Fractional Diffusion Based on Riemann-Liouville Fractional Derivatives†

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A fractional diffusion equation based on Riemann−Liouville fractional derivatives is solved exactly. The initial values are given as fractional integrals. The solution is obtained in terms of *H*-functions. It differs from the known solution of fractional diffusion equations based on fractional integrals. The solution of fractional diffusion based on a Riemann−Liouville fractional time derivative does not admit a probabilistic interpretation in contrast with fractional diffusion based on fractional integrals. While the fractional initial value problem is well defined and the solution finite at all times, its values for $t \to 0$ are divergent.

Anomalous subdiffusive transport appears to be a universal experimental phenomenon.[1−3] Examples occur in widely different systems ranging from amorphous semiconductors[4,5] through polymers[6−8] and composite heterogeneous films[9] to porous media.[10,11] Theoretical investigations into anomalous diffusion and continuous time random walks have been a major focus of H. Schers research for many years.[4,10,12,13] The purpose of this paper is to discuss a theoretical approach based on the replacement of the time derivative in the diffusion equation with a derivative of noninteger order (fractional derivative).

Many investigators have proposed the use of fractional time derivatives for subdiffusive transport on a purely mathematical or heuristic basis.[8,14−20] From the perspective of theoretical physics this proposal touches upon fundamental principles such as locality, irreversibility, and invariance under time translations because fractional derivatives are non-local operators that are not invariant under time reversal.[21] These issues are generally avoided in heuristic and mathematical proposals, but were discussed recently in the context of long time limits and coarse graining.[21] It was found that fractional time derivatives with orders between 0 and 1 may generally appear as infinitesimal generators of a coarse grained macroscopic time evolution.[20−24]

Differential equations involving fractional derivatives raise a second basic problem, related to the first, that will be the focus of this paper. The second problem is whether to replace the integer order derivative by a Riemann−Liouville, by a Weyl, by a Riesz, by a Grünwald, or by a Marchaud fractional derivative (see refs 25−28 for definitions of these different derivatives). Different authors have introduced different derivatives depending on the physical situation.[21,29−31]

Given the basic objective of introducing fractional derivatives into the diffusion equation the present paper will be concerned with the equation

$$\mathrm{D}^{\alpha}_{0+} f(\boldsymbol{r}, t) = C_{\alpha}\Delta f(\boldsymbol{r}, t) \tag{1}$$

where $f(\boldsymbol{r}, t)$ denotes the unknown field and C_α denotes the fractional diffusion constant with dimensions [cm/s$^\alpha$]. The fractional derivative operator, denoted as D^{α}_{0+}, is the Riemann−Liouville derivative of order α and with lower limit $\alpha \in \mathbb{R}$. It is defined as[25,28]

$$(\mathrm{D}^{\alpha}_{a+} f)(x) = \frac{\mathrm{d}}{\mathrm{d}x}(\mathrm{I}^{1-\alpha}_{a+} f)(x) \tag{2}$$

where

$$(I^{\alpha}_{a+} f)(x) = \frac{1}{\Gamma(\alpha)}\int_a^x (x-y)^{\alpha-1} f(y)\,\mathrm{d}y \tag{3}$$

is the Riemann−Liouville fractional integral with order α and lower limit a. Although many authors have investigated fractional diffusion problems[14−16,19,32,33] it seems that eq 1 has not been solved previously. In fact, it was recently questioned whether an approach using eq 1 is consistent.[29,34] It is the purpose of this paper to solve eq 1 exactly, thereby establishing its consistency for appropriate initial conditions.

Let me emphasize that eq 1 differs from the popular equation introduced and solved in ref 16. The latter equation is obtained by first rewriting the diffusion equation in integral form as

$$f(\boldsymbol{r}, t) = f_0\delta(\boldsymbol{r}) + C_1\int_0^t \Delta f(\boldsymbol{r}, t')\,\mathrm{d}t' \tag{4}$$

where C_1 is the usual diffusion constant, $\delta(\boldsymbol{r})$ is the Dirac measure at the origin, and where the initial condition $f(\boldsymbol{r}, 0) = f_0\delta(\boldsymbol{r})$ has been incorporated. Then the integral on the right hand side is replaced by a fractional Riemann−Liouville integral to arrive at the fractional integral form

$$\begin{aligned} f(\boldsymbol{r}, t) &= f_0\delta(\boldsymbol{r}) + \frac{C_\alpha}{\Gamma(\alpha)}\int_0^t (t-t')^{\alpha-1}\Delta f(\boldsymbol{r}, t')\,\mathrm{d}t' \\ &= f_0\delta(\boldsymbol{r}) + C_\alpha(\mathrm{I}^{\alpha}_{0+}\Delta f)(\boldsymbol{r}, t) \end{aligned} \tag{5a}$$

or, upon differentiating both sides, at

$$\frac{\partial}{\partial t} f(\boldsymbol{r}, t) = C_\alpha(\mathrm{D}^{1-\alpha}_{0+}\Delta f)(\boldsymbol{r}, t) \tag{5b}$$

where C_α is again a fractional diffusion constant. For $\alpha = 1$ this reduces to eq 4. The exact solution of eq 5 is known and given by eq 22 below.

In refs 35, 36, it was shown that eqs 5 have a rigorous relation with continuous time random walks of the kind investigated

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frequently by Harvey Scher.[4,10,12,13] More precisely, eq 1 was found to correspond exactly to a continuous time random walk with the long tailed waiting time density[35,36]

$$\psi(t;\alpha,\tau_0) = \frac{1}{\tau_0}\left(\frac{t}{\tau_0}\right)^{\alpha-1} E_{\alpha,\alpha}\left(-\frac{t^\alpha}{\tau_0^\alpha}\right) \tag{6}$$

where τ_0 is a time constant. Here $E_{a,b}(x)$ denotes the generalized Mittag−Leffler function defined by

$$E_{a,b}(x) = \sum_{k=0}^{\infty} \frac{x^k}{\Gamma(ak+b)} \tag{7}$$

for all $a > 0$ and $b \in \mathrm{C}$. For $\alpha = 1$ this reduces to an exponential waiting time density. For $0 < \alpha < 1$ these waiting time densities have a long tail decaying as $\psi(t) \sim t^{-1-\alpha}$ for $t \to \infty$. Interestingly, $\psi(t) \sim t^{\alpha-1}$ diverges algebraically for $t \to 0$. It follows from refs 20−24 that among the the waiting time densities with long tails the densities $\psi(t;\alpha,\tau_0)$ represent important universality classes for continuous time random walks.

Note that eqs 5 and 1 are not equivalent. The difference between eqs 5 and 1 has to do with the initial conditions. An appropriate initial condition is found by analyzing the stationary case. One finds that the fractional integral

$$\mathrm{I}_{0+}^{(1-\alpha)} f(\boldsymbol{r}, 0+) = f_{0,\alpha}\delta(\boldsymbol{r}) \tag{8}$$

is preserved during the time evolution. This is a nonlocal initial condition. It implies the divergence of $f(\boldsymbol{r}, t)$ as $t \to 0$, as is characteristic for fractional stationarity.[20,24]

Equation 1 with initial condition (eq 8) can be solved exactly by Fourier−Laplace techniques. Let the Fourier transformation be defined as

$$\mathcal{F}\{f(\boldsymbol{r})\}(\boldsymbol{q}) = \int_{\mathrm{R}^d} e^{i\boldsymbol{q}\cdot\boldsymbol{r}} f(\boldsymbol{r})\mathrm{d}\boldsymbol{r} \tag{9}$$

Fourier and Laplace transformation of eq 1 now yields

$$f(\boldsymbol{q}, u) = \frac{f_{0,\alpha}}{C_\alpha \boldsymbol{q}^2 + u^\alpha} \tag{10}$$

Inverting the Laplace transform gives

$$f(\boldsymbol{q}, t) = f_{0,\alpha} t^{(\alpha-1)} E_{\alpha,\alpha}(-C_\alpha \boldsymbol{q}^2 t^\alpha) \tag{11}$$

Setting $\boldsymbol{q} = 0$ shows that $f(\boldsymbol{r}, t)$ cannot be a probability density because its normalization would depend on t. Hence eq 1 does not admit a probabilistic interpretation contrary to eq 5.

To obtain $f(\boldsymbol{r}, t)$ it is advantageous to first invert the Fourier transform in eq 10 and only later the Laplace transform. The Fourier transform may be inverted by noting the formula[37]

$$(2\pi)^{-d/2}\int e^{i\boldsymbol{q}\cdot\boldsymbol{r}}\left(\frac{|\boldsymbol{r}|}{m}\right)^{1-(d/2)} K_{(d-2)/2}(m|\boldsymbol{r}|)\,\mathrm{d}\boldsymbol{r} = \frac{1}{\boldsymbol{q}^2 + m^2} \tag{12}$$

which leads to

$$f(\boldsymbol{r}, u) = f_{0,\alpha}(2\pi C_\alpha)^{-d/2}\left(\frac{r}{\sqrt{C_\alpha}}\right)^{1-(d/2)} u^{\alpha(d-2)/4} K_{(d-2)/2}\left(\frac{r u^{\alpha/2}}{\sqrt{C_\alpha}}\right) \tag{13}$$

with $r = |\boldsymbol{r}|$. To invert the Laplace transform it is convenient to use the relation

$$\mathcal{M}\{f(t)\}(s) = \frac{\mathcal{M}\{\mathcal{L}\{f(t)\}(u)\}(1-s)}{\Gamma(1-s)} \tag{14}$$

between the Laplace transform and the Mellin transform

$$\mathcal{M}\{f(t)\}(s) = \int_0^\infty t^{s-1} f(t)\,\mathrm{d}t \tag{15}$$

of a function $f(t)$. Setting $A = r/\sqrt{C_\alpha}$, $\lambda = \alpha/2$, $\nu = (d-2)/2$, and $\mu = \alpha(d-2)/4$ and using the general relation

$$\mathcal{M}\{x^q g(bx^p)\}(s) = \frac{1}{p} b^{-(s+q)/p} g\left(\frac{s+q}{p}\right) \quad (b,p > 0) \tag{16}$$

leads to

$$\mathcal{M}\{f(r,u)\}(s) = \frac{f_{0,\alpha}}{\lambda}(2\pi C_\alpha)^{-d/2} A^{1-(d/2)} A^{-(s+\mu)/\lambda}\,\mathcal{M}\{K_\nu(u)\}((s+\mu)/\lambda) \tag{17}$$

The Mellin transform of the Bessel function reads[38]

$$\mathcal{M}\{K_\nu(x)\}(s) = 2^{s-2}\Gamma\left(\frac{s+\nu}{2}\right)\Gamma\left(\frac{s-\nu}{2}\right) \tag{18}$$

Inserting this, using eq 14, and restoring the original variables then yields

$$\mathcal{M}\{f(r,t)\}(s) = \frac{f_{0,\alpha}}{\alpha(r^2\pi)^{d/2}}\left(\frac{r}{2\sqrt{C_\alpha}}\right)^{2(1-(1/\alpha))} \times \left(\frac{r}{2\sqrt{C_\alpha}}\right)^{2s/\alpha} \frac{\Gamma\left(\frac{d}{2} - \left(1-\frac{1}{\alpha}\right) - \frac{s}{\alpha}\right)\Gamma\left(\frac{1}{\alpha} - \frac{s}{\alpha}\right)}{\Gamma(1-s)} \tag{19}$$

for the Mellin transform of f. Comparing this with the definition of the general H-function in eqs 23, 24 allows one to identify the H-function parameters as $m = 0$, $n = 2$, $p = 2$, $q = 1$, $A_1 = A_2 = 1/\alpha$, $a_1 = 1 - (d/2) + (1 - (1/\alpha))$, $a_2 = (1 - (1/\alpha))$, $b_1 = 0$, and $B_1 = 1$, if $(\alpha d/2) - (\alpha - 1) > 0$. Then the result becomes

$$f(r,t) = \frac{f_{0,\alpha}}{\alpha(r^2\pi)^{d/2}}\left(\frac{r}{2\sqrt{C_\alpha}}\right)^{2(1-(1/\alpha))} H_{21}^{02}\left(\left(\frac{2\sqrt{C_\alpha}}{r}\right)^{2/\alpha} t \,\middle|\, \begin{matrix}\left(1-\frac{d}{2}+\left(1-\frac{1}{\alpha}\right),\frac{1}{\alpha}\right),\left(\left(1-\frac{1}{\alpha}\right),\frac{1}{\alpha}\right)\\ (0,1)\end{matrix}\right) \tag{20}$$

This may be simplified using eqs 35−37 to become finally

$$f(r,t) = \frac{f_{0,\alpha}\,t^{(\alpha-1)}}{(r^2\pi)^{d/2}} H_{12}^{20}\left(\frac{r^2}{4C_\alpha t^\alpha}\,\middle|\,\begin{matrix}(\alpha,\alpha)\\ (d/2,1),(1,1)\end{matrix}\right) \tag{21}$$

This result should be compared with the known solution

$$f(r,t) = \frac{f_0}{(r^2\pi)^{d/2}} H_{12}^{02}\left(\frac{r^2}{4C_\alpha t^\alpha}\,\middle|\,\begin{matrix}(1,\alpha)\\ (d/2,1),(1,1)\end{matrix}\right) \tag{22}$$

of eq 5 in which case $f(\boldsymbol{r}, t)$ is also a probability density.

In summary, this paper has shown that fractional diffusion based on Riemann−Liouville derivatives requires a fractional initial condition given by eq 8. With this initial condition the fractional Cauchy problem can be solved exactly in terms of H-functions, and the solution is similar to the exact solution of the fractional integral form in eq 5. However contrary to eq 5 whose solution is a probability density, and which is related to continuous time random walks, the solution of eq 1 does not admit a probabilistic interpretation.

The reader may ask why it is important for theoretical physics and chemistry to investigate different forms of fractional diffusion. An answer was given already in refs 20−24, and recently again in refs 39, 40. In these works it was found that fractional time derivatives arise generally as infinitesimal generators of the time evolution when taking a long-time scaling limit. Hence the importance of investigating fractional equations arises from the necessity to sharpen the concepts of equilibrium, stationary states, and time evolution in the long time limit.

APPENDIX: H-FUNCTIONS

The H-function or order $(m, n, p, q) \in \mathbb{N}^4$ and parameters $A_i \in \mathbb{R}_+ (i = 1, ..., p)$, $B_i \in \mathbb{R}_+ (i = 1, ..., q)$, $a_i \in \mathbb{C} (i = 1, ..., p)$, and $b_i \in \mathbb{C} (i = 1, ..., q)$ is defined for $z \in \mathbb{C}$, $z \neq 0$ by the contour integral[41−45]

$$H^{m,n}_{p,q}\left(z\,\middle|\,\begin{matrix}(a_1, A_1), ..., (a_p, A_p)\\(b_1, B_1), ..., (b_q, B_q)\end{matrix}\right) = \frac{1}{2\pi i}\int_{L} \eta(s) z^{-s}\,\mathrm{d}s \tag{23}$$

where the integrand is

$$\eta(s) = \frac{\prod_{i=1}^{m}\Gamma(b_i + B_i s)\prod_{i=1}^{n}\Gamma(1 - a_i - A_i s)}{\prod_{i=n+1}^{p}\Gamma(a_i + A_i s)\prod_{i=m+1}^{q}\Gamma(1 - b_i - B_i s)} \tag{24}$$

In eq 23, $z^{-s} = \exp\{-s \log |z| - i \arg z\}$ and $\arg z$ is not necessarily the principal value. The integers m, n, p, q must satisfy

$$0 \le m \le q, \qquad 0 \le n \le p \tag{25}$$

and empty products are interpreted as being unity. The parameters are restricted by the condition

$$\mathrm{P}_a \cap \mathrm{P}_b = \varnothing \tag{26}$$

where

$$\begin{aligned}\mathrm{P}_a &= \{\text{poles of } \Gamma(1 - a_i - A_i s)\} = \\ &\left\{\frac{1 - a_i + k}{A_i} \in \mathbb{C} :\ i = 1, ..., n; k \in \mathbb{N}_0\right\}\\ \mathrm{P}_b &= \{\text{poles of } \Gamma(b_i + B_i s)\} = \\ &\left\{\frac{-b_i - k}{B_i} \in \mathbb{C} :\ i = 1, ..., m; k \in \mathbb{N}_0\right\}\end{aligned} \tag{27}$$

are the poles of the numerator in eq 24. The integral converges if one of the following conditions holds:[45]

$$L = L(c - i\infty, c + i\infty; \mathrm{P}_a, \mathrm{P}_b); \quad |\arg z| < C\pi/2; \quad C > 0 \tag{28a}$$

$$L = L(c - i\infty, c + i\infty; \mathrm{P}_a, \mathrm{P}_b); \quad |\arg z| = C\pi/2; \quad C \ge 0; \quad cD < -\mathrm{Re}\, F \tag{28b}$$

$$L = L(-\infty + i\gamma_1, -\infty + i\gamma_2; \mathrm{P}_a, \mathrm{P}_b); \quad D > 0; \quad 0 < |z| < \infty \tag{29a}$$

$$L = L(-\infty, + i\gamma_1, -\infty + i\gamma_2; \mathrm{P}_a, \mathrm{P}_b); \quad D = 0; \quad 0 < |z| < E^{-1} \tag{29b}$$

$$L = L(-\infty, + i\gamma_1, -\infty + i\gamma_2; \mathrm{P}_a, \mathrm{P}_b); \quad D = 0; \quad |z| = E^{-1}; C \ge 0; \mathrm{Re}\, F < 0 \tag{29c}$$

$$L = L(\infty + i\gamma_1, \infty + i\gamma_2; \mathrm{P}_a, \mathrm{P}_b); \quad D > 0; \quad 0 < |z| < \infty \tag{30a}$$

$$L = L(\infty + i\gamma_1, \infty + i\gamma_2; \mathrm{P}_a, \mathrm{P}_b); \quad D = 0; \quad |z| > E^{-1} \tag{30b}$$

$$L = L(\infty + i\gamma_1, \infty + i\gamma_2; \mathrm{P}_a, \mathrm{P}_b); \quad D = 0; \quad |z| = E^{-1}; C \ge 0; \mathrm{Re}\, F < 0 \tag{30c}$$

where $\gamma_1 < \gamma_2$. Here $L(z_1, z_2; G_1, G_2)$ denotes a contour in the complex plane starting at z_1 and ending at z_2 and separating the points in G_1 from those in G_2, and the notation

$$C = \sum_{i=1}^{n} A_i - \sum_{i=n+1}^{p} A_i + \sum_{i=1}^{m} B_i - \sum_{i=m+1}^{q} B_i \tag{31}$$

$$D = \sum_{i=1}^{q} B_i - \sum_{i=1}^{p} A_i \tag{32}$$

$$E = \prod_{i=1}^{p} A_i^{A_i} \prod_{i=1}^{q} B_i^{-B_i} \tag{33}$$

$$F = \sum_{i=1}^{q} b_i - \sum_{i=1}^{p} a_j + (p - q)/2 + 1 \tag{34}$$

was employed. The H-functions are analytic for $z \neq 0$ and multivalued (single valued on the Riemann surface of $\log z$).

A change of variables in eq 23 shows

$$H^{m,n}_{p,q}\left(z\,\middle|\,\begin{matrix}(a_1, A_1), ..., (a_p, A_p)\\(b_1, B_1), ..., (b_q, B_q)\end{matrix}\right) = H^{n,m}_{q,p}\left(\frac{1}{z}\,\middle|\,\begin{matrix}(1 - b_1, B_1), ..., (1 - b_q, B_q)\\(1 - a_1, A_1), ..., (1 - a_p, A_p)\end{matrix}\right) \tag{35}$$

which allows us to transform an H-function with $D > 0$ and $\arg z$ to one with $D < 0$ and $\arg(1/z)$. For $\gamma > 0$

$$\frac{1}{\gamma} H^{m,n}_{p,q}\left(z\,\middle|\,\begin{matrix}(a_1, A_1), ..., (a_p, A_p)\\(b_1, B_1), ..., (b_q, B_q)\end{matrix}\right) = H^{m,n}_{p,q}\left(z^{\gamma}\,\middle|\,\begin{matrix}(a_1, \gamma A_1), ..., (a_p, \gamma A_p)\\(b_1, \gamma B_1), ..., (b_q, \gamma B_q)\end{matrix}\right) \tag{36}$$

while for $\gamma \in \mathbb{R}$

$$z^{\gamma} H^{m,n}_{p,q}\left(z\,\middle|\,\begin{matrix}(a_1, A_1), ..., (a_p, A_p)\\(b_1, B_1), ..., (b_q, B_q)\end{matrix}\right) = H^{m,n}_{p,q}\left(z\,\middle|\,\begin{matrix}(a_1 + \gamma A_1, A_1), ..., (a_p + \gamma A_p, A_p)\\(b_1 + \gamma B_1, B_1), ..., (b_q + \gamma B_q, B_q)\end{matrix}\right) \tag{37}$$

holds.

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