Research Article

Rudolf Hilfer*

Mathematical analysis of time flow

Abstract: The mathematical analysis of time flow in physical many-body systems leads to the study of long-time limits. This article discusses the interdisciplinary problem of local stationarity, how stationary solutions can remain slowly time dependent after a long-time limit. A mathematical definition of almost invariant and nearly indistinguishable states on C*-algebras is introduced using functions of bounded mean oscillation. Rescaling of time yields generalized time flows of almost invariant and macroscopically indistinguishable states, that are mathematically related to stable convolution semigroups and fractional calculus. The infinitesimal generator is a fractional derivative of order less than or equal to unity. Applications of the analysis are given to irreversibility and to a physical experiment.

Keywords: Time evolution, local stationarity, semigroups, C*-algebras, fractional time derivative, irreversibility, many-body system, coarse graining, separation of scales, excess wing, dielectric relaxation, glassy dynamics

MSC 2010: Primary: 37L05; secondary: 82D30, 81T27, 46L30, 46L40, 47L90

DOI: 10.1515/anly-2015-5005
Received July 4, 2015; accepted August 4, 2015

Dedicated to the memory of Anatoly Kilbas

1 Introduction

A time evolution for closed quantum systems is usually specified as a one-parameter group of unitary operators on a Hilbert space. Dissipative processes, diffusion or irreversible ageing of natural systems are difficult to accommodate within this theoretical framework.

Many equations of motion for closed systems are formulated as abstract Cauchy problems on a Banach space $\mathfrak{B}$. A state or observable $A \in \mathfrak{B}$ evolves according to

$$\tau\epsilon \frac{\mathrm{d}}{\mathrm{d}t} A(t/\tau) = \mathscr{L} A(t/\tau), \tag{1.1a}$$

$$A(t_0/\tau) = A_{0,\tau} \tag{1.1b}$$

from its initial value $A_{0,\tau}$ at time instant t_0. In (1.1) time is measured in units of τ seconds (such that $t/\tau \in \mathbb{R}$). and ϵ is an energy scale (Joule). Often the infinitesimal generator $\mathscr{L}$ (Liouvillian or Hamiltonian) is an unbounded operator with domain $D(\mathscr{L}) \subset \mathfrak{B}$ (see [16]). Recall that $\mathscr{L}$ corresponds to a vector field in classical mechanics, to a selfadjoint operator in quantum mechanics, and to a derivation on an algebra of observables in field theories [5]. Existence of a physical time evolution is equivalent to the existence of global solutions of (1.1) under various assumptions. Many examples of (1.1) do not have global solutions, particularly when the physical system is infinite.

Define $\mathfrak{B} = \mathfrak{A}$ to be the C*-algebra of observables of a physical system, and assume that it has an identity, unless explicitly stated otherwise. Equations (1.1) give formally

$$\mathscr{T}^{t/\tau} \mathcal{K}_{A_{0,\tau}}\left(\frac{t_0}{\tau}\right) = T^{t/\tau} A_{0,\tau} \tag{1.2}$$

***Corresponding author: Rudolf Hilfer:** Insitut für Computerphysik, Universität Stuttgart, 70569 Stuttgart, Germany

if the maps $T^s : \mathfrak{A} \to \mathfrak{A}$ and $\mathscr{T}^s : \mathfrak{A} \to \mathfrak{A}$ are defined as

$$T^{t/\tau} A = \exp\left(\frac{\mathscr{L} t}{\epsilon\tau}\right) A, \tag{1.3a}$$

$$\mathscr{T}^{t/\tau} \mathcal{K}_A(s) = \mathcal{K}_A\left(s + \frac{t}{\tau}\right), \tag{1.3b}$$

and the orbit maps $\mathcal{K}_A : \mathbb{R} \to \mathfrak{A}$ are defined as

$$\mathcal{K}_A(s) : s \mapsto T^s A$$

for each fixed $A \in \mathfrak{A}$. In (1.3), where T^s with $s \in \mathbb{R}$ is a one-parameter family of *-automorphisms of $\mathfrak{A}$, it remains to give meaning to the formal exponential, such that the orbit maps $\mathcal{K}_A : \mathbb{R} \to \mathfrak{A}$ are continuous for every $A \in \mathfrak{A}$.

Given the *-automorphisms $(T^s)_{s\in\mathbb{R}}$ this family of operators is expected to fulfill the time evolution law

$$T^{t/\tau} T^{s/\tau} = T^{(t+s)/\tau} \tag{1.4}$$

with $T^0 = \mathbf{1}$ being the identity. Let the maps $t \mapsto T^t$ be continuous as mappings from $\mathbb{R}$ into the space $\mathfrak{B}(\mathfrak{A})$ of all bounded operators on $\mathfrak{A}$ endowed with the strong operator topology [34, 60]. One then has continuous orbits $\mathcal{K}_A(s)$ and the operator family $(T^s)_{s\in\mathbb{R}}$ is then a strongly continuous one-parameter group (C_0-group) on $\mathfrak{A}$. Recall that physical experiments do not observe or measure the selfadjoint observables A directly, but only their expectation values, denoted as $\langle \mathsf{z}, A\rangle \in \mathbb{R}$, with respect to a state z of the physical system. Indeed, states are generally defined as positive and normalized linear functionals on the algebra $\mathfrak{A}$ of observables [5]. A linear functional $\mathsf{z} : \mathfrak{A} \to \mathbb{R}$ is called positive, if $\langle \mathsf{z}, A^*A\rangle \geq 0$ holds for all $A \in \mathfrak{A}$, and normalized if $\|\mathsf{z}\| = \sup\{|\langle \mathsf{z}, A\rangle|, \|A\| = 1\} = 1$. Most of the discussion below concerns states and it is necessary to define their time evolution.

States are elements of the topological dual $\mathfrak{A}^* = \{\mathsf{z} : \mathfrak{A} \to \mathbb{C} : \mathsf{z} \text{ is linear and continuous}\}$. The notation $\langle \mathsf{z}, A\rangle$ is used for the value $\mathsf{z}(A) \in \mathbb{R}$ of a selfadjoint $A \in \mathfrak{A}$ in the state z. The adjoint time evolution $T^{*t} : \mathfrak{A}^* \to \mathfrak{A}^*$ with $t \in \mathbb{R}$ consists of all adjoint operators $(T^t)^*$ on the dual space $\mathfrak{A}^*$ (see [50, 51]). Let $\mathsf{Z} \subset \mathfrak{A}^*$ denote the set of all states. The orbit maps for states $\mathcal{K}_{\mathsf{z}} : \mathbb{R} \to \mathsf{Z}$ are defined as

$$\mathcal{K}_{\mathsf{z}}(s) : s \mapsto (T^s)^* \mathsf{z} = T^{*s} \mathsf{z}$$

for states $\mathsf{z} \in \mathsf{Z} \subset \mathfrak{A}^*$. If T^t is strongly continuous then

$$|\langle (T^{*t} - \mathbf{1})\mathsf{z}, A\rangle| = |\langle \mathsf{z}, T^t A - A\rangle| \leq \|\mathsf{z}\|\, \|T^t A - A\|$$

shows that the adjoint time evolution T^{*t} is weak*-continuous in the sense that the maps $\langle A\rangle_{\mathsf{z}} : \mathbb{R} \to \mathbb{R}$,

$$t \mapsto \langle A\rangle_{\mathsf{z}}(t) = \langle \mathsf{z}, T^t A\rangle = \langle T^{*t}\mathsf{z}, A\rangle \tag{1.5}$$

are continuous for all $A \in \mathfrak{A}$, $\mathsf{z} \in \mathsf{Z}$. These maps are the time evolutions of all expectation values. In other words, for a C_0-group $(T^s)_{s\in\mathbb{R}}$ the orbit maps $\mathcal{K}_{\mathsf{z}}(s)$ are continuous from $\mathbb{R}$ into the space $\mathfrak{B}(\mathfrak{A}^*)$ of all bounded operators on $\mathfrak{A}^*$ endowed with the weak*-topology [34, 53] and the adjoint family $(T^{*s})_{s\in\mathbb{R}}$ is a C_0^*-group. Note that the adjoint time evolution T^{*t} need not be strongly continuous unless $\mathfrak{A}$ is reflexive. The relation between the time evolution of states and observables is

$$\begin{aligned}
\langle \mathcal{K}_{\mathsf{z}}(t_0), T^t \mathcal{K}_A(t_0)\rangle &= \langle \mathcal{K}_{\mathsf{z}}(t_0), \mathscr{T}^t \mathcal{K}_A(t_0)\rangle \\
&= \langle \mathcal{K}_{\mathsf{z}}(t_0), \mathcal{K}_A(t_0 + t)\rangle \\
&= \langle \mathcal{K}_{\mathsf{z}}(t_1 - t), \mathcal{K}_A(t_1)\rangle \\
&= \langle \mathscr{T}^{-t} \mathcal{K}_{\mathsf{z}}(t_1), \mathcal{K}_A(t_1)\rangle \\
&= \langle T^{*t} \mathcal{K}_{\mathsf{z}}(t_1), \mathcal{K}_A(t_1)\rangle,
\end{aligned} \tag{1.6}$$

where $t_1 = t_0 + t \in \mathbb{R}$. The adjoint time evolution of states is related to right translations along the orbits in state space in the same way as the time evolution of observables is related to left translations along orbits in the algebra.

Equation (1.1a) combined with (1.6) for the adjoint time evolution states formally the proportionality

$$\pm\tau\frac{\mathrm{d}}{\mathrm{d}t} = \pm\frac{\mathscr{L}}{\epsilon}$$

of the infinitesimal generator $\mathrm{d}/\mathrm{d}t$ of time translations and the infinitesimal generator $\mathscr{L}$ of changes of the physical system. Independently of the manner in which one attaches a meaning to the formal exponential, equation (1.2) says that the time evolution of a physical system is a translation along orbits corresponding to the changes of the system, specifically

$$\begin{aligned}(\text{left shift along } \mathfrak{A}\text{-orbit}) &= (\text{change of observable}),\\ (\text{right shift along } \mathsf{Z}\text{-orbit}) &= (\text{change of state}),\end{aligned}$$

where the first equation reflects the Heisenberg picture, while the second corresponds to the Schrödinger picture.

2 Problems and objective

It has been recently suggested that several unsolved and superficially unrelated problems in open quantum systems, classical mechanics and quantum field theory are related to two fundamental questions associated with the theoretical framework described in the introduction [31].

Problem 1. Are there global solutions of problem (1.1), i.e. solutions for all $t \in \mathbb{R}$?

Problem 2. If global solutions of problem (1.1) exist, how can invariant solutions still change with time?

Whenever thermodynamic observables such as temperature, pressure or densities change slowly with time, it is necessary to analyze the mathematical transition from local (short time) stationarity to global (long time) instationarity. The same mathematical transition is needed for the discussion of the relation between different representations of canonical (anti-)commutation relations [16].

The two problems require to analyze the set of states that are invariant under the time evolution. If the thermodynamic observables change, then there must exist many invariant states, and "long"-time averages cannot be unique. The objective of this paper is to introduce a mathematical framework for the analysis of such questions. Such a framework has remained elusive [56]. The main results are stated as two mathematical theorems. The second objective of this paper is to apply the results of the mathematical analysis to a physical experiment.

3 Invariance and indistinguishability

Definition 1. A state $\mathsf{z} \in \mathfrak{A}^*$ is called invariant, if

$$\langle \mathsf{z}, T^t A\rangle = \langle \mathsf{z}, A\rangle \tag{3.1}$$

holds for all $A \in \mathfrak{A}$ and $t \in \mathbb{R}$.

The expectation values $\langle A\rangle_{\mathsf{z}}(t) = \langle \mathsf{z}, A\rangle$ of all observables $A \in \mathfrak{A}$ are then constant. The set of invariant states $\mathsf{B}_0 \subset \mathfrak{A}^*$ over $\mathfrak{A}$ is convex and compact in the weak*-topology [5]. The same holds for the set of all states $\mathsf{Z} \supset \mathsf{B}_0$. Invariant states are fixed points of the adjoint time evolution T^{*t} as seen from (1.5).

Definition 2. A state $\mathsf{z} \in \mathsf{Z}$ is called a BMO-state if all maps $\langle A\rangle_{\mathsf{z}} : \mathbb{R} \to \mathbb{R}$ are in $\mathrm{BMO}(\mathbb{R})$ for all $A \in \mathfrak{A}$. The Banach space $\mathrm{BMO}(\mathbb{R})$ of functions with bounded mean oscillation on $\mathbb{R}$ is defined as the linear space

$$\mathrm{BMO}(\mathbb{R}) = \{f \in L^1_{\mathrm{loc}}(\mathbb{R}), \|f\|_{\mathrm{BMO}} < \infty\},$$

where $L^1_{\text{loc}}(\mathbb{R})$ is the space of locally integrable functions $f : \mathbb{R} \to \mathbb{R}$. The BMO-norm is defined as

$$\|f\|_{\text{BMO}} = \inf_C \left\{ \int_I |f(x) - f_I| \, \mathrm{d}x \le C|I|, \text{ for all } I \right\},$$

where $I \subset \mathbb{R}$ denotes intervals of length $|I|$ and

$$f_I = \frac{1}{|I|} \int_I f(x) \, \mathrm{d}x$$

denotes the average of f over the interval I.

The set of all BMO-states

$$\mathsf{B} = \{\mathsf{z} \in \mathsf{Z} : \|\langle A \rangle_\mathsf{z}\|_{\text{BMO}} < \infty \text{ for all } A \in \mathfrak{A}\}$$

is convex by linearity. Because $\mathsf{B} \subset \mathsf{Z}$ is a subset of a weak*-compact set, it is also weak*-compact. Hence a decomposition theory into extremal BMO-states exists by virtue of the Krein–Milman theorem [5]. The set of invariant states is denoted by

$$\mathsf{B}_0 = \{\mathsf{z} \in \mathsf{B} : \|\langle A \rangle_\mathsf{z}\|_{\text{BMO}} = 0 \text{ for all } A \in \mathfrak{A}\},$$

and it is a subset $\mathsf{B}_0 \subset \mathsf{B}$. BMO-states allow to define the concept of almost invariance. A BMO-state will be called ε-almost invariant, or almost invariant with accuracy ε, if the expectation values of all observables are stationary to within an accuracy ε.

Definition 3. The set B_ε of all ε-almost invariant states is defined as

$$\mathsf{B}_\varepsilon = \{\mathsf{z} \in \mathsf{B} : \|\langle A \rangle_\mathsf{z}\|_{\text{BMO}} < \varepsilon \text{ for all } A \in \mathfrak{A}\}.$$

The one-parameter family B_ε of ε-almost invariant states is a family of subsets of B. The accuracy ε measures temporal fluctuations away from the time average.

The following inclusions of classes of states used in the following are summarized for orientation and convenience:

$$\mathsf{K}_\beta \subset \mathsf{B}_0 \subset \mathsf{B}_\varepsilon \subset \mathsf{B}_\infty = \mathsf{B} \subset \mathsf{Z} \subset \mathfrak{A}^*,$$

where $0 < \varepsilon < \infty$ and the sets of KMS-states K_β at inverse temperature $\beta > 0$ are defined as states $\mathsf{z} \in \mathsf{Z}$ such that the KMS-condition [6]

$$\langle \mathsf{z}, T^{t/\tau}(A)B \rangle = \langle \mathsf{z}, BT^{t/\tau + \mathrm{i}\epsilon\beta}(A) \rangle$$

holds for all $t/\tau \in \mathbb{R}$ and $A, B \in \mathfrak{A}$. The KMS-states are invariant states for all $\beta \ge 0$, but KMS-states for infinite volume systems at different β are often disjoint [6]. For $\beta = 0$ the KMS-states are trace states, that are defined by $\langle \mathsf{z}, AB \rangle = \langle \mathsf{z}, BA \rangle$ for all $A, B \in \mathfrak{A}$. Because KMS-states are Gibbs states, they are usually interpreted as equilibrium states. Extremal states correspond to pure thermodynamic phases [6].

Two states are called indistinguishable, if they cannot be distinguished by measurements. Let $m < \infty$ denote the maximal number of experiments that can be performed to distinguish the states of the system. Let $\{A_i\}_{i=1}^m \subset \mathfrak{A}$ denote the observables in these experiments, and let η_i $(i = 1, \ldots, m)$ be the accuracies, that can be attained for measuring A_i.

Definition 4. Two states $\mathsf{z}, \mathsf{z}' \in \mathsf{Z}$ with

$$|\langle \mathsf{z}, A_i \rangle - \langle \mathsf{z}', A_i \rangle| = |\langle \mathsf{z} - \mathsf{z}', A_i \rangle| < \eta_i \le \eta = \max_{i=1,\ldots,m} \eta_i$$

for all $i = 1, \ldots, m$ are called (experimentally) indistinguishable with respect to the observables $A_1, \ldots, A_m$.

The sets of indistinguishable states

$$\mathsf{N}(\mathsf{z}; \{A_i\}_{i=1}^m; \eta) = \{\mathsf{z}' \in \mathfrak{A}^* : |\langle \mathsf{z} - \mathsf{z}', A_i \rangle| < \eta_i, \ i = 1, \ldots, m\}$$

are η-neighborhoods of z in the weak*-topology [17]. The algebra $\mathfrak{M}$ generated by the elements $A_1, \dots, A_m \in \mathfrak{A}$ will be called macroscopic algebra.

In the following, $0 < \eta_i < \infty$ and $0 < \eta = \max_i \eta_i < \infty$ will be assumed. The η-neighborhoods of ε-almost invariant states $\mathsf{N}(\mathsf{z}; \{A_i\}_{i=1}^m, \eta) \cap \mathsf{B}_\varepsilon$ with $\mathsf{z} \in \mathsf{B}_0$ for small $\varepsilon, \eta \to 0$ will be the candidates for local (in time) stationary states.

4 Representation dependent invariant measures on BMO-states

The set of BMO-states B is weak*-compact. Its open subsets are the elements of the weak*-topology restricted to B. They generate the σ-algebra $\mathcal{B}$ of Borel sets on B. Let $\mathsf{z} \in \mathsf{B}_0 \subset \mathsf{B}$ denote an invariant state so that (3.1) holds for all $t \in \mathbb{R}$, $A \in \mathfrak{A}$. An invariant probability measure on B corresponding to the invariant state z is constructed using the GNS-construction [5] and a resolution of the identity on $\mathcal{B}$. The GNS-representation $(\mathfrak{H}_\mathsf{z}, \pi_\mathsf{z}, \Omega_\mathsf{z}, U_\mathsf{z}^t)$ is the cyclic representation canonically associated with an invariant state $\mathsf{z} \in \mathsf{B}$ and the time evolution T^t on the C*-algebra $\mathfrak{A}$. It is uniquely determined by the two requirements

$$\begin{aligned} U_\mathsf{z}^t\, \pi_\mathsf{z}(A)\, U_\mathsf{z}^{-t} &= \pi_\mathsf{z}(T^t A) \quad \text{for } A \in \mathfrak{A},\ t \in \mathbb{R}, \\ U_\mathsf{z}^t\, \Omega_\mathsf{z} &= \Omega_\mathsf{z} \qquad\quad \text{for } t \in \mathbb{R}. \end{aligned}$$

The scalar product in $\mathfrak{H}_\mathsf{z}$ will be denoted as $(\ ,\ )$. A resolution of the identity [53, p. 301] on the Borel σ-algebra $\mathcal{B}$ is a mapping $P : \mathcal{B} \to \mathfrak{B}(\mathfrak{H}_\mathsf{z})$ with the following properties.

1. $P(\emptyset) = 0$, $P(\mathsf{B}) = \mathbf{1}$.
2. Each $P(\mathsf{G})$ is a self-adjoint projector.
3. $P(\mathsf{G} \cap \mathsf{G}') = P(\mathsf{G})P(\mathsf{G}')$.
4. If $\mathsf{G} \cap \mathsf{G}' = \emptyset$ then $P(\mathsf{G} \cup \mathsf{G}') = P(\mathsf{G}) + P(\mathsf{G}')$.
5. For every $\psi \in \mathfrak{H}_\mathsf{z}$ and $\phi \in \mathfrak{H}_\mathsf{z}$ the set function $P_{\psi,\phi} : \mathcal{B} \to \mathbb{C}$ defined by

$$P_{\psi,\phi}(\mathsf{G}) = \big(P(\mathsf{G})\psi, \phi\big)$$

is a complex regular Borel measure on $\mathcal{B}$.

Because the projectors are self-adjoint, the set function $P_{\psi,\psi}$ is a positive measure for every $\psi \in \mathfrak{H}_\mathsf{z}$. For $\psi = \phi = \Omega_\mathsf{z}$ the resulting measure

$$P_{\Omega_\mathsf{z},\Omega_\mathsf{z}} = \big(P(\mathsf{G})\Omega_\mathsf{z}, \Omega_\mathsf{z}\big) =: P_\mathsf{z}$$

is an invariant probability measure on the measurable space $(\mathsf{B}, \mathcal{B})$ associated with the invariant BMO-state $\mathsf{z} \in \mathsf{B}$. The triple $(\mathsf{B}, \mathcal{B}, P_\mathsf{z})$ is a probability space. The probability measure P_z is invariant under the adjoint time evolution T^{*t} on B.

5 Long time limit

This section solves Problem 2. It uses ε-almost invariant and η-indistinguishable states to do that. The result implies global existence of a rescaled time evolution on an algebra $\mathfrak{M} \subset \mathfrak{A}$ of macroscopic observables.

Let $\mathsf{S} \subset \mathsf{B}_0$ be a subset of (strictly) invariant states. An example could be KMS-states within some temperature interval $\bigcup_{\beta \in [\beta_1 \beta_2]} \mathsf{K}_\beta$. Define a weak*-neighborhood

$$\mathsf{G} = \mathsf{B}_\varepsilon \cap \Big(\bigcup_{\mathsf{z} \in \mathsf{S}} \mathsf{N}(\mathsf{z}, \{A_i\}_{i=1}^m; \eta) \Big) = \mathsf{G}(\mathsf{S}, \mathfrak{M}, \varepsilon, \eta) \tag{5.1}$$

of ε-almost invariant η-indistinguishable states near S. The time translations $\mathscr{T}^{-t/\tau}$ with time scale τ translate any initial state $\mathsf{z} \in \mathsf{G}$ along its orbit according to

$$\mathscr{T}^{-t/\tau} \mathcal{K}_\mathsf{z}\Big(\frac{t_0}{\tau}\Big) = \mathcal{K}_\mathsf{z}\Big(\frac{t_0 - t}{\tau}\Big),$$

where t_0 denotes the initial instant, $\mathcal{K}_z(t_0/\tau) = z$ and $\tau > 0$ the time scale. Discretizing time as

$$t = k\tau$$

with $k \in \mathbb{Z}$ such that $t_0 = 0$ produces discretized orbits $\mathcal{K}_z(-k)$, $k \in \mathbb{N}$ for all $z \in G$ as iterates of $\mathcal{T}^1$. For every initial state $z \in G$ define

$$w_G(z) = \min\{k \geq 1 : \mathcal{T}^{-k}\mathcal{K}_z(0) \in G\}$$

as the first return time of z into the set G. For all invariant $z \in B_0$ one has $w_G(z) = 1$. For states z that never return to G one sets $w_G(z) = \infty$. For all $k \geq 1$ let

$$G_k = \{z \in G : w_G(z) = k\}$$

denote the subset of states with recurrence time $1 \leq k \leq \infty$ with $k = \infty$ interpreted as

$$G_\infty = G \setminus \bigcup_{k\in\mathbb{N}} G_k.$$

The states $z \in S$ generate a family of resolutions of the identity P_z resulting in a family of invariant measures on $(B, \mathcal{B})$. Their mixture with a measure $\nu(z)$ on S,

$$Q = \int\limits_S P_z \, d\nu(z), \tag{5.2}$$

is again an invariant measure on $(B, \mathcal{B})$. The numbers

$$p(k) = \frac{Q(G_k)}{Q(G)} \tag{5.3}$$

define a discrete probability density on $\mathbb{N} \cup \{\infty\}$. It may be interpreted as a weighted probability of recurrence into the neighborhood G of the set $S \subset B_0$.

The time evolution of almost invariant states can be defined by addition of random recurrence times. Let $p_N(k)$ be the probability density of the sum

$$W_N = w_1 + \cdots + w_N$$

of $N \geq 1$ independent and identically distributed random recurrence times $w_i \geq 1$. Let $p(k)$ from (5.3) be the common probability density of all w_i. Then, with $N \geq 2$ and $p_1(k) = p(k)$,

$$p_N(k) = (p_{N-1} * p)(k) = \sum_{m=0}^{k} p_{N-1}(m)p(k-m) \tag{5.4}$$

is an N-fold convolution of the discrete recurrence time density in (5.3). The family of distributions $p_N(k)$ obeys

$$p_N(\infty) + \sum_{k=1}^{\infty} p_N(k) = 1$$

for all $N \geq 1$ and the discrete analogue of (1.4)

$$p_{N+M}(k) = (p_N * p_M)(k)$$

holds for all $N, M \geq 1$. The individual states in G are indistinguishable within the given accuracy η, but each of them may have a different first recurrence time. It is then natural to assign the average

$$\mathcal{S}^{-1} = \sum_{k=1}^{\infty} p(k)\, \mathcal{T}^{-k}$$

over all recurrence times as the duration of a single time step in the induced time evolution on G. If a macroscopic time evolution with a rescaled time exists then this requires to consider the iterations

$$\mathcal{S}^{-N} = \mathcal{S}^{-(N-1)}\mathcal{S}^{-1} = \sum_{k=1}^{\infty} p_N(k)\, \mathcal{T}^{-k} \tag{5.5}$$

in the limit $N \to \infty$. These may be interpreted as the age of the indistinguishable states in G after N iterations or time steps. The long-time or large N limit of $\mathscr{S}^N$ is governed by local limit theorems for convolutions [3, 11, 14, 20, 21, 35].

Theorem 1. *Let $p_N(k)$ be the probability density of random recurrence times specified in* (5.4). *Then there exist constants $D_N \geq 0$, $D \geq 0$ and $0 < \alpha \leq 1$ such that*

$$\lim_{N\to\infty} \sup_k \left| D_N p_N(k) - \frac{\tau}{D^{1/\alpha}} h_\alpha\left(\frac{k\tau}{D_N D^{1/\alpha}}\right)\right| = 0, \tag{5.6}$$

where

$$\alpha = \sup\left\{0 < \beta < 1 : \sum_{k=1}^{\infty} k^\beta p(k) < \infty\right\} \tag{5.7}$$

if $\sum_{k=1}^{\infty} kp(k)$ diverges, while

$$\alpha = 1,$$

if $\sum_{k=1}^{\infty} kp(k)$ converges. For $\alpha = 1$ the function $h_\alpha(x)$ is $h_1(x) = \delta(x-1)$. For $0 < \alpha < 1$ the function $h_\alpha(x)$ is

$$h_\alpha(x) = \begin{cases} 0 & \textit{for } x \leq 0, \\ \dfrac{1}{x}\displaystyle\sum_{j=0}^{\infty} \frac{(-1)^j x^{-\alpha j}}{j!\,\Gamma(-\alpha j)} & \textit{for } x > 0. \end{cases}$$

Proof. Existence of a limiting distribution for $W_N/D_N > 0$ is known to be equivalent to stability of the limit [14]. If the limit distribution is nondegenerate, this implies that the rescaling constants D_N have the form

$$D_N = (N\Lambda(N))^{1/\alpha}, \tag{5.8}$$

where $\Lambda(N)$ is a slowly varying function [55], defined by the requirement that

$$\lim_{x\to\infty} \frac{\Lambda(bx)}{\Lambda(x)} = 1$$

holds for all $b > 0$. That the number α obeys (5.7) is proven in [14, p. 179]. It is bounded as $0 < \alpha \leq 1$ because the rescaled random variables $W_N/D_N > 0$ are positive.

To prove (5.6) note that the characteristic function of W_N is the N-th power

$$\langle \mathrm{e}^{\mathrm{i}\xi W_N}\rangle = [p(\xi)]^N = \sum_k \mathrm{e}^{\mathrm{i}\xi k} p_N(k)$$

because the characteristic functions $p(\xi) = \langle \mathrm{e}^{\mathrm{i}\xi w_j}\rangle$ of w_j are identical for all $j = 1, \ldots, N$. Inverse Fourier transformation gives

$$p_N(k) = \frac{1}{2\pi}\int_{-\pi}^{\pi} \mathrm{e}^{-\mathrm{i}\xi k}[p(\xi)]^N \,\mathrm{d}\xi = \frac{\tau}{2\pi D_N D^{1/\alpha}} \int_{-\pi D_N D^{1/\alpha}}^{\pi D_N D^{1/\alpha}} \mathrm{e}^{-\mathrm{i}\xi x}\left[p\left(\frac{\xi\tau}{D_N D^{1/\alpha}}\right)\right]^N \mathrm{d}\xi,$$

where

$$x = x_{kN} = \frac{k\tau}{D_N D^{1/\alpha}}$$

and ξ was substituted with $(\xi\tau)/(D_N D^{1/\alpha})$. Let $h_\alpha(\xi)$ denote the characteristic function of $h_\alpha(x)$ so that

$$h_\alpha(x) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{e}^{-\mathrm{i}x\xi} h_\alpha(\xi)\,\mathrm{d}\xi$$

holds.

Following [35], the difference $\Delta_N(k)$ in (5.6) can be decomposed and bounded from above as

$$
\begin{aligned}
\Delta_N(k) &= \left| D_N p_N(k) - \frac{\tau}{D^{1/\alpha}} h_\alpha\left(\frac{k\tau}{D_N D^{1/\alpha}}\right)\right| = \left| D_N p_N(k) - \frac{\tau}{D^{1/\alpha}} h_\alpha(x)\right| \\
&= \frac{\tau}{2\pi D^{1/\alpha}} \left| \int_{-\pi D_N D^{1/\alpha}}^{\pi D_N D^{1/\alpha}} \mathrm{e}^{-\mathrm{i}\xi x}\left[p\left(\frac{\xi\tau}{D_N D^{1/\alpha}}\right)\right]^N \mathrm{d}\xi - \int_{-\infty}^{\infty} \mathrm{e}^{-\mathrm{i}\xi x}\, h_\alpha(\xi)\,\mathrm{d}\xi \right| \\
&= \frac{\tau}{2\pi D^{1/\alpha}} \left| \int_{|\xi|<B} \mathrm{e}^{-\mathrm{i}\xi x}\left[p\left(\frac{\xi\tau}{D_N D^{1/\alpha}}\right)^N - h_\alpha(\xi)\right] \mathrm{d}\xi + \int_{B\le|\xi|<\eta D_N D^{1/\alpha}} \mathrm{e}^{-\mathrm{i}\xi x}\left[p\left(\frac{\xi\tau}{D_N D^{1/\alpha}}\right)^N - h_\alpha(\xi)\right] \mathrm{d}\xi \right. \\
&\qquad \left. + \int_{\eta\le\frac{|\xi|}{D_N D^{1/\alpha}}<\pi} \mathrm{e}^{-\mathrm{i}\xi x}\left[p\left(\frac{\xi\tau}{D_N D^{1/\alpha}}\right)^N - h_\alpha(\xi)\right] \mathrm{d}\xi - \int_{|\xi|\ge\pi D_N D^{1/\alpha}} \mathrm{e}^{-\mathrm{i}\xi x}\, h_\alpha(\xi)\,\mathrm{d}\xi \right| \\
&\le \frac{\tau}{2\pi D^{1/\alpha}} \left(\int_{|\xi|<B} \left| p\left(\frac{\xi\tau}{D_N D^{1/\alpha}}\right)^N - h_\alpha(\xi)\right| \mathrm{d}\xi + \int_{B\le|\xi|<\eta D_N D^{1/\alpha}} \left| p\left(\frac{\xi\tau}{D_N D^{1/\alpha}}\right)\right|^N \mathrm{d}\xi \right. \\
&\qquad \left. + \int_{\eta\le\frac{|\xi|}{D_N D^{1/\alpha}}<\pi} \left| p\left(\frac{\xi\tau}{D_N D^{1/\alpha}}\right)\right|^N \mathrm{d}\xi + \int_{|\xi|\ge B} h_\alpha(\xi)\,\mathrm{d}\xi \right)
\end{aligned} \tag{5.9}
$$

with constants B, η to be specified below. The terms involving $h_\alpha(\xi)$ from the second and third integral have been absorbed in the fourth integral. The four integrals are now discussed further individually.

The first integral in (5.9) converges uniformly to zero for $N \to \infty$, because $p(k)$ belongs to the domain of attraction of a stable law with index α, as already noted above.

To estimate the second integral in (5.9), note that the characteristic function $p(\xi)$ belongs to the domain of attraction for index α if and only if it behaves for $|\xi| \to 0$ as [35]

$$|p(\xi)| = \exp\left\{-c|\xi|^\alpha \Lambda\left(\frac{1}{|\xi|}\right)\right\},$$

where $c > 0$ and $\Lambda(x)$ is a slowly varying function at infinity obeying

$$\lim_{N\to\infty} \frac{N\Lambda(D_N)}{D_N^\alpha} = 1.$$

By the representation theorem for slowly varying functions [4, p. 12] there exist functions $d(y)$ and $\varepsilon(y)$ such that the function $\Lambda(y)$ can be represented as

$$\Lambda(y) = d(y)\exp\left\{-\int_b^y \frac{\varepsilon(u)}{u}\,\mathrm{d}u\right\}$$

for some $b > 0$ where $d(y)$ is measurable and $d(y) \to d \in (0, \infty)$ as well as $\varepsilon(u) \to 0$ hold for $y \to \infty$. As a consequence

$$\frac{\Lambda(\lambda y)}{\Lambda(y)} = \frac{d(\lambda y)}{d(y)} \exp\left\{-\int_y^{\lambda y} \frac{\varepsilon(u)}{u}\,\mathrm{d}u\right\}$$

so that, with $\lambda = |\xi|^{-1}$ and $y = D_N$,

$$\frac{\Lambda(D_N/|\xi|)}{\Lambda(D_N)} = |\xi|^{o(1)}(1 + o(1))$$

is obtained for $N \to \infty$. Therefore, there exists for any $\gamma < \alpha$ a positive number $c(\gamma)$ independent of N such that

$$|p_N(\xi)| = \left|p\left(\frac{\xi}{D_N}\right)\right|^N = \left|\exp\left\{-\frac{cN}{D_N^\alpha}\frac{\Lambda(D_N)}{\Lambda(D_N)}\Lambda\left(\frac{D_N}{\xi}\right)|\xi|^\alpha\right\}\right| \le \exp\{-c(\gamma)|\xi|^\gamma\}$$

for sufficiently large N. If N is sufficiently large, it is then possible to choose an $\eta > 0$ (and find $\tilde{c}(\gamma)$) such that

$$\int\limits_{B\le|\xi|<\eta D_N D^{1/\alpha}} \left|p\left(\frac{\xi\tau}{D_N D^{1/\alpha}}\right)\right|^N \mathrm{d}\xi \le \int\limits_{B\le|\xi|<\eta D_N D^{1/\alpha}} \exp\left\{-\tilde{c}\left(\frac{\alpha}{2}\right)|\xi|^{\frac{\alpha}{2}}\right\}\mathrm{d}\xi \le \int\limits_{|\xi|\ge B} \exp\left\{-\tilde{c}\left(\frac{\alpha}{2}\right)|\xi|^{\frac{\alpha}{2}}\right\}\mathrm{d}\xi$$

and this converges to zero for $B \to \infty$.

The third integral in (5.9) is estimated by noting that $|p(\xi)| < 1$ for $0 < |\xi| < 2\pi/\tau$. Hence, there is a positive constant $c > 0$ such that

$$|p(\xi)| \le \mathrm{e}^{-c}$$

for $\eta \le |\xi| \le \pi$. Consequently, with (5.8),

$$\int\limits_{\eta\le\frac{|\xi|}{D_N D^{1/\alpha}}<\pi} \left|p\left(\frac{\xi\tau}{D_N D^{1/\alpha}}\right)\right|^N \mathrm{d}\xi \le 2\pi\mathrm{e}^{-cN}[N\Lambda(N)D]^{1/\alpha}$$

converges to zero as $N \to \infty$.

Finally, the fourth integral in (5.9) converges to zero, because the characteristic function $h_\alpha(\xi)$ is integrable on $\mathbb{R}$. In summary, all four terms in (5.9) vanish for $N \to \infty$, and (5.6) holds. □

Theorem 2. *Let $\mathsf{S} \subset \mathsf{B}_0$ be a subset of invariant states and let $\mathsf{G}(\mathsf{S}, \mathfrak{M}, \varepsilon, \eta)$ be a weak*-neighborhood of ε-almost invariant η-indistinguishable states near S as defined in (5.1). Let P_z be a family of resolutions of the identity, ν a mixing measure, and Q the invariant measure defined in (5.2). Let α, D, D_N and Λ depend on these data as implied by Theorem 1. Then the long-time evolutions*

$$\lim_{\substack{\tau\to\infty,\, N\to\infty\\ [N\Lambda(N)D]^{1/\alpha}/\tau=a}} \mathscr{S}^{-N} = \mathscr{T}_\alpha^{-a} = \int_0^\infty h_\alpha(x)\,\mathscr{T}^{-xa}\,\mathrm{d}x \tag{5.10}$$

are one-sided stable convolution semigroups $\mathscr{T}_\alpha^{-a}$ with long-time parameter $a \ge 0$.

Proof. If the limit in Theorem 1 exists and is nondegenerate so that $D \ne 0$, then the rescaling constants D_N have the form

$$D_N = (N\Lambda(N))^{1/\alpha},$$

where $\Lambda(N)$ is a slowly varying function [55], i.e.

$$\lim_{x\to\infty}\frac{\Lambda(bx)}{\Lambda(x)} = 1$$

for all $b > 0$. Theorem 1 shows that

$$p_N(k) \approx \frac{1}{D_N D^{1/\alpha}} h_\alpha\left(\frac{k}{D_N D^{1/\alpha}}\right)$$

holds for sufficiently large N and all τ. Inserting this into (5.5), one finds

$$\mathscr{S}^{-N} = \sum_{k=1}^{\infty} h_\alpha\left(\frac{k\tau}{D_N D^{1/\alpha}}\right)\mathscr{T}^{-k}\,\frac{[k-(k-1)]\tau}{D_N D^{1/\alpha}}$$

with $D_N \ge 0$ and $D \ge 0$. Introducing the long-time scaling limit

$$\lim_{\tau\to\infty,\, N\to\infty}\frac{D_N D^{1/\alpha}}{\tau} = \lim_{\tau\to\infty,\, N\to\infty}\frac{[N\Lambda(N)D]^{1/\alpha}}{\tau} = a$$

and the macroscopic time parameter a, the result follows. Note that $a \ge 0$ because $D_N \ge 0$ and $D \ge 0$. □

The result shows that a proper mathematical formulation of local stationarity necessitates a generalization of the left-hand side in (1.1a). The time evolution in (1.1) is implicitly restricted to be a translation along the orbit. The general result is that the integration of infinitesimal system changes requires to consider convolutions instead of just translations along the orbit [20, 30]. Convolutions along the orbits reduce to the traditional translations in the special case $\alpha = 1$ as discussed below.

6 Discussion and application

6.1 Abundance of invariant states

The family B_ε of ε-almost invariant BMO-states with $0 \le \varepsilon \le \infty$ has provided a mathematical concept for formulating questions concerning the abundance of time-invariant states and their embedding in the set of all states. Its definition reflects the experimental reality that observations are always performed by integration of experimental data over time intervals. BMO-states allow for singular expectation values, thereby establishing for the first time a general framework to discuss Problem 1 above. Finally, the result in (5.10) provides an answer to Problem 2 by showing that the left-hand side in (1.1a) cannot be considered a time translation along the orbits of the underlying dynamics. Instead the left-hand side is in general the infinitesimal generator of a convolution along rescaled orbits of ε-almost invariant states. The orbits of ε-almost invariant states can approach the manifold of invariant states of the physical system or subsystem of interest at every point for any length of time without being trapped.

6.2 The limit $\alpha \longrightarrow 1$ and irreversibility

Equation (5.10) generalizes time flow to a convolution. For $\alpha \to 1^-$ the convolution reduces to a translation because

$$h_1(x) = \lim_{\alpha\to 1^-} h_\alpha(x) = \delta(x-1) \tag{6.1}$$

and therefore

$$\mathscr{T}^{-a}_{1,\Lambda} = \int_0^\infty \delta\left(\frac{t}{a\tau} - 1\right) \mathscr{T}^{-t/\tau} \frac{\mathrm{d}t}{a\tau} = \mathscr{T}^{-a} \tag{6.2}$$

is a right translation. Here $(t/\tau) \in \mathbb{R}$ denotes a time instant, while $a \ge 0$ is an age or duration. This shows that induced right translations do not form a group, but only a semigroup.

The absence of inverse elements from semigroups raises a generalized perspective on time reversibility because it suggests a reversed formulation of the problem of irreversibility [19, 20, 28, 29]. The well-known problem of irreversibility may be formulated as follows:

Problem 3 (The normal irreversibility problem). Assume that time is reversible. Explain how and why time irreversible equations arise in physics.

Fundamental (microscopic) theories of modern physics generally postulate invariance under the Poincaré group. This expresses the symmetry of the fundamental laws of nature as an isometry of spacetime. Because the time translations are a subgroup of the Poincaré group, this postulates time reversibility and raises the normal irreversibility problem. The explanation of macroscopically irreversible behavior for macroscopic nonequilibrium states of subsystems is due to Boltzmann. It is based on the applicability of statistical mechanics and thermodynamics, the large separation of scales, the importance of low entropy initial conditions, and probabilistic reasoning [43].

The normal problem of irreversibility, which ensues from assuming $t \in \mathbb{R}$, is not only related to the second law of equilibrium thermodynamics. The deeper problem with assuming $t \in \mathbb{R}$ is that an experiment (requiring the preparation of an initial state within an infinity of η-indistinguishable initial states for a dynamical system) cannot be repeated yesterday, but only tomorrow [28]. While it is possible to translate the spatial position of a physical system forward and backward in space, it is not possible to translate the temporal position of a physical system backwards in time. Note that translating an experiment backward in time is not the same as reversing the momenta of all particles in a physical system. This was emphasized in [28, 29]. These observations combined with (6.1) and (6.2) suggest to reformulate the normal irreversibility problem above as follows:

Problem 4 (The reversed irreversibility problem). Assume that time evolution is always irreversible. Explain how and why time reversible equations arise in physics.

The reversed irreversibility problem was introduced in [28]. Its solution is given by Theorem 1 combined with (6.1) and (6.2). Even if $a \geq 0$, time reversibility may arise as a purely mathematical symmetry of model equations in the case $\alpha = 1$. The case $\alpha = 1$ is expected to occur more frequently in applications than the case $0 < \alpha < 1$, because the limit law for $\alpha = 1$ in Theorem 1 has a larger domain of attraction than for $0 < \alpha < 1$ [3, 11, 14, 35]. This explains why equations of motion of the form of (1.1) with time reversal symmetry arise more frequently in physics.

6.3 Infinitesimal generators

The generalized (coarse grained) time evolution operators $\mathscr{T}^a_{\alpha,\Lambda}$ found in Theorem 1 and (5.10) are convolution operators. In the following, their dependence on a slowly varying function Λ will be suppressed to write $\mathscr{T}^a_\alpha$ for short. The notation $\mathscr{T}^a_1 = \mathscr{T}^a$ will also be used.

The operators $\mathscr{T}^a_\alpha$ form a family of strongly continuous semigroups on $C_0(G)^*$ provided that the translations $\mathscr{T}$ inside the integral in (5.10) are strongly continuous [48, 51] and $\int_0^\infty \|\mathscr{T}^s\| h_\alpha(s/a)/a \, ds < \infty$. In this case the infinitesimal generators $\mathscr{T}^a_\alpha$ for $0 < \alpha \leq 1$ are defined by

$$\mathrm{A}_\alpha \widetilde{\varrho} = \operatorname*{s-lim}_{a \to 0} \frac{\mathscr{T}^a_\alpha \widetilde{\varrho} - \widetilde{\varrho}}{a}$$

for all $\widetilde{\varrho} \in C_0(G)^*$ for which the strong limit s-lim exists. In general, the infinitesimal generators A_α are unbounded operators. If $\mathrm{A} = -\mathrm{d}/\mathrm{d}a$ denotes the infinitesimal generator of the translation $\mathscr{T}^a$, then

$$\mathrm{A}_\alpha = -(-\mathrm{A})^\alpha = -\left(\frac{\mathrm{d}}{\mathrm{d}a}\right)^\alpha \tag{6.3}$$

are fractional time derivatives [23, 58]. The action of A_α on mixed states can be represented in different ways. Frequently an integral representation

$$\mathrm{A}_\alpha \widetilde{\varrho} = \lim_{\epsilon \to o} C \int_\epsilon^\infty a^{-\alpha-1} (\mathbf{1} - \mathscr{T}^a) \widetilde{\varrho} \, \mathrm{d}a$$

of Marchaud type [1, 45] is used. The integral representation

$$\mathrm{A}_\alpha \widetilde{\varrho} = \lim_{\epsilon \to o} C \int_\epsilon^\infty a^{-\alpha} \mathrm{A} (\mathbf{1} - a\mathrm{A})^{-1} \widetilde{\varrho} \, \mathrm{d}a$$

in terms of the resolvent of A (see [40]) defines the same fractional derivative operator [57]. Representations of Grünwald–Letnikov type are also well known [58].

6.4 An application to experiment

This section discusses a direct application of the mathematical analysis to an experiment. In physics the time evolutions from (5.10) with $\alpha < 1$ have been linked to observations of so-called “anomalous dynamics” or “strange kinetics” for many years (see [22, 33, 37, 46] for reviews). A specific example is the dielectric relaxation in glasses [25, 26]. It is briefly discussed and selected here for two reasons: Firstly, because experimental data over up to 19 decades in time are available for this example [44], and secondly, because the generic appearance of excess wings [41] at high frequencies has remained obscure theoretically. Excess wings are found not only in dielectric spectroscopy, but recently also in depolarized light scattering experiments [52].

Let $f(t)$ with $f(0) = 1$ denote the normalized, electrical dipolar polarization observed in a dielectric relaxation experiment [13, 41]. Then the complex frequency dependent dielectric susceptibility is $\varepsilon(u) = 1 - u\widehat{f}(u)$, where $\widehat{f}(u)$ denotes the Laplace transform of $f(t)$, $u = -2\pi\mathrm{i}\nu$, $\mathrm{i}^2 = -1$, and ν is the frequency [25, p. 402, (18)]. It is generally accepted that coarse graining the microscopic equations (1.1) for a fluid weakly perturbed by a time-varying electromagnetic field at sufficiently high temperatures gives rise to a coarse grained evolution $\mathscr{T}_\alpha^a$ with $\alpha = 1$ for the relaxation function f in linear response theory [2, 13, 42]. The macroscopic relaxation function f expresses an average over microscopic orientational dipole-dipole fluctuations. The function f obeys the normalized Debye equation for dielectric relaxation [13, Chapter III, §10]

$$\frac{\mathrm{d}f}{\mathrm{d}a} = -f \tag{6.4}$$

with initial value $f(0) = 1$. Writing $a = t/\tau$ for the dimensionless duration, the solution is the exponential (Debye) relaxation function

$$f(t) = \exp\left(-\frac{t}{\tau}\right) \qquad \text{(Debye)} \tag{6.5a}$$

and its associated dielectric function

$$\varepsilon(u) = \frac{1}{1 + u\tau}. \tag{6.5b}$$

While this form is confirmed experimentally at high temperatures, deviations are found at lower temperatures. This is illustrated for the imaginary part of $\varepsilon(u)$ in the upper curve in Figure 1. The comparison between theoretical results and experimental data is done numerically.

Many alternatives to the Debye relaxation have been suggested phenomenologically to improve the fit to the data. A time-honored example is the stretched exponential Kohlrausch relaxation [38, 39], revived by Williams and Watts [59]:

$$f(t) = \exp\left(-\left[\frac{t}{\tau}\right]^\alpha\right) \qquad \text{(KWW)},$$
$$\varepsilon(u) = 1 - H_{11}^{11}\left([u\tau]^\alpha \,\middle|\, \begin{matrix}(1,1)\\(1,\alpha)\end{matrix}\right),$$

where $\tau > 0$ is the relaxation time and $0 < \alpha \le 1$ is the stretching exponent. The dielectric susceptibility for the stretched exponential was only found 150 years later by the author [27] in terms of special functions defined through inverse Mellin-transforms of Γ-functions and known as H-functions [12].

A popular alternative to stretching in time is to stretch in frequency. In this case a stretching exponent α is introduced into (6.5b) rather than into (6.5a). This leads to the Cole–Cole (CC) relaxation [8]

$$f(t) = \mathrm{E}_\alpha\left(-\left[\frac{t}{\tau}\right]^\alpha\right), \tag{6.6a}$$

$$\varepsilon(u) = \frac{1}{1 + (u\tau)^\alpha} \qquad \text{(CC)}, \tag{6.6b}$$

where

$$E_a(x) = \sum_{k=0}^{\infty} \frac{x^k}{\Gamma(ak+1)}$$

is the Mittag-Leffler function [47]. It is well known that the relaxation function $f(t)$ for Cole–Cole relaxation is intimately related to eigenfunctions of the fractional derivatives from (6.3) [15, 23]. Unfortunately, the Cole–Cole form (6.6b) exhibits a symmetric α-peak, while asymmetric α-peaks are observed experimentally for many materials [41]. Therefore, a second way to introduce the stretching exponent α into the Debye function (6.5b), known as the Cole–Davidson (CD) form, was introduced in [9]:

$$f(t) = \frac{\Gamma(\alpha, t/\tau)}{\Gamma(\alpha)}, \tag{6.7a}$$

$$\varepsilon(u) = \frac{1}{(1 + u\tau)^\alpha} \qquad \text{(CD)}, \tag{6.7b}$$

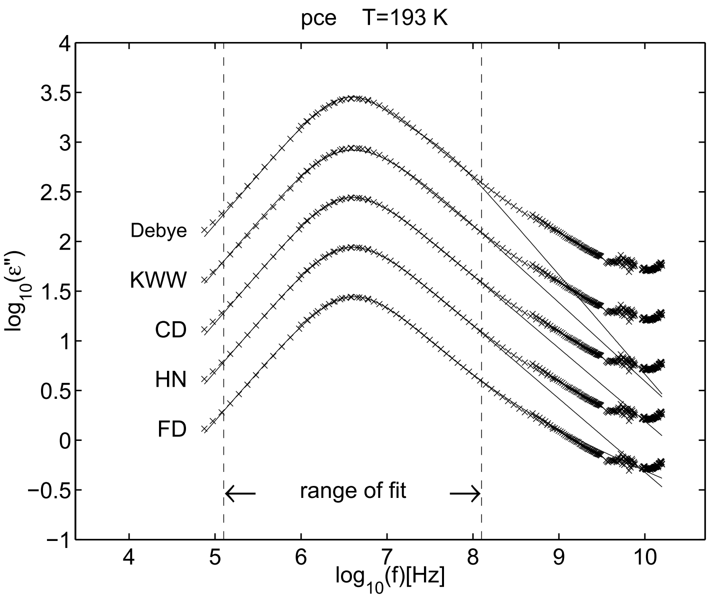

Figure 1. Five different fits to the imaginary part $\operatorname{Im}\varepsilon(u)$ ($u = -2\pi i\nu$) of the complex dielectric function of propylene carbonate at $T = 193$ K as a function of frequency. Experimental data represented by crosses are taken from [54]. The fitting functions corresponds to an exponential (Debye), stretched exponential (KWW), Cole–Davidson [10, 24] Havriliak–Negami [18, 27] and the fractional dynamics (FD) relaxation from (6.11). The range over which the data were fitted is indicated by dashed vertical lines. For clarity the data were displaced vertically by half a decade each. The original location of the data corresponds to the curve labelled FD.

where

$$\Gamma(a, x) = \int_a^\infty y^{a-1} e^{-y}\, \mathrm{d}y$$

denotes the complementary incomplete Gamma function, Finally, the CC-form and CD-form are combined into the popular Havriliak–Negami (HN) form given as

$$f(t) = 1 - \frac{1}{\Gamma(\beta)} H^{11}_{12}\left(\left[\frac{t}{\tau}\right]^\alpha \middle| \begin{matrix}(1,1)\\ (\beta,1)(0,\alpha)\end{matrix}\right), \tag{6.8a}$$

$$\varepsilon(u) = \frac{1}{(1+[u\tau]^\alpha)^\beta} \qquad \text{(HN)} \tag{6.8b}$$

with three fit parameters. The explicit formula (6.8a) for the Havriliak–Negami relaxation function was first given in [27].

The functional forms (6.5), (6.6), (6.7), and (6.8) are used universally almost without exception to fit broadband dielectric data [41, Table 3.1]. A quantitative comparison of the different forms is shown for propylene carbonate at $T = 193$ K in Figure 1. It is found that all of the functional forms (6.5), (6.6), (6.7), and (6.8) deviate from the experimental data at high frequency or give an unsatisfactory fit. Therefore, a weighted sum of two or more of these functional forms is routinely used to fit the excess wing in glass forming materials. An example with seven fit parameters (four exponents, two relaxation times and one mixing parameter that determines the relative weight of the two HN-functions) is seen, e.g., in [41, p. 66, Figure 3.5].

Generalized time evolution operators yield a generalized Mittag-Leffler-type function with only three parameters (instead of seven for HN), that allows to fit both, the asymmetric peak and the excess wing with a single stretching exponent. Every coarse grained evolution operator $\mathscr{T}^a_\beta$ with time scale $\tau > 0$ and order $\beta(\tau)$ factorizes as

$$\mathscr{T}^{\tau a}_{\beta(\tau)} = \mathscr{T}^{\tau_1 a}_{\beta(\tau)} \mathscr{T}^{(\tau-\tau_1)a}_{\beta(\tau)} = \mathscr{T}^{\tau_1 a}_{\beta(\tau)} \mathscr{T}^{\tau_2 a}_{\beta(\tau_1+\tau_2)} \tag{6.9}$$

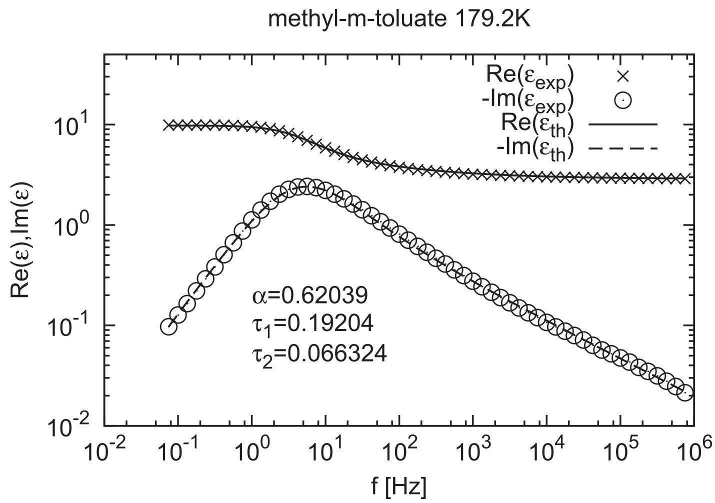

Figure 2. Theoretical FD-fit $\varepsilon_{\mathrm{th}}$ using (6.11) to the real and imaginary part of the complex dielectric function $\varepsilon_{\mathrm{exp}}$ of experimental data for methyl-m-toluate at 179.2 K as a function of frequency [7]. The data are from [49].

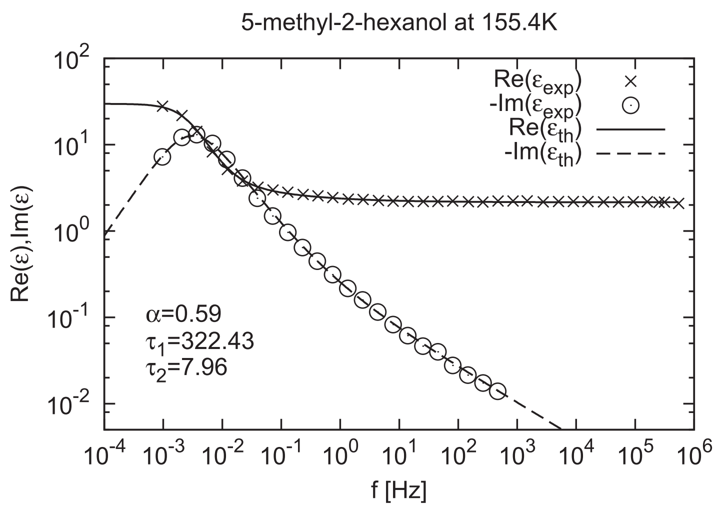

Figure 3. Theoretical FD-fit $\varepsilon_{\mathrm{th}}$ using (6.11) to the real and imaginary part of the complex dielectric function $\varepsilon_{\mathrm{exp}}$ of experimental data of 5-methyl-2-hexanol at 155.4 K as a function of frequency [7]. The data are from [36].

with $\tau = \tau_1 + \tau_2$ by virtue of the basic law from (1.4). Assume now that one of the factors in (6.9), say the second, becomes approximately fractional in the sense that

$$\mathscr{T}^{\tau_2 a}_{\beta(\tau_1+\tau_2)} \approx \mathscr{T}^{\tau_2 a}_{\alpha(\tau_2)}$$

holds in the weak* or strong topology with

$$\lim_{\tau_2\to 0} \alpha(\tau_2) = \beta(\tau).$$

The resulting composite time evolution $\mathscr{T}^{\tau_1 a}_{\beta(\tau)} \mathscr{T}^{\tau_2 a}_{\alpha(\tau_2)}$ was studied in [25, 26] for the case $\beta(\tau) = 1$. Computing the infinitesimal generator of $\mathscr{T}^{\tau_1 a}_{1} \mathscr{T}^{\tau_2 a}_{\alpha(\tau_2)}$ and following the steps to derive the Debye equation yields the differential equation [25, 26]

$$\tau_1 \frac{\mathrm{d}f}{\mathrm{d}a} + \tau_2^{\alpha} \mathrm{A}_{\alpha} f = -f \tag{6.10}$$

with A_α from (6.3) and initial value $f(0) = 1$. It reduces to (6.4) for $\alpha = 1$. The three-parameter function solving (6.10) and its associated dielectric function read

$$f(t) = \mathrm{E}_{(1,1-\alpha),1}\left(-\frac{a}{\tau_1}, -\frac{\tau_2^\alpha}{\tau_1}a^{1-\alpha}\right), \tag{6.11a}$$

$$\varepsilon(u) = \frac{1 + (u\tau_2)^\alpha}{1 + (u\tau_2)^\alpha + u\tau_1} \qquad \text{(FD)}, \tag{6.11b}$$

where

$$\mathrm{E}_{(a_1,a_2),b}(z_1, z_2) = \sum_{k=0}^{\infty} \sum_{\substack{\ell_1 \geq 0 \\ \ell_1+\ell_2=k}} \sum_{\ell_2 \geq 0} \frac{k!}{\ell_1!\ell_2!} \frac{z_1^{\ell_1} z_2^{\ell_2}}{\Gamma(b + a_1\ell_1 + a_2\ell_2)},$$

where $a_1, a_2 > 0$ and $b, z_1, z_2 \in \mathbb{C}$ is the binomial Mittag-Leffler function [15, 32]. In Figure 1 the resulting fit is denoted as "fractional dynamics" (FD). It reproduces the high frequency wing even outside the range of fitting, while this is not the case for the other four curves, labelled Debye, KWW, CD, and HN.

Figures 2 and 3 show FD-fits to methyl-m-toluate and 5-methyl-2-hexanol as two additional examples. Figures 2 and 3 show both the imaginary part, but also the real part of the complex dielectric susceptibility function. The excess wing, best visible in the imaginary part, is characteristic for anomalous dynamics or glassy dynamics [41, 52]. 5-methyl-2-hexanol and methyl-m-toluate are just two from many real physical examples where the mathematical analysis of time flow has become experimentally important.

References

[1] A. V. Balakrishnan, Fractional powers of closed operators and the semigroups generated by them, *Pacific J. Math.* **10** (1960), 419–353.

[2] R. Balescu, *Equilibrium and Nonequilibrium Statistical Mechanics*, Wiley, New York, 1975.

[3] H. Bergström, *Limit Theorems for Convolutions*, Wiley, New York, 1963.

[4] N. H. Bingham, C. M. Goldie and J. L. Teugels, *Regular Variation*, Cambridge University Press, Cambridge, 1987.

[5] O. Bratteli and D. Robinson, *Operator Algebras and Quantum Statistical Mechanics I*, Springer, Berlin, 1979.

[6] O. Bratteli and D. Robinson, *Operator Algebras and Quantum Statistical Mechanics II*, Springer, Berlin, 1981.

[7] S. Candelaresi, *Fraktionale Ansätze in dielektrischer Breitbandspektroskopie*, Master's thesis, Universität Stuttgart, 2008.

[8] K. S. Cole and R. H. Cole, Dispersion and absorption in dielectrics, *J. Chem. Phys.* **9** (1941), 341–351.

[9] D. W. Davidson and R. H. Cole, Dielectric relaxation in glycerine, *J. Chem. Phys.* **18** (1950), 1417–1418.

[10] D. W. Davidson and R. H. Cole, Dielectric relaxation in glycerol, propylene glycol and n-propanol, *J. Chem. Phys.* **19** (1951), 1484–1490.

[11] W. Feller, *An Introduction to Probability Theory and Its Applications, vol. II*, Wiley, New York, 1971.

[12] C. Fox, The G and H functions as symmetrical Fourier kernels, *Trans. Am. Math. Soc.* **98** (1961), 395–429.

[13] H. Fröhlich, *Theory of Dielectrics: Dielectric Constant and Dielectric Loss*, Oxford University Press, Oxford, 1949.

[14] B. V. Gnedenko and A. N. Kolmogorov, *Limit Distributions for Sums of Independent Random Variables*, Addison-Wesley, Cambridge, 1954.

[15] R. Gorenflo, A. Kilbas, F. Mainardi and S. Rogosin, *Mittag-Leffler Functions, Related topics and Applications*, Springer, Heidelberg, 2014.

[16] R. Haag, *Local Quantum Physics*, Springer, Berlin, 1992.

[17] R. Haag and D. Kastler, An algebraic approach to quantum field theory, *J. Math. Physics* **5** (1964), 848–861.

[18] S. Havriliak and S. Negami, A complex plane analysis of α-dispersions in some polymer systems, *J. Polym. Sci. C* **14** (1966), 99–117.

[19] R. Hilfer, An extension of the dynamical foundation for the statistical equilibrium concept, *Physica A* **221** (1995), 89–96.

[20] R. Hilfer, Foundations of fractional dynamics, *Fractals* **3** (1995), 549–556.

[21] R. Hilfer, Fractional dynamics, irreversibility and ergodicity breaking, *Chaos Solitons Fractals* **5** (1995), 1475–1484.

[22] R. Hilfer, *Applications of Fractional Calculus in Physics*, World Scientific, Singapore, 2000.

[23] R. Hilfer, Fractional time evolution, in: *Applications of Fractional Calculus in Physics*, World Scientific, Singapore (2000), 87–130.

[24] R. Hilfer, Analytical representations for relaxation functions of glasses, *J. Noncryst. Solids* **305** (2002), 122–126.

[25] R. Hilfer, Experimental evidence for fractional time evolution in glass forming materials, *Chem. Phys.* **284** (2002), 399–408.

[26] R. Hilfer, Fitting the excess wing in the dielectric α-relaxation of propylene carbonate, *J. Phys. Condens. Matter* **14** (2002), 2297–2301.
[27] R. Hilfer, H-function representations for stretched exponential relaxation and non-Debye susceptibilities in glassy systems, *Phys. Rev. E* **65** (2002), 061510.
[28] R. Hilfer, Remarks on fractional time, in: *Time, Quantum and Information*, Springer, Berlin (2003), 235–240.
[29] R. Hilfer, Threefold introduction to fractional derivatives, in: *Anomalous Transport: Foundations and Applications*, Wiley-VCH, Weinheim (2008), 17–74.
[30] R. Hilfer, Foundations of fractional dynamics: A short account, in: *Fractional Dynamics: Recent Advances*, World Scientific, Singapore (2011), 207–227.
[31] R. Hilfer, Time automorphisms on C*-algebras, *Mathematics* **3** (2015), 626–643.
[32] R. Hilfer, Y. Luchko and Z. Tomovski, Operational method for the solution of fractional differential equations with generalized Riemann–Liouville fractional derivatives, *Fract. Calc. Appl. Anal.* **12** (2009), 299–318.
[33] R. Hilfer, R. Metzler, A. Blumen and J. Klafter (eds.), Special issue on strange kinetics, *Chem. Phys.* **284** (2002), 1–541.
[34] E. Hille and R. Phillips, *Functional Analysis and Semi-Groups*, American Mathematical Society, Providence, 1957.
[35] I. A. Ibragimov and Y. V. Linnik, *Independent and Stationary Sequences of Random Variables*, Wolters-Nordhoff Publishing, Groningen, 1971.
[36] O. E. Kalinovskaya and J. K. Vij, The exponential dielectric relaxation dynamics in a secondary alcohol's supercooled liquid and glassy states, *J. Chem. Phys.* **112** (2000), 3262–3266.
[37] R. Klages, G. Radons and I. M. Sokolov (eds.), *Anomalous Transport*, Wiley-VCH, Weinheim, 2008.
[38] R. Kohlrausch, Theorie des elektrischen Rückstandes in der Leidener Flasche, *Ann. Phys.* **167** (1854), 56–82.
[39] R. Kohlrausch, Theorie des elektrischen Rückstandes in der Leidener Flasche, *Ann. Phys.* **167** (1854), 179–214.
[40] H. Komatsu, Fractional powers of operators II: Interpolation spaces, *Pacific J. Math.* **21** (1967), 89–111.
[41] F. Kremer and A. Schönhals (eds.), *Broadband Dielectric Spectroscopy*. Springer, Berlin, 2003.
[42] R. Kubo, M. Toda and N. Hashitsume, *Statistical Physics, vol. II*, Springer, Berlin, 1978.
[43] J. Lebowitz, Statistical mechanics: A selective review of two central issues, *Rev. Mod. Phys.* **71** (1999), 346–357.
[44] P. Lunkenheimer, U. Schneider, R. Brand and A. Loidl, Glassy dynamics, *Contemp. Phys.* **41** (2000), 15–36.
[45] A. Marchaud, Sur les derivees et sur les differences des fonctions de variables reelles, *J. Math. Pures Appl. (9)* **6** (1927), 337–425.
[46] R. Metzler and J. Klafter, The random walk's guide to anomalous diffusion: A fractional dynamics approach, *Phys. Rep.* **339** (2000), 1–77.
[47] G. Mittag-Leffler, Sur l'integrale de Laplace–Abel, *C.R. Acad. Sci. Paris* **135** (1902), 937–939.
[48] E. Nelson, A functional calculus using singular Laplace integrals, *Trans. Amer. Math. Soc.* **88** (1958), 400–413.
[49] A. I. Nielsen, T. Christensen, B. Jakobsen, K. Niss, N. Boye Olsen, R. Richert and J. C. Dyre, Prevalence of approximate sqrt(t) relaxation for the dielectric alpha process in viscous organic liquids, *J. Chem. Phys.* **130** (2009), 154508.
[50] A. Pazy, *Semigroups of Linear Operators and Applications to Partial Differential Equations*, Springer, Berlin, 1983.
[51] R. S. Phillips, On the generation of semigroups of linear operators, *Pacific J. Math.* **2** (1952), 343–369.
[52] N. Petzold, R. Schmidtke, R. Kahlau, D. Bock, R. Meier, B. Micko, D. Kruk and E. Rössler, Evolution of the dynamic susceptibility in molecular glass formers: Results from light scattering, dielectric spectroscopy and NMR, *J. Chem. Phys.* **138** (2013), 12A510.
[53] W. Rudin, *Functional Analysis*, McGraw-Hill, New York, 1973.
[54] U. Schneider, P. Lunkenheimer, R. Brand and A. Loidl, Broadband dielectric spectroscopy on glass-forming propylene carbonate, *Phys. Rev. E*, **59** (1999), 6924–6936.
[55] E. Seneta, *Regularly Varying Functions*, Springer, Berlin, 1976.
[56] H. Spohn, *Large Scale Dynamics of Interacting Particles*, Springer, Berlin, 1991.
[57] J. Stafney, Integral representations of fractional powers of infinitesimal generators, *Illinois J. Math.* **20** (1976), 124–133.
[58] U. Westphal, Fractional powers of infinitesimal generators of semigroups, in: *Applications of Fractional Calculus in Physics*, World Scientific, Singapore (2000), 131–170.
[59] G. Williams and D. C. Watts, Non-symmetrical dielectric relaxation behaviour arising from a simple empirical decay function, *Trans. Faraday Soc.* **66** (1970), 80–85.
[60] K. Yosida, *Functional Analysis*, Springer, Berlin, 1965.