Letter

Consistent Hamiltonian models for space-momentum diffusion

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We develop a unified Hamiltonian approach to the diffusion of a particle coupled to a dissipative environment, an archetypal model widely invoked to interpret condensed phase phenomena, such as polymerization and cold-atom diffusion in optical lattices. By appropriate choices of the coupling functions, we reformulate phenomenological diffusion models by adding otherwise ignored space-momentum terms. We thus numerically predict a variety of diffusion regimes, from diffusion saturation to superballistic diffusion. With reference to ultracold atoms in optical lattices, we also show that time correlated external noises prevent superdiffusion from exceeding Richardson's law. Some of these results are unexpected and call for experimental validation.

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In recent years a growing number of experimental [1-6]and theoretical studies [7-12] addressed the problem of cold atoms diffusing in optical lattices generated by counterpropagating laser beams. Such systems are subjected to intrinsic damping mechanisms often modeled by power-law momentum-dependent friction terms. For this reason, the diffusion of cold atoms has been addressed mostly in the momentum space. However, a consistent treatment of spatial diffusion requires a fully Hamiltonian formalism to correctly account for the coupling of the space and momentum variables. We do so by adopting a well-established approach [13,14], whereby a cold atom can be modeled as an open system nonlinearly coupled to a "heat bath" of classical oscillators, which mimic the light fields. By an appropriate choice of the coupling functions, we thus generalize known phenomenological space-momentum diffusion models, discuss their limitations, and point to a richer phenomenology for further experimental work.

The current models predict three spatial diffusion regimes for cold atoms: normal, Lévy, and Obukhov-Richardson diffusion [15]. The last two regimes imply momentum diffusion to explain the superdiffusive mean-square displacements (MSDs), $\langle x^2(t) \rangle \sim t^{\alpha}$ with $1 < \alpha \leq 3$. The question then rises, whether in a more rigorous Hamiltonian model, inertia can combine with environmental fluctuations to produce an even faster time growth of the MSD. As a matter of fact, MSD measurements in one-dimensional ensembles of ultracold ⁸⁷Rb atoms return exponents significantly faster than ballistic diffusion, $\alpha = 2$ [16–18]. Remarkably, the current phenomenology also fails to reproduce the opposite limiting situation, namely, diffusion saturation, $\alpha = 0$ [19–21]. Such a mechanism is expected to play a key role in the interpretation of polymerization experiments [22–24], that is another topic of current interest. In conclusion, at least two distinct classes of ongoing experiments, i.e., on cold atoms and polymerization, call for a more consistent approach to space-momentum diffusion. The multipurpose microscopic Hamiltonian approach proposed here will be tested against cold-atom diffusion in optical lattices.

Model. To best model the coupling of spatial and momentum variables, we started with a heuristic Hamiltonian consisting of a system part, which depends only on the state variables (x, p), and a bath part, which depends on both the state of system and the bath variables $\{q_i, p_i\}$, namely

$$H(x, p; \{q_j, p_j\}) = \frac{p^2}{2m} + U(x) + \sum_{j=1}^{\infty} \left[\frac{p_j^2}{2m_j} + \frac{1}{2} m_j \omega_j^2 \right] \times \left(q_j - \frac{c_j}{m_j \omega_j^2} F(x, p) \right)^2 .$$
(1)

Here, U(x) is a substrate potential and F(x, p) an arbitrary function of the system variables [13]. To the best of our knowledge, a few authors did address the case of nonlinear coupling functions [25–29], but a discussion of their impact on the asymptotic diffusion of the system is lacking. We also remark that in Eq. (1) any hypothetical coupling between bath momenta and the system variables has been eliminated by means of a suitable canonical transformation [25].

The relevant canonical equations of motion are readily obtained from Eq. (1). The set of inhomogeneous linear differential equations for the bath variables can be solved exactly. Substituting the solutions for the bath variables into the equations of motion for the system (see the Supplement Material [30] for details), we finally arrive at the following nonlinear generalized Langevin equation (GLE),

$$\dot{p} = -\partial_x U + G(t)\partial_x F, \quad \dot{x} = p/m - G(t)\partial_p F,$$
 (2)

$$G(t) = -\int_0^t \Gamma(t-s)[\partial_x F\dot{x}(s) + \partial_p F\dot{p}(s)]ds + \varepsilon(t), \quad (3)$$

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with ∂_{ψ} denoting the partial derivative with respect to $\psi = x, p$. The zero-mean valued colored noise $\varepsilon(t)$ depends on the bath preparation and is related to the memory kernel $\Gamma(t)$ through the fluctuation-dissipation relationship (FDR), $\langle \varepsilon(t)\varepsilon(t') \rangle = k_B T \Gamma(t - t')$, where k_B is the Boltzmann constant and *T* the bath temperature. The reduced system of Eqs. (2) and (3) is thus the result of a systematic elimination of the bath degrees of freedom; the standard GLE is recovered by choosing for F(x, p) a linear function of the system coordinate *x* [13].

To focus on the interplay of space and momentum diffusion, we consider here the Markovian limit of Eqs. (2) and (3) with $\Gamma(t) = 2\delta(t)$. Under this simplifying assumption, the internal noise $\varepsilon(t)$ is replaced by the white noise $\xi(t)$ and the particle dynamics is governed by two Langevin equations (LEs), one for each canonical coordinate, *x* and *p* [30], i.e.,

$$\dot{p} = -(1 - \partial_x F \,\partial_p F)\partial_x U - (\partial_x F)^2 p/m + \partial_x F \xi(t), \qquad (4)$$

$$\dot{x} = (1 + \partial_x F \,\partial_p F)p/m - (\partial_p F)^2 \partial_x U - \partial_p F \,\xi(t).$$
(5)

Equations (4) and (5) exhibit a few unanticipated *x*-*p* coupling terms. The time derivative \dot{x} is not easy to evaluate, because the momentum *p* enters the Hamiltonian not only through the kinetic energy, but also through the coupling function F(x, p). As a consequence, the usual definition of velocity, $\dot{x} = p/m$, no longer holds and the spatial diffusion coefficient cannot be derived from the generalized Green-Kubo relation. To appreciate the implications of the above reduced LEs, we refer to the recent nonlinear friction model with $F = F(\dot{x})$ proposed by Smith and co-workers [29] to study Lévy walks in Sisyphus cooling. In their heuristic work, both the momentum-dependent cooling force and the *x*-*p* crossing terms are amiss.

Nonlinear frictions. The occurrence of nonlinear dissipative forces was explained by Klimontovich in his theory of nonlinear Brownian motion [31]. Here, to avoid technical complications, we restrict our analysis to particle diffusion in the absence of substrate potentials, that is, for U(x) = 0.

First, we investigate the remarkable kinetics of the particle subjected to a *momentum-dependent friction* with coupling function $F(x, p) = f_1(p)x = \gamma_0[1 + (p/p_s)^2]^{\frac{\mu}{2}}x$. p_s is the momentum unit and γ_0 the tunable friction strength. Equations (4) and (5) now read

$$\dot{p} = -m^{-1}f_1^2(p)p + f_1(p)\xi(t),$$

$$\dot{x} = m^{-1}p + pf_1(p)\partial_p f_1(p)x - \partial_p f_1(p)x\xi(t).$$
(6)

These LEs can be advocated to model (i) for $\mu = -1$, Sisyphus cooling of atoms trapped in an optical lattice [1], and (ii) for $\mu = 1$, particle dynamics in frictional environments at higher velocities [31]. Our numerical results yield polymerization for the former and diffusion for the latter. Both are effects of the driving force acting on the spatial coordinate *x* generated by the momentum-dependent coupling function F(x, p) [see the second term on the right-hand side of Eq. (6) for *x*]. In passing we notice that *p* is distributed according to a modified Boltzmann statistics with finite variance [32]. For $\mu < 0$ ($\mu > 0$) $\langle p^2 \rangle$ is larger (smaller) than mk_BT ; a smaller momentum variance leads to a faster spatial diffusion.



FIG. 1. (a) $\langle p^2 \rangle$, C_{xp} , and (b) $\langle x^2 \rangle$ vs *t* for a free particle, initially at rest, and subject to momentum-dependent friction $f_1(p)$ with $\mu = -1$ and different γ_0 . In both panels, the results obtained by integrating the full Eq. (6) (black squares) are compared with those obtained from the same equation after discarding the *x*-*p* corrections, i.e., setting $\dot{x} = p/m$ (red squares). Other simulation parameters are $k_BT = 1, m = 1$, and $p_s = 1$.

In Fig. 1 we plotted the time-dependence of momentum, $\langle p^2(t) \rangle$, and space variance, $\langle x^2(t) \rangle$, and the space-momentum cross-correlation function, $C_{xp} = \langle x(t)p(t)\rangle / \sqrt{\langle x^2(t)\rangle \langle p^2(t)\rangle}$, in view of an experimental validation of the model [6]. Our numerical results are compared with those obtained by ignoring the cross terms of Eq. (6), i.e., imposing $\dot{x} = p/m$ as often done in the literature. This "simplification" of Eq. (6) leads invariably to normal diffusion laws with $\alpha = 1$. By contrast, the curves $\langle x^2(t) \rangle$, obtained by integrating the full Eq. (6), approach an horizontal asymptote, which can be lowered by increasing the friction strength γ_0 . We anticipate here that when modeling cold-atom diffusion, γ_0 quantifies the depth of the optical lattice potential [11]. The additional force term in Eq. (6) can only be responsible for this instance of nodiffusion dynamics of a force-free particle [33]. Indeed, the multiplicative noise in the same equation would enhance the diffusivity of the particle [34,35]. Within our self-consistent Hamiltonian approach, the asymptotic MSD of a free particle coupled to a heat bath with momentum-dependent friction is thus determined by conflicting corrections to its spacemomentum dynamics, none of which can be discarded a priori.

Spatial-dependent friction is also a key ingredient in many studies on the XY model [36], diffusion of colloidal particles [37], nuclear fission [38], etc. In contrast with earlier heuristic treatments, which had recourse to either nonlinear coordinate-dependent frictions [39] or coordinate-dependent diffusion coefficients [34,40], we discuss here the outcome of a self-consistent approach, where the process is driven simultaneously by space-dependent friction and multiplicative noise. In the framework of our Hamiltonian scheme, one can set $\partial_x F = f_2(x) = \gamma_0 [1 + (x/x_s)^2]^{\frac{\mu}{2}}$, with $x_s = 1$ and γ_0 and μ free model parameters, to recover the LE [41],

$$\dot{p} = -m^{-1}f_2^2(x)p + f_2(x)\xi(t), \quad \dot{x} = \frac{p}{m}.$$
 (7)

Of course, the coupling function may have a more complicated form than the power law assumed here. Note that models with power-law space-dependent diffusivity [37] have been widely employed, e.g., to characterize the response of a single particle to femto-Newton forces [42]. Moreover, GLEs with power-law time-memory functions have also been proposed [20].

For our choice of $f_2(x)$, we reformulate the LE (7) as

$$m[1 + (x/x_s)^2]^{-\frac{\mu}{2}}\ddot{x} = -\gamma_0[1 + (x/x_s)^2]^{\frac{\mu}{2}}\dot{x} + \xi(t), \quad (8)$$

and estimate the asymptotic MSD for large displacements in two limiting cases:

(i) $\mu = 1$. Squaring the solution, $x\sqrt{1 + (x/x_s)^2} + x_s \ln[x/x_s + \sqrt{1 + (x/x_s)^2}] = 2\gamma_0^{-1} \int_0^t \xi(t') dt'$, of the overdamped limit of Eq. (8), $\gamma_0 \sqrt{1 + (x/x_s)^2} \dot{x} = \xi(t)$, and imposing the Gaussian approximation, $\langle x^4(t) \rangle = 3 \langle x^2(t) \rangle^2$, yield $\langle x^2(t) \rangle \sim \sqrt{t}$. This subdiffusion law has been widely investigated in various contexts, such as stochastic single files [43] and chaotic dynamical systems [44].

(ii) $\mu = -1$. The approximate identity, $\langle (x/x_s)\sqrt{1 + (x/x_s)^2} \rangle + \langle \ln[x/x_s + \sqrt{1 + (x/x_s)^2}] \rangle =$

 $(2k_BT/m)t^2/x_s^2$, which holds in the underdamped limit under the further equilibration condition, $\langle \dot{x}^2(t \to \infty) \rangle = k_BT/m$, points to a ballistic diffusive law, $\langle x^2(t) \rangle \sim t^2$. This result can be explained by noticing that as MSD grows, the friction decreases (frictionless limit), and the effective particle mass $m/f_2(x)$ increases. Moreover, in contrast with Ref. [41], the ballistic diffusion recovered here has no effect on the geometric entropic forces.

To gain further insight into the nonlocal spatiotemporal contributions to the free-particle diffusion, we ran a Monte Carlo simulation of the GLEs (2) and (3) [30] with a tunable friction exponent μ and non-Ohmic memory function $\Gamma(t)$ [45,46]. Extensive simulations confirm the analytical predictions we extracted from Eq. (7) for $\mu = \pm 1$; more in general, anomalous diffusion with any exponent $0 < \alpha \leq 2$ can be associated with an appropriate power law of the space-dependent friction [47–50]. This provides an alternative approach to anomalous diffusion.

Application to cold-atom diffusion. When modeling cold atoms diffusing in dissipative optical lattices, the classical heat bath of Eq. (1) is meant to reproduce the diffusive effects of counterpropagating laser fields [8]. Most semiclassical LEs proposed in the current literature on this topic [7-12] can be rewritten as $\dot{p} = F(p) + \sqrt{2D(p)}\xi(t)$, $\dot{x} = p/m$, with cooling force $F(p) = -\bar{\alpha}p/[1 + (p/p_s)^2]$, diffusion coefficient $D(p) = D_1 + D_2/[1 + (p/p_s)^2]$, and an appropriate choice of the parameters D_1 , D_2 , and $\bar{\alpha}$ [1]. In our approach, the friction term F(p) would correspond to choosing $F(x, p) = xf_1(p)$ with $\mu = -1$. For $D_2 = \bar{\alpha} k_B T$, the momentum-dependent dissipation and fluctuation terms in the first LE do satisfy the FDR [51], as they do in our Eq. (4). However, in sharp contrast with Eq. (5), no x-p cross terms were ever added in the LE for x, $\dot{x} = p/m$. Within the Hamiltonian framework advocated here, this may work only for fast particles (i.e., large momenta), where $\partial f_1 / \partial p \simeq 0$.

More importantly, the additional white noise with strength D_1 in the phenomenological LE for p is not associated with any dissipative term. To restore full consistency within our Hamiltonian scheme, a nonequilibrium term $-xH_e(t)$ ought to be added to the Hamiltonian of Eq. (1) [35], the sim-



FIG. 2. $\langle x^2 \rangle$ vs *t* obtained by numerically integrating Eqs. (9)–(11) for $D_2 = 0$ and (a) $D_1 = 0.5$ and different τ_c ; (b) $\tau_c = 10$ and different D_1 . All other parameters are as in Fig. 1(a). The dashed curve in (a) was obtained by neglecting the *x*-*p* cross terms; α in (b) is the fitting exponent of the asymptotic diffusion law $\langle x^2 \rangle \sim t^{\alpha}$. Other simulation parameters are m = 1, $\bar{\alpha} = 1$, and $p_s = 1$. Averages were taken over 10^4 trajectories with random initial conditions and integration time step 10^{-2} .

plest choice being $H_e(t) = \eta(t)$, with $\eta(t)$ a zero-mean valued Gaussian external noise [30].

Superdiffusion of cold ⁸⁷Rb atoms in optical lattices has been investigated experimentally [5]. It was observed that, as the lattice wells get deeper, the atom diffusion grows more sensitive to the time correlation of two competing fluctuating mechanisms, that is, spontaneous photon emission and photon absorption from the counterpropagating laser beams. To account for this effect, we model the external noise $\eta(t)$ by an Ornstein-Uhlenbeck process with correlation time τ_c . By eliminating the bath variables from the corresponding full Hamiltonian, Eq. (1), we obtain the self-consistent set of LEs [30],

$$\dot{p} = -\frac{\bar{\alpha}}{1 + (p/p_s)^2} p + \sqrt{\frac{\bar{\alpha}}{1 + (p/p_s)^2}} \xi(t) + \eta(t) + \frac{\bar{\alpha}px}{p_s^2 [1 + (p/p_s)^2]^2} \eta(t),$$
(9)

$$\dot{x} = \frac{p}{m} - \frac{\bar{\alpha}px}{p_s^2 [1 + (p/p_s)^2]^2} p + \frac{\sqrt{\bar{\alpha}px}}{p_s^2 [1 + (p/p_s)^2]^{3/2}} \xi(t) + \left(\frac{\bar{\alpha}(px)^2}{p_s^4 [1 + (p/p_s)^2]^3}\right) \eta(t),$$
(10)

$$\dot{\eta} = -\frac{1}{\tau_c}\eta(t) + \frac{1}{\tau_c}\tilde{\xi}(t),\tag{11}$$



FIG. 3. Anomalous diffusion exponent α vs D_1 for different dynamical regimes of Eqs. (9)–(11) numerically integrated in Fig. 2.

where $\langle \tilde{\xi}(t) \rangle = \langle \xi(t) \rangle = 0$, $\langle \tilde{\xi}(t) \tilde{\xi}(t') \rangle = 2D_1 \delta(t - t')$, and $\langle \xi(t) \xi(t') \rangle = 2D_2 \delta(t - t')$.

Due to the presence of strongly nonlinear coupling terms, we had recourse to a Monte Carlo algorithm to integrate Eqs. (9)-(11) [30]. Assuming large p values such as in the earlier literature [7–12], we imposed $D_2 = 0$ for $\bar{\alpha}$ constant and varied the parameters D_1 and τ_c of the external noise. In Fig. 2 we illustrate the dependence of the MSD curves $\langle x^2(t) \rangle$ on the correlation time τ_c . We found that the fitting diffusion exponent α is insensitive of τ_c for $\bar{\alpha}\tau_c > 1$. For the sake of a comparison, we also reported the results obtained from the same equations after discarding all the x-p dependent terms. The D_1 dependence of the fitting diffusion exponent of Fig. 2(b) is summarized in Fig. 3 for three distinct dynamical regimes modeled by Eqs. (9)–(11). We notice immediately that there exists a threshold value D_{th} of the constant D_1 below which diffusion is suppressed. This threshold mechanism is due to the trapping effect exerted by the additional harmonic force in Eq. (10): Diffusion sets in only for suitably large D_1 values, when the amplification action of the multiplicative noise prevails. Furthermore, our data clearly show that when the x-p cross terms in Eq. (9) are neglected, the largemomentum dynamics of the particle becomes frictionless; hence, Richardson's law with $\alpha = 3$. However, upon retaining all x-p correlated terms appearing in Eqs. (9) and (10), the particle MSD seems to exceed Richardson's law. However, this effect sets in at large D_1 , only for vanishingly small values of τ_c (white noise limit), where values $\alpha \ge 4$ have been observed. Examples of superdiffusion beyond Richardson's law have been reported for lipid bilayer membranes and actively moving biological cells [52].

Comparison with experiments. Experiments on cold-atom diffusion in optical lattices have clearly demonstrated that the anomalous diffusion exponent α is controlled by the



FIG. 4. Diffusion of cold atoms in an optical lattice: α vs Φ_{opt} (see text). Results obtained by numerically integrating Eqs. (9)–(11) for $D_1 = 1$ and $\tau_c = 10$, with (circles) and without *x*-*p* cross terms (squares). Other simulation parameters are m = 1, and $p_s = 1$.

depth of the optical potential Φ_{opt} . In particular, for deeper lattices, α may grow significantly larger than 2 [5]. Earlier theoretical works suggest that decreasing Φ_{opt} is equivalent to increasing the intensity of the external fluctuations D_1 . This connection becomes apparent in our model. The numerical integration of Eqs. (9)–(11) yields the phase diagram of Fig. 3, with two sharp transition at $D_1 = 0.2$ and $D_1 = 1.0$, in close agreement with other theoretical models [9,11,15]. Moreover, from Eq. (9) one extracts an explicit expression for the optical potential, namely, $\Phi(p) = \int^p \bar{\alpha} p' dp' / [1 + (p'^2/p_s^2)] =$ $\Phi_{\text{opt}} \ln[1 + (p/p_s)^2]$, where $\Phi_{\text{opt}} = \bar{\alpha} p_s^2/2$. The diffusion exponent α as a function of Φ_{opt} is displayed in Fig. 4 for one parameter choice of Fig. 2(b). The relevance of the x-p cross terms is remarkable at large Φ_{opt} values. Recalling that the diffusion law in our model is not very sensitive to the correlation time τ_c of the external noise, we conclude that our approach can nicely interpret recent experimental findings [5].

In summary, we have developed a consistent Hamiltonian approach to model the nonlinear coupling of a particle to a heat bath. The coupling function is a function of the particle coordinates, position and momentum, depending on the process at hand. By systematically eliminating the bath degrees of freedom, we have recovered various self-consistent Langevin equations, which resemble phenomenological equations already utilized in the literature of diffusive processes, except for the presence of additional (and so far overlooked) space-momentum cross terms. We hope the present numerical investigation of the effects of such "missing" terms can inspire further experimental validation work.

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$$\frac{\partial W}{\partial t} = \frac{\partial}{\partial p} \left[\left(f_1^2(p) \frac{p}{m} + f_1(p) \frac{\partial}{\partial p} f_1(p) \right) W \right].$$

- Its stationary solution is the modified Boltzmann distribution, $W_{\text{st}}(p) = \frac{N}{f_1(p)} \exp(-\frac{p^2}{2mk_BT})$, with *N* the appropriate normalization constant.
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