# Interpretation of a fractional diffusion equation with nonconserved probability density in terms of experimental systems with trapping or recombination

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The fractional diffusion equation that is constructed replacing the time derivative with a fractional derivative, $_0D_t^\alpha f = C_\alpha \partial^2 f / \partial x^2$, where $_0D_t^\alpha$ is the Riemann-Liouville derivative operator, is characterized by a probability density that decays with time as $t^{\alpha-1}$ ($\alpha < 1$) and an initial condition that diverges as $t \to 0$ [R. Hilfer, J. Phys. Chem. B **104**, 3914 (2000)]. These seemingly unphysical features have obstructed the application of the fractional diffusion equation. The paper clarifies the meaning of these properties adopting concrete physical interpretations of experimentally verified models: the decay of free-carrier density in a semiconductor with an exponential distribution of traps, and the decay of ion-recombination isothermal luminescence. We conclude that the fractional difusion equation is a suitable representation of diffusion in disordered media with dissipative processes such as trapping or recombination involving an initial exponential distribution either in the energy or spatial axis. The fractional decay does not consider explicitly the starting excitation and ultrashort time-scale relaxation that forms the initial exponential distribution, and therefore it cannot be extrapolated to $t = 0$.

## I. INTRODUCTION

The modeling of the dynamics of anomalous processes by means of differential equations that involve derivatives of fractional order has provided interesting results in recent years in a variety of fields of science and engineering [1,2]. For example, physical models based on fractional derivatives have been amply studied in relation to scaling concepts, carrier transport in heterogeneous or disordered media [3], and the relaxation of complex systems [4]. We highlight as the most studied and applied model the fractional diffusion equations (FDE), which play an important role in describing anomalous diffusion [3].

In this paper, we discuss the physical interpretation of a FDE based on the replacement of the time derivative in the ordinary diffusion equation with a derivative of noninteger order,

$$_0D_t^\alpha f(x,t) = C_\alpha \frac{\partial^2 f(x,t)}{\partial x^2}. \qquad (1)$$

Here, $f$ is a probability distribution and $C_\alpha$ is the fractional diffusion coefficient, which takes the form

$$C_\alpha = K_0 \tau_\alpha^{1-\alpha} \qquad (2)$$

with respect to the ordinary diffusion coefficient $K_0$ (in $cm^2/s$) and a time constant $\tau_\alpha$. $_0D_t^\alpha$ is the fractional Riemann-Liouville derivative operator of order $0 < \alpha < 1$ and with lower limit $t = 0$. It is defined as [3,5]

$$_aD_x^\gamma[f(x)] = \frac{1}{\Gamma(\alpha)}\frac{d}{dx}{_aI_x^{1-\gamma}}, \qquad (3)$$

where

$$_aI_x^\beta[f(x)] = \frac{1}{\Gamma(\beta)}\int_a^x (x-y)^{\beta-1}f(y)dy \qquad (4)$$

is the Riemann-Liouville fractional integral operator of order $\beta$ with a lower real limit $a$ and $\Gamma(\beta)$ is the gamma function [1]. The fractional time derivative can be written

$$_0D_t^\alpha f(t) = \frac{1}{\Gamma(1-\alpha)}\frac{d}{dt}\left\{\int_0^t \frac{f(t')}{(t-t')^\alpha}dt'\right\} \qquad (5)$$

and its Laplace transform is

$$L\{_0D_t^\alpha f(t)\} = u^\alpha f(u) - f_0, \qquad (6)$$

where $f_0$ is defined below in Eq. (7).

The properties of Eq. (1) have been widely studied in the literature. The solutions in different dimensions of space under certain initial conditions and the moments of the distribution have been obtained [4–9]. Feldman *et al.* [10] considered Eq. (1) for the fractional generalization of the Liouville equation for dissipative systems, and Tarasov [6] provided a fractional analog for the normalization conditions for distribution functions. The FDE (1) was used also to define a non-exponential relaxation process that is not based on a distribution of relaxation times [4].

Equation (1) was discussed as a natural generalized diffusion equation suggesting that it could describe anomalous diffusion process [5,11–16]. However, those possible applications have been scarcely developed because of a problem of physical interpretation, owing to the fact that the $f(x,t)$ is not a normalized function [14,16] and the initial condition could not be the usual one. Indeed, it was found [5] that $f(x,t)$ is divergent for $t \to 0$. The structure of the initial condition was described in the context of the Cauchy problem by Trujillo *et al.*, see Lemma 9 in [7], implying

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$$f_0 = \left. \left( {}_0I_t^{1-\alpha} f \right)(t) \right|_{t=0} = \lim_{t \to 0} t^{1-\alpha} f(t). \qquad (7)$$

Therefore, the solution diverges as $f(t) \propto t^{\alpha-1}$ as $t \to 0$. This fact has been discussed in several works. Hilfer related the nonlocal form of the initial condition [5] to the fractional stationarity concept [12,17] which establishes, in addition to the conventional constants, a second class of stationary states that obey power-law time dependence. Recently, Ryabov [9] discussed the solution and initial conditions of Eq. (1) in different dimensions and concluded that "virtual sources" of the diffusion agent are required at the origin, meaning that some mass is injected at the origin. However, this injection is not defined by the boundary conditions of the problem. Thus, the author [9] concludes that the presence of such "virtual sources" in solutions is physically meaningless and one needs to resolve this contradiction. Also, recently I showed [18] that the FDE (1) describes the diffusion of free carriers in multiple trapping (MT) with an exponential distribution of gap states, but the question of the initial distribution was not considered in that work [18].

In this paper, we consider the apparent contradiction mentioned by Ryabov [9] by adopting definite physical interpretations that are experimentally verified, and we realize the properties of the formal model with a fractional time derivative. We first discuss the initial condition of Eq. (1) and the decay law that is intrinsic to this equation even in spatially homogeneous conditions. In relation with the MT interpretation [18], thereafter we revise the heuristic arguments for describing the transient decay of an injected pulse of free carriers in amorphous semiconductors [19–21] in order to discuss the meaning of the initial condition of the FDE. We also discuss the power-law decay of isothermal luminiscence for ion recombination in condensed media [22–24]. We conclude that Eq. (1) is a physical representation of diffusion in disordered media with dissipative processes such as trapping or recombination.

## II. INITIAL CONDITION AND DECAY LAW OF THE FDE

Let us consider the implications of Eq. (1) for a diffusion process. Taking into account that the diffusive flux of particles (normalized by the total number density) is given by

$$j = -K_0 \frac{\partial f}{\partial x}, \qquad (8)$$

Eq. (1) can be written as a generalized equation of continuity,

$$\tau_\alpha^{\alpha-1} {}_0D_t^\alpha f(x,t) = -\frac{\partial j}{\partial x}. \qquad (9)$$

Integrating between two impermeable boundaries, such that $j(x_1) = j(x_2) = 0$ (it may be, for example, $x_1 = 0$ and $x_2 = +\infty$), we obtain

$$ {}_0D_t^\alpha \left[ \int_{x_1}^{x_2} f(x,t)dx \right] = 0. \qquad (10)$$

In the ordinary diffusion problem, the equivalent of Eq. (10) (with the normal time derivative) states the conservation of the number of particles: Since no particles are crossing the boundaries, their number must be constant. However, Eq. (10) does not state the conservation of the probability, but instead the condition

$$ {}_0I_t^{1-\alpha} \left[ \int_{x_1}^{x_2} f(x,t)dx \right] = \text{const.} \qquad (11)$$

The fractional integral of the total probability is preserved during the evolution. Therefore, Eq. (1) involves a peculiar and nontrivial temporal dynamics. To make the point clearer, let us consider the expression of the FDE in Fourier-Laplace space (with variables $x \to q$ and $t \to u$),

$$f(u,q) = \frac{f_{0,\alpha}}{\tau_\alpha^{\alpha-1} u^\alpha + K_0 q^2}. \qquad (12)$$

The case $q \to 0$ describes a situation in which the carrier density is homogenous, i.e., there is no diffusion at all, and this is the only case that we will consider in this paper. From Eq. (12) we obtain

$$f(u,0) = f_{0,\alpha} \tau_\alpha^{1-\alpha} u^{-\alpha}. \qquad (13)$$

Now the time decay of the probability in spatially homogeneous conditions is given by

$$f(t) = \frac{f_{0,\alpha} \tau_\alpha^{-\alpha+1}}{\Gamma(\alpha)} t^{\alpha-1}. \qquad (14)$$

The last result, the decay law for Eq. (1), shows that the number of particles decreases with time.

It has been remarked in the literature [5,7] that Eq. (1) requires an initial condition for the Green function of the form

$$ {}_{0+}I_t^{1-\alpha} [f(x,0+)] = f_{0,\alpha} \delta(x), \qquad (15)$$

where $\delta(x)$ is the Dirac measure at the origin and $f_{0,\alpha}$ is a constant. Indeed, as pointed out above, it has been shown rigorously [7] that the initial condition $f(t)$ satisfies the condition

$$\lim_{x \to 0+} t^{1-\alpha} f(t) = \text{const.} \qquad (16)$$

It is clear that the temporal dynamics in the FDE implies the divergence of $f(t)$ as $t \to 0$ [5,7], a point that will be discussed further in Sec. IV.

## III. ANOMALOUS FDE WITH CONSERVED PROBABILITY DENSITY

Note that another FDE has been amply studied in the literature of anomalous diffusion [3,5,14,16,25–27], namely

$$\frac{\partial}{\partial t} f(x,t) = K_\alpha \left( D_0^{1-\alpha} \frac{\partial^2 f}{\partial x^2} \right)(x,t). \qquad (17)$$

This last equation is rigorously related [5,28] to the continuous-time random walk formalism of Scher and Montroll [29]. Equations (1) and (17), obviously, are not equivalent [5]. We can see this inequality, for instance, comparing
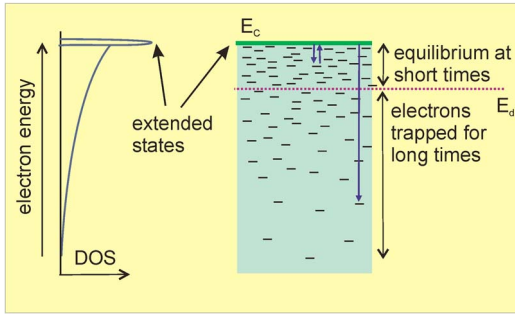
FIG. 1. (Color online) Energy diagram of a semiconductor with a transport level (indicated by the energy of the conduction band, $E_c$) and localized states in the band gap. The left diagram shows the density of states (DOS) and the right diagram shows the trapping-detrapping kinetics as a function of the depth of the localized state in the band gap. At time $t$ after injection, states shallower than the demarcation level $E_d$, defined in Eq. (21), are able to frequently detrap and retrap electrons and establish thermal equilibrium, while electrons trapped at states deeper than $E_d$ have a negligible probability of being released.

the expression of the FDE (17) in Fourier-Laplace space, which is

$$f(q,u) = \frac{f_{0,\alpha} u^{\alpha-1}}{u^\alpha + K_0 \tau_\alpha^{1-\alpha} q^2}, \tag{18}$$

and the corresponding expression for Eq. (1) given by Eq. (12).

Equation (17) has the initial condition $f(x,0) = f_0 \delta(x)$ and the probability is conserved as in ordinary diffusion. Notice that shifting the Riemann-Liouville derivative to either side of the diffusion equation changes drastically the resulting temporal dynamics.

## IV. DECAY OF FREE CARRIERS IN SEMICONDUCTORS BY TRAPPING IN BAND-GAP STATES

The multiple trapping (MT) model applies to a semiconductor with a band of extended states and a tail of localized states, which usually takes the exponential form, as schematically shown in Fig. 1,

$$g(E) = \frac{N_t}{k_B T_c} \exp[(E - E_c)/k_B T_c], \tag{19}$$

where $N_t$ is the total density of traps, $E_c$ is the lower band edge energy, $k_B$ is Boltzmann's constant, and $T_c$ characterizes the broadening of the distribution, with $\alpha = T/T_c < 1$ at temperature $T$. The density of conduction-band electrons can be written $n_c = N_c f_c$, where $N_c$ is the total density and $f_c$ the fractional occupancy of conduction-band states. The mobility is assumed to drop abruptly at the energy of transition from localized to extended states, so that $J = N_c j$ is the diffusive flux of conduction-band electrons, where $j$ is described by Eq. (8), where $K_0$ is the diffusion coefficient of the electrons in extended states.

This model explained long tails of the electrical current observed in time-of-flight experiments [30], the time depen-

dence of transient photocurrents [21], and it was used for deriving drift mobilities [31]. MT has been widely used to describe carrier transport in amorphous semiconductors and is also of current research interest, for example in nanostructured semiconductors such as networks of $TiO_2$ nanoparticles used in dye-sensitized solar cells [32–34]. When MT is represented by a FDE, as shown in [18], the characteristic time in Eq. (2) takes the value

$$\tau_\alpha = \left( \alpha \Gamma(\alpha) \Gamma(1-\alpha) \frac{N_t}{N_c} \right)^{1/\alpha} \nu_0^{-1}. \tag{20}$$

In the following, we review the heuristic arguments for the evolution of free-carrier density in MT provided by Tiedje and Rose [19,20] and Orenstein and Kastner [21] (see also [31]), and we will show that these arguments explain and give a simple interpretation to the temporal decay of the probability of the FDE, described in Eq. (14), which is a crucial issue in the interpretation of the FDE [11,14–16]. As already mentioned, we consider that both free and trapped electron densities are homogeneous in the semiconductor. This assumption corresponds to the experimental technique of transient photocurrents in conditions of homogeneous light absorption [35]. Furthermore, we note that the disappearance of carriers by recombination is completely neglected in this model.

If at time $t=0$, $n_0$ excess carriers are optically excited into the transport states of a semiconductor, they are rapidly thermalized in the extended states, and at a time of the order of one trapping time ($\approx 10^{-13} - 10^{-12}$ s), they will be trapped in the localized states. The distribution of trapped carriers is proportional to the density of states for an energy-independent capture cross section. Trapped carriers are subsequently released into the transport states. The energy depth below the transport band becomes a key factor for the frequency of the transitions between a given trap and the conduction band, because the probability of detrapping to the conduction band is proportional to $\exp[(E - E_c)/k_B T]$. After a time $t$, the shallower states release and retrap electrons a large number of times, so that these states effectively obtain a thermal distribution. In contrast, the electrons in deeper levels remain frozen, as indicated in Fig. 1. The demarcation energy level $E_d(t)$ above which electrons are released at time $t$ is determined by

$$t = \nu_0^{-1} \exp\{[E_c - E_d(t)]/k_B T\}. \tag{21}$$

According to Eq. (21), the demarcation level sinks with time into the distribution of localized states, depopulating the states above $E_d$ and adding more trapped charge to the states below $E_d$ in addition to the already present frozen-in charge, so that the total density of excess carriers remains constant, as indicated in Fig. 2. This $E_d(t)$ acts in the way of a quasi-Fermi-level, and the bulk of the injected charge will be concentrated near $E_d(t)$. The distribution of carriers in the band gap is described by [36]

$$n_t(E,t) = \phi(t) g(E) F[E, E_d(t)] \tag{22}$$

in terms of a time-dependent occupancy factor $\phi(t)$, which ensures that the intitial excitation is conserved, and the Fermi-Dirac distribution function,
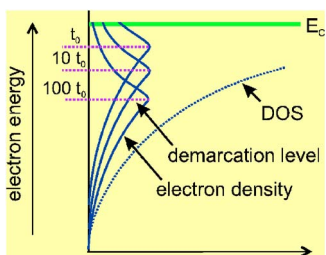
FIG. 2. (Color online) Energy diagram of a semiconductor showing the exponential density of states (DOS) and the energy distribution of carriers. The peak of the distribution is at the demarcation level $E_d$ and sinks deeper in the band gap with time.

$$F(E, E_F) = \left[1 + e^{(E - E_F)/k_B T}\right]^{-1}. \tag{23}$$

The conservation of carriers can be stated as

$$\phi(t) \int_{-\infty}^{E_c} g(E) F[E, E_d(t)] dE = n_0. \tag{24}$$

Equation (24) can be transformed to

$$\phi(t)(\nu_0 t)^{-a} N_t \alpha \int_0^{\nu_0 t} \frac{x^{\alpha - 1}}{1 + x} dx = n_0. \tag{25}$$

Provided that $\nu_0 t \gg 1$, Eq. (25) gives

$$\phi(t) = \frac{n_0}{N_t \alpha \Gamma(\alpha) \Gamma(1 - \alpha)} (\nu_0 t)^{\alpha}. \tag{26}$$

The occupancy factor $\phi(t)$ in Eq. (26) increases with time to compensate for the sinking of the quasi-Fermi-level $E_d(t)$. Equation (26) is valid starting at times of the order $t \approx 10^{-10}$ s, much longer than individual trapping times. Alternatively, this means that at the initial instant $t_0 > 0$ where the Eq. (26) applies, the quasi-Fermi-level must be several times $k_B T$ below the conduction band. In this case, the density of electrons in the conduction band is a small tail of the total distribution,

$$n_c(t) = N_c \phi(t) e^{[E_d(t) - E_c]/k_B T}, \tag{27}$$

such that $n_c \ll n_0$, see Fig. 2. Using Eqs. (21) and (26), it is obtained that the occupancy factor of electrons in extended states $f_c = n_c / N_c$ decreases with time as

$$f_c(t) = \frac{n_0}{N_t \alpha \Gamma(\alpha) \Gamma(1 - \alpha)} (\nu_0 t)^{\alpha - 1}. \tag{28}$$

As a physical interpretation of the decay process of the type $t^{\alpha - 1}$ for the free electrons, we remark that in Eq. (28) the dependence $t^{-1}$ is due to the sinking of the demarcation level by the release of trapped carriers, while the dependence $t^{\alpha}$ arises from the increasing occupancy at $E_d(t)$ by retrapping of the released carriers.

We must emphasize that these considerations leading to the decay law of Eq. (28) were explained and confirmed experimentally in the 1980s [19,21]. Now we remark that in Eq. (28) we have recovered the decay law that is obtained from the FDE, Eq. (14). Hence, the decay law that results from the assumption of thermalization in the MT model

[19,21,31] corresponds to the rigorous solution of the homogenous case of the FDE (1).

This physical model allows us to understand the physical significance of the seemingly strange temporal structure of the FDE (1). Normally one expects that an equation of motion describes the dynamics of the whole system. However, Eq. (1) is a different kind of equation of motion, in the sense that a part of the dynamics is not described by it. In the general kinetics of the trapping process, the distribution of carriers in the band gap continuously shifts in the energy space, while maintaining the shape and total area (Fig. 2). This is a dissipative dynamics in which the total energy [determined by the electrochemical potential of the electrons $E_d(t)$] decreases with time while the number density is conserved. The relaxation in the full energy space is not described explicitly in Eq. (1), which only contains the resulting evolution of the carriers in extended states, in correspondence with the requirements of the experimental techniques that monitor carrier transport detecting only the free carriers. The disappearance of the probability for $f_c$ takes on a perfectly valid physical meaning as a decrease of the number of free carriers with time. In conclusion, the fractional derivative maps a complex local dynamics of a many-body problem into the diffusion equation.

When using the FDE to represent the dissipative process, such process must consistently be active in the initial conditions, i.e., a $\delta$ sheet of charge injected at some point cannot be stationary but must be decreasing with time, which explains Eqs. (15) and (16). Concerning the divergence of the solution of the FDE as $t \to 0$, which is remarked in the literature [5,8], it must be recognized that in the physical system described above, the decay law Eq. (28) cannot be extrapolated to $t = 0$, because Eq. (28) requires a minimal time for the formation of the initial exponential distribution of electrons at $E_d < E_c$ and a thermal distribution at the levels close to $E_c$, as indicated in connection with Eq. (26). Therefore, the divergence of the initial condition at $t \to 0$ is not an impediment for the application of the FDE of Eq. (1) in the description of experiments.

We next consider another physical problem with certain points in common with the decay of free electrons in semiconductors with traps. The model for ion-geminate recombination allows for a discussion of the apparent divergence of the initial condition in configurational space, instead of the energy axis.

## V. THE DECAY LAW IN GEMINATE-ION RECOMBINATION

Many organic compounds supported in a solid matrix can be easily oxidized by a light pulse. The electrons initially trapped in the surrounding medium, when returning to the ionized species, cause a time-dependent luminescence that has been known, for more than half a century [22], to follow a decay law of the form $t^{-m}$, with $m$ close to 1. Such power-law decays have been recently observed by transient absorption spectroscopy in polymer/fullerene composites [37,38], which are of current interest for organic solar cells.

Debye and Edwards were the first to propose a model for ion-recombination luminescence [22]. In this model, they de-

rived the initial distribution of electrons based on the idea of diffusion of trapped electrons towards the recombination center, with a recombination rate of the form $t^{-m}$. The model was later criticized by Abell and Mozumber [23] on the grounds that the distribution function would diverge as $t \to 0$. The latter authors modified some assumptions of the theory but then the resulting decay law was of the form $t^{-3/2}$ as in standard diffusion, in contrast with the observation of an exponent $m \approx 1$. We consider here a related model of Tachiya and Mozumber [24] which explains the general decay law $t^{-m}$ from the assumption of tunneling recombination. This model describes well the recent experimental observation of the photovoltage decay of electrons injected from photo-oxidized dye molecules into ultrathin $TiO_2$ layers [39].

The model considers the usual exponential rate law for spatial-dependent tunneling recombination of electrons with respect to the distance $r$ to the recombination center,

$$k(r) = \nu_0 e^{-r/a}, \tag{29}$$

where $\nu_0$ and $a$ are parameters. The value of $\nu_0$ is of the order of $10^{-15}$ s$^{-1}$, and the value of $a$ is of the order of tenths of a nanometer. The *initial* probability distribution for the distance between the trapped electron and the parent ion, $f_0(r)$, is well approximated by an exponential function [24]

$$f_0(r) = \frac{1}{b} e^{-r/b}. \tag{30}$$

The probability distribution evolves with time as

$$f(r,t) = e^{-k(r)t} f_0(r), \tag{31}$$

and the integrated probability, representing the fraction of trapped electrons that survive at time $t$, is given by

$$F(t) = \int_0^\infty e^{-k(r)t} f_0(r) dr, \tag{32}$$

which can be written as

$$F(t) = \frac{a}{b} \gamma(\nu_0 t, a/b)(\nu_0 t)^{-a/b} \tag{33}$$

in terms of the incomplete gamma function $\gamma(x,z)$, which for $x \to \infty$ reduces to the gamma function $\Gamma(z)$. Therefore, at times $\nu_0 t \gg 1$, the electron density decays in the power-law form as

$$F(t) = \frac{a}{b} \Gamma(a/b)(\nu_0 t)^{-a/b}. \tag{34}$$

The time-dependent luminescence is given by $I(t) \propto dF/dt \propto t^{-1-a/b}$, and $m \approx 1$ is obtained provided that $a \ll b$.

As mentioned above, this system bears a strong resemblance with the relaxation of free carriers in amorphous semiconductors discussed in the foregoing. In the present system, the probability of recombination decreases exponentially with the distance from the recombination center. Consequently, the front of the initial exponential distribution recedes with time from $r=0$, see Fig. 1 in [24] and also [40]. The picture is similar to Fig. 2, but in this case the profile changes in *configuration space* and the total number of initial carriers formed by the external excitation is not conserved.

The power law decay in Eq. (34), if extrapolated to $t \to 0$ gives an unphysical divergence, and this led to some discussion in the literature [23], as remarked before. However, while Eq. (34) is useful for explaining the experimental observation of power-law decays of total carrier density over several decades in time [37–39], it should be recognized that the complete model starts from an exponential distribution at $t=0$, i.e., without a divergence, a fact which is recovered as $F(0)=1$ in the general expression Eq. (33). If indeed $\nu_0$ is of the order $10^{-15}$ s$^{-1}$, the time domain where the decay *departs* from the power law occurs at such short time scales that usually it will not be experimentally observable, unless using the fastest detection techniques available. In any case, Eq. (34) must be accompanied by the restriction $\nu_0 t \gg 1$, as clearly remarked in Ref. [24].

## VI. CONCLUSION

An apparent contradiction of the physical applications of a fractional time derivative has been clarified. The physical models related to the fractional time derivative are aimed to describe the decay of a selected kind of carrier with an initial distribution of an exponential form, either in the energy or spatial axis. The physical models that represent the decay as a power law in time are restricted to account for the dissipative processes of trapping and/or recombination and do not consider explicitly the necessary starting excitation and ultrashort time-scale relaxation that forms the initial exponential distribution. For this reason, the models with a fractional time derivative or power law decay in time cannot be extrapolated to $t=0$.

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