P H Y S I C A L R E V I E W L E T T E R S

Master Equation for a Quantum Particle in a Gas

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The equation for the quantum motion of a Brownian particle in a gaseous environment is derived by means of S-matrix theory. This quantum version of the linear Boltzmann equation accounts nonperturbatively for the quantum effects of the scattering dynamics and describes decoherence and dissipation in a unified framework. As a completely positive master equation it incorporates both the known equation for an infinitely massive Brownian particle and the classical linear Boltzmann equation as limiting cases.

How is the motion of a quantum particle affected by collisions with ambient gas molecules? The well-established field of quantum Brownian motion [1,2] provides an answer provided the interaction can be linearized and the particle state is close to classical. One is faced with a rather different situation if the Brownian particle is in a highly nonclassical motional state, say, due to the passage through an interferometer or the entanglement with another controlled degree of freedom. Such correlations are becoming experimentally accessible in the emerging field of molecular quantum optics, where the quantum nature of molecular motion is tested and exploited [3].

In order to assess the (partial) loss of coherence in the case of strong, nonclassical correlations in the motional state, it is necessary to provide a detailed, nonperturbative account of the microscopic scattering process. At the same time, for the effects of decoherence to be relevant at all, the gas density must be sufficiently low, so that the environmental gas can be safely taken as not self-interacting and Markovian [4]. In analogy to the classical case [5] the associated description of a *single* distinguished tracer particle within an ideal gas may be called a *linear* Boltzmann equation. It should not be confused with the linearized Boltzmann equations for the reduced single particle-gas state, obtained perturbatively from its multiparticle description.

The investigation of the loss of coherence due to gas collisions was initiated by Joos and Zeh [6], who considered the limiting case of an infinitely massive tracer particle. This theory, which was later refined [7,8] and tested experimentally [9], describes the pure spatial "localization" of an extended coherent matter wave into a mixture with reduced spatial coherence, but it cannot account for dissipation. The situation is much more involved if the tracer mass M is finite and comparable to the gas mass m so that the ratio m/M must not be neglected. In this case localization occurs both in position and in momentum [10], and the appropriate kinetic equation must describe the full interplay of decohering and thermalizing dynamics.

So far, the most important advancement in this direction is the proposal by Diósi [11] of an equation based on a combination of scattering theory and heuristic arguments. A more recent development is the theory by Vacchini [12] in terms of the dynamic structure factor of the medium, an approach limited to the (weak coupling) Born approximation, like those in [13,14].

This Letter presents the full quantum version of the linear Boltzmann equation, describing the whole range of collisional effects from decoherence to dissipation. It provides a transparent and stringent derivation, and a discussion of its implications and limits. The only essential premise is the Markov assumption, which implies—in the spirit of Boltzmann's classic derivation—that both the rate and the effect of individual two-particle scattering events are separately physically meaningful, while subsequent collisions with the "same" gas molecule are negligibly unlikely. Accordingly, the gas may be taken to be ideal (not self-interacting), stationary (diagonal in momentum), and uniform in position space, thus covering, e.g., thermal states of bosons and fermions, but no liquids.

In operator form the master equation reads $\partial_t \rho = (i\hbar)^{-1}[\mathsf{H}, \rho] + \mathcal{L}\rho$, with $\mathsf{H} = \mathsf{P}^2/(2M)$ the free Hamiltonian and

$$\mathcal{L}\rho = \int d\boldsymbol{Q} \int_{Q^\perp} \frac{d\boldsymbol{K}}{Q} \left\{ \mathsf{L}_{Q,K}\rho \mathsf{L}_{Q,K}^\dagger - \frac{1}{2}\rho \mathsf{L}_{Q,K}^\dagger \mathsf{L}_{Q,K} - \frac{1}{2}\mathsf{L}_{Q,K}^\dagger \mathsf{L}_{Q,K}\rho \right\}. \quad (1)$$

Here the integration is over all momentum transfers $\boldsymbol{Q}$, and for fixed $\boldsymbol{Q}$ also over the perpendicular plane $\boldsymbol{Q}^\perp = \{\boldsymbol{K} \in \mathbb{R}^3 : \boldsymbol{K} \cdot \boldsymbol{Q} = 0\}$. The Lindblad (jump) operators have the form

$$\mathsf{L}_{Q,K} = e^{i\mathsf{R}\cdot\boldsymbol{Q}/\hbar}F(\boldsymbol{K}, \mathsf{P}; \boldsymbol{Q}) \quad (2)$$

where R and P are the position and the momentum operator of the Brownian tracer particle. The function F, which is operator valued in (2), contains all the details of the collisional interaction with the gas. It involves the elastic scattering amplitude $f(\boldsymbol{p}_{\text{out}}, \boldsymbol{p}_{\text{in}})$, the momentum distribution function $\mu(\boldsymbol{p})$ of the gas [15], and its number density n_{gas}. It is convenient to denote relative momenta by

$$\mathrm{rel}\,(\boldsymbol{p}, \boldsymbol{P}) := \frac{m_*}{m}\boldsymbol{p} - \frac{m_*}{M}\boldsymbol{P},$$

with $m_* = mM/(M + m)$ the reduced mass. Moreover, for given $\boldsymbol{Q} \neq 0$ let us denote the parallel and the perpendicular contribution of a vector (operator) $\boldsymbol{P}$ by $\boldsymbol{P}_{\|\boldsymbol{Q}} = (\boldsymbol{P}\cdot\boldsymbol{Q})\boldsymbol{Q}/Q^2$ and by $\boldsymbol{P}_{\perp\boldsymbol{Q}} = \boldsymbol{P} - \boldsymbol{P}_{\|\boldsymbol{Q}}$, respectively. With these definitions

$$F(\boldsymbol{K}, \boldsymbol{P}; \boldsymbol{Q}) = \frac{\sqrt{n_{\mathrm{gas}}m}}{m_*}f\left(\mathrm{rel}(\boldsymbol{K}_{\perp\boldsymbol{Q}}, \boldsymbol{P}_{\perp\boldsymbol{Q}}) - \frac{\boldsymbol{Q}}{2}, \mathrm{rel}(\boldsymbol{K}_{\perp\boldsymbol{Q}}, \boldsymbol{P}_{\perp\boldsymbol{Q}}) + \frac{\boldsymbol{Q}}{2}\right)\mu\left[\boldsymbol{K}_{\perp\boldsymbol{Q}} + \left(1 + \frac{m}{M}\right)\frac{\boldsymbol{Q}}{2} + \frac{m}{M}\boldsymbol{P}_{\|\boldsymbol{Q}}\right]^{1/2}. \quad (3)$$

This implies that both the scattering amplitude and the distribution function attain an operator character in (2), and that the particle momentum contributes with the part perpendicular to the momentum exchange to the former and with the parallel one to the latter. For physically reasonable interactions, the function F decreases sufficiently fast as $Q \to 0$ so that (1) is well defined.

Note that the form of $\mathcal{L}\rho$ fits the general structure of a translation-invariant and completely positive master equation, as characterized by Holevo [16] (see [17] for a discussion), although the summation in Ref. [16] is here replaced by the integrations in (1). We will see below that the master equation assumes a more intuitive form in the momentum representation.

My first aim is to provide a derivation of (1)–(3). To that end, let us first define the positive operator Γ which yields the total collision rate. As in classical mechanics, the rate is determined by the gas density, the modulus of the relative velocity $v = |\mathrm{rel}(\boldsymbol{p}, \boldsymbol{P})|/m_*$, and the total scattering cross section $\sigma(\boldsymbol{p}_{\mathrm{in}})$. Denoting the improper momentum eigenvectors of tracer and gas by $|\boldsymbol{P}\rangle$ and $|\boldsymbol{p}\rangle$ we have

$$\Gamma = \int \mathrm{d}\boldsymbol{P}\mathrm{d}\boldsymbol{p}\, n_{\mathrm{gas}}v(\boldsymbol{p}, \boldsymbol{P})\sigma(\mathrm{rel}(\boldsymbol{p}, \boldsymbol{P}))|\boldsymbol{P}\rangle\langle\boldsymbol{P}| \otimes |\boldsymbol{p}\rangle\langle\boldsymbol{p}|.$$

Indeed, for separable particle-gas states the expectation value of Γ yields the average total collision rate experienced by the Brownian particle.

Let us now see how a single collision changes the motional state of the tracer particle according to scattering theory. If tracer and gas are uncorrelated before the collision, the outgoing tracer state is $\rho' = \mathrm{Tr}_{\mathrm{gas}}(\mathsf{S}[\rho \otimes \rho_{\mathrm{gas}}]\mathsf{S}^\dagger)$, where $\mathsf{S} = \mathsf{I} + i\mathsf{T}$ is the two-particle scattering operator and a partial trace over the gas has to be performed. Employing unitarity, $\mathsf{S}^\dagger\mathsf{S} = \mathsf{I}$, one can express the change of the state $\Delta\rho = \rho' - \rho$ as

$$\Delta\rho = \frac{i}{2}\mathrm{Tr}_{\mathrm{gas}}([\mathsf{T} + \mathsf{T}^\dagger, \rho \otimes \rho_{\mathrm{gas}}]) + \mathrm{Tr}_{\mathrm{gas}}(\mathsf{T}[\rho \otimes \rho_{\mathrm{gas}}]\mathsf{T}^\dagger)$$
$$- \frac{1}{2}\mathrm{Tr}_{\mathrm{gas}}(\mathsf{T}^\dagger\mathsf{T}[\rho \otimes \rho_{\mathrm{gas}}] + [\rho \otimes \rho_{\mathrm{gas}}]\mathsf{T}^\dagger\mathsf{T}) \quad (4)$$

The first term generates a constant coherent modification of the unitary evolution and can be absorbed in the Hamiltonian H. This energy shift due to "forward scattering" is usually accounted for by a modified index of refraction [18] and can be disregarded since the gas density is uniform.

The momentum representation of the remaining incoherent part of (4) can be expressed entirely in terms of the kernel

$$\langle\boldsymbol{P}|\mathrm{Tr}_{\mathrm{gas}}(\mathsf{T}[|\boldsymbol{P}_0\rangle\langle\boldsymbol{P}'_0| \otimes \rho_{\mathrm{gas}}]\mathsf{T}^\dagger)|\boldsymbol{P}'\rangle. \quad (5)$$

If the momentum-diagonal representation of the gas state

$$\rho_{\mathrm{gas}} = \frac{(2\pi\hbar)^3}{\Omega}\int \mathrm{d}\boldsymbol{p}_0\mu(\boldsymbol{p}_0)|\boldsymbol{p}_0\rangle\langle\boldsymbol{p}_0|, \quad (6)$$

is inserted, the kernel (5) assumes the simple form $\delta(\boldsymbol{P} - \boldsymbol{P}_0 - \boldsymbol{P}' + \boldsymbol{P}'_0)J(\boldsymbol{P}, \boldsymbol{P}'; \boldsymbol{P} - \boldsymbol{P}_0)$, where the function

$$J(\boldsymbol{P}, \boldsymbol{P}'; \boldsymbol{Q}) = \frac{(2\pi\hbar)^3}{\Omega}\int \mathrm{d}\boldsymbol{p}_0\mu(\boldsymbol{p}_0)\langle\mathrm{rel}(\boldsymbol{p}_0 - \boldsymbol{Q}, \boldsymbol{P})|$$
$$\times \mathsf{T}_0|\mathrm{rel}(\boldsymbol{p}_0, \boldsymbol{P} - \boldsymbol{Q})\rangle\langle\mathrm{rel}(\boldsymbol{p}_0, \boldsymbol{P}' - \boldsymbol{Q})|$$
$$\times \mathsf{T}_0^\dagger|\mathrm{rel}(\boldsymbol{p}_0 - \boldsymbol{Q}, \boldsymbol{P}')\rangle \quad (7)$$

is given in terms of the single particle operator T_0 for the relative coordinates. Its momentum matrix elements are related to the scattering amplitude by $\langle\boldsymbol{p}_f|\mathsf{T}_0|\boldsymbol{p}_i\rangle = (\pi\hbar)^{-1}\delta(p_f^2 - p_i^2)f(\boldsymbol{p}_f, \boldsymbol{p}_i)$ [19]. However, inserting this into (7) one arrives at an ill-defined expression, which involves the arbitrary normalization volume Ω and, for $\boldsymbol{P} = \boldsymbol{P}'$, the *square* of a δ-function. As is well understood, the reason for this is our use of a momentum representation of ρ_{gas} in (5). Plane waves are not in the domain of the operator $\mathsf{S}_0 = \mathsf{I}_0 + i\mathsf{T}_0$, which maps incoming asymptotes to outgoing ones (and, by extension, leaves the outgoing ones invariant).

A possible resort would be therefore to choose a representation of ρ_{gas} which permits a decomposition into in-states and out-states. Indeed, a decomposition into Gaussian wave packets admits a conventional but tedious calculation of the state change in the limiting case of an infinitely massive Brownian particle, $m/M = 0$, as demonstrated in [8]. In the same article it is shown that the identical result can be obtained directly by keeping the diagonal representation. In this case the extension of T_0, beyond the physically acceptable domain of incoming states, must be complemented by a consistent, physically motivated replacement rule:

$$\frac{(2\pi\hbar)^3}{\Omega}|\langle\boldsymbol{p}_f|\mathsf{T}_0|\boldsymbol{p}_i\rangle|^2 \to \delta\left(\frac{p_f^2 - p_i^2}{2}\right)\frac{|f(\boldsymbol{p}_f, \boldsymbol{p}_i)|^2}{\sigma(\boldsymbol{p}_i)|\boldsymbol{p}_i|}. \quad (8)$$

In the present case of a finite mass ratio this yields a well-

defined kernel (7) for $P = P'$, and thus implies $\int dQ \tilde{J}(P + Q, P + Q; Q) = 1$, which ensures the conservation of the norm of ρ'. For $P \neq P'$ an extension of the rule (8) to different pairs of incoming and outgoing relative momenta is required. It can be constructed at no additional cost if the momentum change of the two pairs is the same, as is the case in

$$X = \frac{(2\pi\hbar)^3}{\Omega} \langle p_f + q | T_0 | p_i + q \rangle \langle p_i - q | T_0^\dagger | p_f - q \rangle g(q)$$

with arbitrary positive function g. Inserting the formal square root of the replacement rule (8) yields the square root of a product of two energy conserving δ-functions with arguments $\frac{p_f^2 - p_i^2}{2} \pm (p_f - p_i) \cdot q$. They imply that the parallel component $q_\parallel \equiv q_{\parallel(p_f - p_i)}$ of the momentum separation must be zero, which restricts q integrations to the plane perpendicular to the momentum change $p_f - p_i$. This restriction is equally effected by replacing the vectors q with their projection $q_\perp \equiv q - q_\parallel$ onto that plane. This way the form of the arguments in each individual scattering amplitude already ensures the conservation of energy and one is left with a single proper Dirac function $\delta(\frac{p_f^2 - p_i^2}{2})$. Hence, as a natural generalization of (8), we have

$$X \rightarrow \delta\left(\frac{p_f^2 - p_i^2}{2}\right) \frac{f(p_f + q_\perp, p_i + q_\perp)}{\sqrt{\sigma(p_i + q_\perp)|p_i + q_\perp|}}$$
$$\times \frac{f^*(p_f - q_\perp, p_i - q_\perp)}{\sqrt{\sigma(p_i - q_\perp)|p_i - q_\perp|}} g(q_\perp). \tag{9}$$

It turns into the known replacement rule as $q \rightarrow 0$. The only freedom in this construction is a possible phase factor from taking the roots, but symmetry considerations lead to the above choice of no additional phase.

Being able to evaluate traces with a momentum-diagonal representation of the gas, we can now combine the operator T for the effect of a single collision with the rate operator Γ to obtain the temporal evolution (in interaction picture putting aside the contribution of H). The temporal change $\partial_t \rho$ is obtained from $\Delta\rho$ in (4) by replacing T with $T\Gamma^{1/2}$,

$$\partial_t \rho \cong \mathrm{Tr}_{\mathrm{gas}}(T\Gamma^{1/2}[\rho \otimes \rho_{\mathrm{gas}}]\Gamma^{1/2}T^\dagger)$$
$$- \frac{1}{2}\mathrm{Tr}_{\mathrm{gas}}(\Gamma^{1/2}T^\dagger T\Gamma^{1/2}[\rho \otimes \rho_{\mathrm{gas}}])$$
$$- \frac{1}{2}\mathrm{Tr}_{\mathrm{gas}}([\rho \otimes \rho_{\mathrm{gas}}]\Gamma^{1/2}T^\dagger T\Gamma^{1/2}). \tag{10}$$

This is suggested by a quantum trajectory unravelling of $\partial_t \rho$ [20], where each trajectory is first weighted by the probability of a collision event to take place in an infinitesimal time interval before being scattered. To obtain the momentum representation of (10) we have to evaluate

$$\langle P | \mathrm{Tr}_{\mathrm{gas}}(T\Gamma^{1/2}[|P_0\rangle\langle P_0'| \otimes \rho_{\mathrm{gas}}]\Gamma^{1/2}T^\dagger) | P' \rangle. \tag{11}$$

Using again the momentum-diagonal gas state (6) re-

duces the kernel (11) to the form $\delta(P - P_0 - P' + P_0')M_{\mathrm{in}}(P, P'; P - P_0)$. It follows that the momentum representation of (10) is

$$\partial_t \rho(P, P') = \int dQ M_{\mathrm{in}}(P, P'; Q)\rho(P - Q, P' - Q)$$
$$- \frac{1}{2}[M_{\mathrm{out}}^{\mathrm{cl}}(P) + M_{\mathrm{out}}^{\mathrm{cl}}(P')]\rho(P, P') \tag{12}$$

with

$$M_{\mathrm{in}}(P, P'; Q) = \int dp_0 \frac{(2\pi\hbar)^3}{\Omega} \mu(p_0)\frac{n_{\mathrm{gas}}}{m_*}$$
$$\times \sqrt{|p_i + q|\sigma(p_i + q)}$$
$$\times \sqrt{|p_i - q|\sigma(p_i - q)}\langle p_f + q | T_0 | p_i + q \rangle$$
$$\times \langle p_i - q | T_0^\dagger | p_f - q \rangle$$

and $M_{\mathrm{out}}^{\mathrm{cl}}(P) := \int dQ M_{\mathrm{in}}(P + Q, P + Q; Q)$. Here I introduced $p_i := \mathrm{rel}(p_0, \frac{P+P'}{2} - Q)$ and $p_f := p_i - Q$ as functions of p_0, and defined $q := \mathrm{rel}(0, \frac{P-P'}{2})$.

In order to evaluate M_{in}, the integration is now transformed to dp_i. Incidentally, this suggests a natural factorization of the μ distribution into a product of square roots, $\sqrt{\mu(p_0)\mu(p_0)}$, since p_0 can be equally expressed as a function of P or of P'. Applying the replacement rule (9) projects q to $q_\perp \equiv q_{\perp(p_f - p_i)}$ not only in the scattering amplitudes, but also in the argument of μ. One obtains the well-defined expression

$$M_{\mathrm{in}}(P, P'; Q) = \frac{n_{\mathrm{gas}}}{m_*}\left(\frac{m}{m_*}\right)^3 \int dp_i \delta\left(\frac{p_f^2 - p_i^2}{2}\right)$$
$$\times f(p_f + q_\perp, p_i + q_\perp)$$
$$\times f^*(p_f - q_\perp, p_i - q_\perp)$$
$$\times \mu^{1/2}\left(p_i + \frac{m}{M}(p_f + P) + \frac{m}{m_*}q_\perp\right)$$
$$\times \mu^{1/2}\left(p_i + \frac{m}{M}(p_f + P') - \frac{m}{m_*}q_\perp\right).$$

As the last step, the transformation $p_i \rightarrow K = \frac{m}{m_*}p_i + \frac{m}{M} \times \frac{P_{\perp Q} + P_{\perp Q}'}{2} - \frac{m}{m_*}\frac{Q}{2}$ factorizes the integrand into P and P' contributions,

$$M_{\mathrm{in}}(P, P'; Q) = \int dK \delta(K \cdot Q)F(K, P - Q; Q)$$
$$\times F^*(K, P' - Q; Q), \tag{13}$$

with F given in (3). By returning to the Schrödinger picture and noting $\int dK \delta(K \cdot Q) = Q^{-1}\int_{Q^\perp} dK$ one finds that (12) with (13) is the momentum representation of (1), which closes its derivation. Note that the form of $\mathcal{L}$ is a consequence only of the premises and the replacement (8).

The physics described by (1) is easy to discuss in the momentum basis (12). We will see that $M_{\text{out}}^{\text{cl}}(\boldsymbol{P})$ is the rate of a classical particle with momentum $\boldsymbol{P}$ to be scattered by the gas into a different direction or velocity. Hence, the second line in (12) effects a reduction of the coherences $\rho(\boldsymbol{P}, \boldsymbol{P}' \neq \boldsymbol{P})$ determined by the arithmetic mean of the corresponding momenta on the diagonal. The first line in (12), on the other hand, may reduce or produce coherences. While the complex quantity M_{in} cannot be decomposed into classical rates, it is reassuring that it involves an integration over *all* possible, in general nonparallel, pairs of two-particle scattering trajectories which end at the Brownian momenta $(\boldsymbol{P}, \boldsymbol{P}')$, each part conserving the total energy and the momentum with an exchange of $\boldsymbol{Q}$, and weighted by the thus restricted distribution of available gas momenta. (Diósi's equation [11], in comparison, involves the differential cross section $d\sigma/d\Omega = |f|^2$ and therefore at most pairs of identical trajectories.)

After a time long, compared to the time scale of decoherence, the motional state is expected to be practically indistinguishable from a classical state. As such, it should be characterized by the momentum distribution $w(\boldsymbol{P}) = \rho(\boldsymbol{P}, \boldsymbol{P})$ alone, and one expects that the motion of the diagonal elements predicted by (1) is equal to the classical linear Boltzmann equation. Indeed, one obtains from (12)

$$\partial_t w(\boldsymbol{P}) = \int d\boldsymbol{Q} M_{\text{in}}^{\text{cl}}(\boldsymbol{P}, \boldsymbol{Q}) w(\boldsymbol{P} - \boldsymbol{Q}) - M_{\text{out}}^{\text{cl}}(\boldsymbol{P}) w(\boldsymbol{P}),$$
(14)

where $M_{\text{out}}^{\text{cl}}(\boldsymbol{P}) = \int d\boldsymbol{Q} M_{\text{in}}^{\text{cl}}(\boldsymbol{P} + \boldsymbol{Q}; \boldsymbol{Q})$ and

$$M_{\text{in}}^{\text{cl}}(\boldsymbol{P}; \boldsymbol{Q}) = \frac{n_{\text{gas}}}{m_*} \int d\boldsymbol{K} \mu(\boldsymbol{K}) \delta\left(\frac{p_{\text{cf}}^2 - p_{\text{ci}}^2}{2}\right) \frac{d\sigma}{d\Omega}(\boldsymbol{p}_{\text{cf}}, \boldsymbol{p}_{\text{ci}})$$
(15)

is the rate density of a classical Brownian particle to be scattered into momentum $\boldsymbol{P}$ upon a momentum exchange of $\boldsymbol{Q}$. Here $\boldsymbol{p}_{\text{ci}} \equiv \text{rel}(\boldsymbol{K}, \boldsymbol{P} - \boldsymbol{Q})$, $\boldsymbol{p}_{\text{cf}} \equiv \boldsymbol{p}_{\text{ci}} - \boldsymbol{Q}$, and $d\sigma/d\Omega = |f|^2$. Note that the classic form [5] of (14) is obtained by transforming the $\boldsymbol{Q}$ integrals over the Dirac δ into angular integrations over the relative momentum direction. It follows that the stationary solution of (1) is given, for thermal gas states μ, by the corresponding (momentum-diagonal) thermal Brownian state of the classical equation, and that the H theorem applies.

Another border case of (1) is the limit of an infinitely massive tracer. A short calculation confirms that letting m/M approach zero reduces $\mathcal{L}\rho$ to the (corrected version [8] of the) master equation by Gallis and Fleming [7], which attributes the loss of coherence to the amount of position information gained by the colliding gas. Finally, (1) assumes the form of Vacchini's equation [12] if one replaces the true scattering amplitude f in (3) by its Born

approximation f_B (which depends only on the momentum transfer).

In conclusion, I presented the quantum version of the linear Boltzmann equation. It unifies, in the form of a completely positive master equation, the decohering and dissipative dynamics in the motion of a Brownian particle, and it comprises various known dynamic behaviors as limiting forms.

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