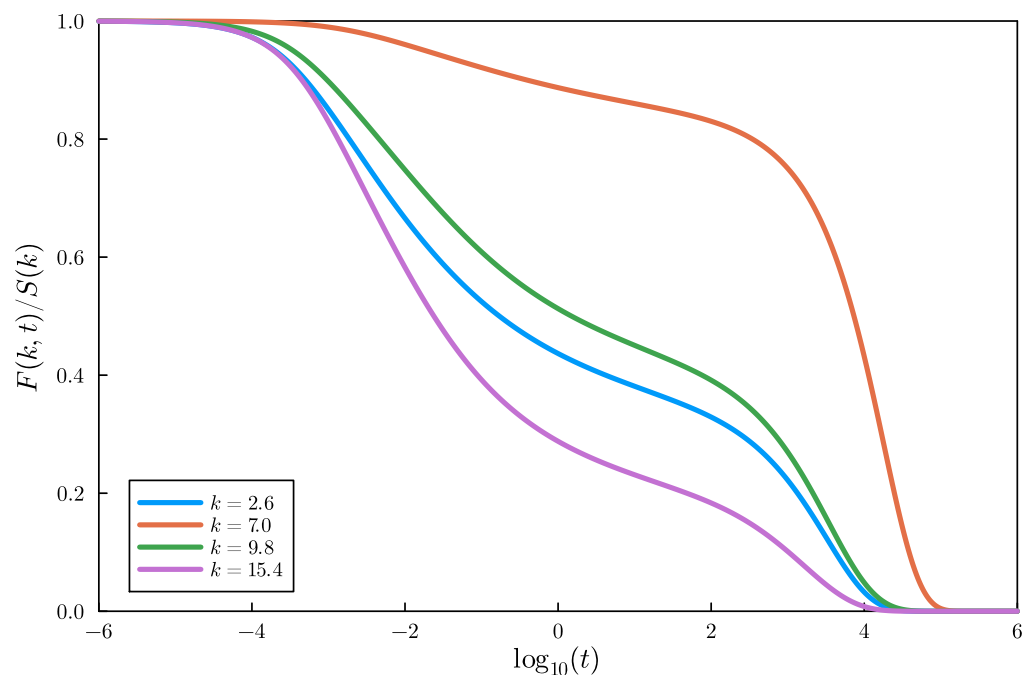


Figure 1: The code above yields this figure, which shows the intermediate scattering function, obtained with MCT, as a function of time for different values of k .

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