

Quantum linear Boltzmann equation with finite intercollision time

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(Received 1 June 2009; published 17 December 2009)

Inconsistencies are pointed out in the usual quantum versions of the classical linear Boltzmann equation constructed for a quantized test particle in a gas. These are related to the incorrect formal treatment of momentum decoherence. We prove that ideal collisions with the molecules would result in complete momentum decoherence, the persistence of coherence is only due to the finite intercollision time. A corresponding quantum linear Boltzmann equation is proposed.

DOI: [10.1103/PhysRevA.80.064104](https://doi.org/10.1103/PhysRevA.80.064104)

PACS number(s): 03.65.Yz, 05.60.Gg, 03.65.Ta, 05.20.Dd

I. INTRODUCTION

Collisional theory of motion of a test particle through a gas had been the oldest classical tool confirming atomic kinetics—e.g., in Brownian motion [1]—or using atomic kinetics—e.g., in Millikan’s experiment [2]. If the gas is dilute and hot, the interaction of the particle with the gas reduces to the repeated independent binary collisions between the particle and the individual molecules. The dynamics of the test particle is fully captured by the differential cross section of the scattering: the linear Boltzmann equation (LBE) describes the particle’s motion. For a quantum particle, the scattering matrix $\hat{1} + i\hat{T}$ is expected to capture the dynamics which we describe by the quantum linear Boltzmann equation (QLBE)

$$\frac{d\hat{\rho}}{dt} = -i[\hat{H}, \hat{\rho}] + \mathcal{L}\hat{\rho}, \quad (1)$$

where $\hat{\rho}$ is the density matrix, \hat{H} is the free Hamiltonian (including an energy shift due to the gas), and \mathcal{L} is the Lindblad [3] superoperator of the non-Hamiltonian evolution. The microscopic derivation of \mathcal{L} does not differ from the classical derivation of the LBE as long as we assume a density matrix $\langle \mathbf{P} | \hat{\rho} | \mathbf{P}' \rangle = \rho(\mathbf{P}) \delta(\mathbf{P} - \mathbf{P}')$ without superpositions in momentum \mathbf{P} . The evolution of the diagonal density matrix is governed by the classical LBE applied to the distribution $\rho(\mathbf{P})$. This is why the classical kinetic theory has so far served as a perfect effective theory for a quantum particle in those innumerable applications where complete momentum decoherence was *a priori* assumed. A stealthy request for a systematic theory emerged in the field of foundations where quantum Brownian motion became the paradigm of how the environment (gas) “measures” the wave function (of the particle) [4–7]. For experiments, a proper quantum Brownian particle was not available before, now it is, cf., e.g., [8].

When we are interested in the local quantum dynamics of the particle, we need the off-diagonal elements $\langle \mathbf{P} | \hat{\rho} | \mathbf{P}' \rangle$ as well, and then the microscopic derivation of \mathcal{L} becomes problematic. The classical derivation resists to a direct quantum extension. The first microscopic derivation was obtained fifteen years ago [9] by the present author. Subsequent works

[10–12] by Vacchini and Hornberger—flavored by transient debates [13,14]—led them to a refined version. Their up-to-date review [15] further elucidates the subject and can serve as an exhaustive source of references for the reader. Microscopic derivations assume typically (though not necessarily) the Maxwell-Boltzmann distribution for the molecular momentum \mathbf{k}

$$\rho_g(\mathbf{k}) = \left(\frac{\beta}{2\pi m} \right)^{3/2} \exp\left(-\frac{\beta \mathbf{k}^2}{2m} \right), \quad (2)$$

where m is the molecular mass and β is the inverse temperature. To overcome the mentioned difficulties with the off-diagonal elements of $\hat{\rho}$, Ref. [9] suggested a formal resolution that became an irreducible part of all subsequent derivations [10–15]. In all variants of QLBE, including a most recent one [16], \mathcal{L} is not a linear functional of $\rho_g(\mathbf{k})$ as it should have been, rather, \mathcal{L} is a quadratic functional of the *square root* of $\rho_g(\mathbf{k})$. In other words, \mathcal{L} is the linear functional of

$$\sqrt{\rho_g(\mathbf{k})\rho_g(\mathbf{k}')} . \quad (3)$$

Therefore, no matter which QLBE is considered, we face the following issue of consistency. Suppose the gas has two independent “components” consisting of the same molecules at same densities while at two different temperatures. Then, their contribution to the QLBE must be additive. But it is not, since \mathcal{L} in the QLBE (1) is linear in the expression (3), not in $\rho_g(\mathbf{k})$ itself.

Before we construct a QLBE that is linear in $\rho_g(\mathbf{k})$, we point out a surprising collisional decoherence effect apparently overlooked for a long time and discovered independently from the present work by Kamleitner and Cresser, too [16]. It may radically alter our understanding what QLBE should be.

A. Single collision decoheres momentum

Let us consider the kinematics of a single collision between the particle and a molecule. As a Gedanken experiment, suppose we prepared the molecule with momentum \mathbf{k} before the collision and we detected it with momentum $\mathbf{k} - \mathbf{Q}$ after the collision, i.e., we know the momentum transfer \mathbf{Q} . Note that the interactions of these preparation and detection devices with the molecule are irrelevant for the reduced

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dynamics of the particle, yet such preparation and detection devices are relevant for our arguments. We get dramatic conclusions for the reduced state $\hat{\rho}$ of the particle since, for elementary kinematic reasons, the observation of \mathbf{Q} is equivalent with the observation of one component of the particle's momentum \mathbf{P} . Let \mathbf{P} stand for the postcollision momentum, then from energy conservation

$$\frac{\mathbf{k}^2}{2m} + \frac{(\mathbf{P} - \mathbf{Q})^2}{2M} = \frac{(\mathbf{k} - \mathbf{Q})^2}{2m} + \frac{\mathbf{P}^2}{2M}, \quad (4)$$

we have the following relationship:

$$\mathbf{P}_{\parallel} = \frac{M}{m} \mathbf{k}_{\parallel} - \left(\frac{M}{m} - 1 \right) \frac{\mathbf{Q}}{2}, \quad (5)$$

where the subscripts \parallel refer to the components parallel to \mathbf{Q} . It is obvious that after our single collision the particle's density matrix $\hat{\rho}$, whatever it was before the collision, becomes perfect diagonal in \mathbf{P}_{\parallel} . Gradually, after many collisions, the state $\hat{\rho}$ becomes a mixture of plane waves, no off-diagonal mechanism will be left at all. This result contradicts to what we have so far learned as QLBE. The art of deriving a plausible QLBE has always been about the nontrivial treatment of the off-diagonal elements. The above decoherence mechanism, however, shows that standard scattering theory and independent collisions do not yield any QLBE for the off-diagonals. The apparent off-diagonal dynamics may well have been artifacts of the square-root phenomenology (3).

In reality, the state of the particle is not necessarily the delocalized one advocated by our decoherence argument. In fact, our argument is relevant for the case of very dilute gas or for very small collision cross sections. Then, the density matrix has no off-diagonal elements at all. Such perfect momentum decoherence assumes perfect energy conservation of the collision process. In quantum scattering, it requires infinite time and this is the loophole where we concentrate. If the intercollision time is moderate long, it still leaves ample room for energy balance uncertainty and momentum coherence.

B. QLBE with finite intercollision time

We outline the derivation of QLBE (1) where \mathcal{L} remains a linear functional of $\rho_g(\mathbf{k})$ and momentum decoherence is described correctly in function of the intercollision time τ . The full quantum derivation introduces the box-normalized single-molecule density matrix

$$\hat{\rho}_g = \frac{(2\pi)^3}{V} \rho_g(\hat{\mathbf{k}}). \quad (6)$$

We follow Ref. [9]. Starting from the precollision initial state $\hat{\rho} \otimes \hat{\rho}_g$, we calculate the collisional change of the particle's reduced state. The parts responsible for in- and out-scattering are respectively given by the following expressions:

$$\Delta \hat{\rho}|_{\text{in}} = V n_g \text{tr}_g \{ \hat{T} \hat{\rho} \otimes \hat{\rho}_g \hat{T}^\dagger \}, \quad (7)$$

$$\Delta \hat{\rho}|_{\text{out}} = -\frac{1}{2} V n_g \text{tr}_g \{ \hat{T}^\dagger \hat{T} \cdot \hat{\rho} \otimes \hat{\rho}_g \}, \quad (8)$$

where the single-collision contributions has been enhanced by the total number $V n_g$ of molecules at density n_g . The standard form of the transition operator for the (spin-independent) scattering with initial laboratory momenta \mathbf{k} and \mathbf{P} , respectively, reads

$$\hat{T} = \frac{1}{2\pi m^*} \int d\mathbf{k} d\mathbf{Q} e^{i\mathbf{Q}\hat{\mathbf{x}}} f(\hat{\mathbf{k}}_f^*, \hat{\mathbf{k}}_i^*) \delta(\hat{E}_{fi}^*) |\mathbf{k} - \mathbf{Q}\rangle \langle \mathbf{k}|, \quad (9)$$

where the ket and bra stand for the momentum eigenstates of the molecule. The function f is the scattering amplitude. The quantities marked by $*$ are the initial and final (pre- and postcollision) c.o.m. momenta, while \hat{E}_{fi}^* is the c.o.m. energy balance, respectively,

$$\hat{\mathbf{k}}_i^* = \frac{M}{M^*} \mathbf{k} - \frac{m}{M^*} \hat{\mathbf{P}},$$

$$\hat{\mathbf{k}}_f^* = \hat{\mathbf{k}}_i^* - \mathbf{Q},$$

$$\hat{E}_{fi}^* = \frac{1}{2m^*} (\hat{\mathbf{k}}_f^*)^2 - \frac{1}{2m^*} (\hat{\mathbf{k}}_i^*)^2. \quad (10)$$

Here, $M^* = M + m$ and $m^* = Mm/M^*$. We substitute the Eqs. (6) and (9) into Eq. (7), yielding

$$\Delta \hat{\rho}|_{\text{in}} = \frac{2\pi n_g}{m^{*2}} \int \int d\mathbf{k} \rho_g(\mathbf{k}) d\mathbf{Q} e^{i\mathbf{Q}\hat{\mathbf{x}}} \times f(\hat{\mathbf{k}}_f^*, \hat{\mathbf{k}}_i^*) \delta(\hat{E}_{fi}^*) \hat{\rho} \bar{f}(\hat{\mathbf{k}}_f^*, \hat{\mathbf{k}}_i^*) \delta(\hat{E}_{fi}^*) e^{-i\mathbf{Q}\hat{\mathbf{x}}}. \quad (11)$$

A similar elaboration applies to $\Delta \hat{\rho}|_{\text{out}}$ as well. And now, we alter (and simplify) the old derivation [9]. In standard quantum scattering theory, both the preparation of precollision and the emergence of postcollision states take infinite long time and the transition operator (9) contains the δ function for perfect energy conservation. In our case, however, the intercollision time τ is finite

$$\tau = \frac{\sqrt{\pi \beta m}}{\sigma n_g}, \quad (12)$$

σ is the total scattering cross section. We take this feature into the account if in \hat{T} , we use the "smoothened" $\delta_\tau(E)$ instead of $\delta(E)$

$$\delta_\tau(E) = \frac{\sin(\tau E/2)}{\pi E}, \quad (13)$$

and we extend the transition amplitude $f(\hat{\mathbf{k}}_f^*, \hat{\mathbf{k}}_i^*)$ off-shell. We shall approximate $d\hat{\rho}/dt|_{\text{in}}$ by $\Delta \hat{\rho}|_{\text{in}}/\tau$, when we substitute Eq. (11), it yields

$$\frac{d\hat{\rho}}{dt} \Big|_{\text{in}} = \frac{2\pi n_g}{\pi m^{*2}} \int \int d\mathbf{k} \rho_g(\mathbf{k}) d\mathbf{Q} e^{i\mathbf{Q}\hat{\mathbf{x}}} \times f(\hat{\mathbf{k}}_f^*, \hat{\mathbf{k}}_i^*) \delta_\tau(\hat{E}_{fi}^*) \hat{\rho} \bar{f}(\hat{\mathbf{k}}_f^*, \hat{\mathbf{k}}_i^*) \delta_\tau(\hat{E}_{fi}^*) e^{-i\mathbf{Q}\hat{\mathbf{x}}}. \quad (14)$$

This form, and a similar form for $d\hat{\rho}/dt|_{\text{out}}$, give $d\hat{\rho}/dt$

$=d\hat{\rho}/dt|_{\text{in}}+d\hat{\rho}/dt|_{\text{out}}$ and explicitly indicate the following Lindblad structure (1) of our QLBE:

$$\begin{aligned} \frac{d\hat{\rho}}{dt} = & -i[\hat{H}, \hat{\rho}] + \frac{2\pi n_g}{\tau m^*2} \int \int d\mathbf{k} \rho_g(\mathbf{k}) d\mathbf{Q} \left(\hat{V}(\mathbf{k}, \mathbf{Q}) \hat{\rho} \hat{V}^\dagger(\mathbf{k}, \mathbf{Q}) \right. \\ & \left. - \frac{1}{2} \{ \hat{V}^\dagger(\mathbf{k}, \mathbf{Q}) \hat{V}(\mathbf{k}, \mathbf{Q}), \hat{\rho} \} \right), \end{aligned} \quad (15)$$

where the Lindblad operators read

$$\hat{V}(\mathbf{k}, \mathbf{Q}) = e^{i\mathbf{Q}\hat{\mathbf{X}}} f(\hat{\mathbf{k}}_f^*, \hat{\mathbf{k}}_i^*) \delta_\tau(\hat{E}_{fi}^*). \quad (16)$$

The cautious reader might notice that the *average* intercollision time τ depends on $\rho_g(\mathbf{k})$, so that \mathcal{L} 's exact linear dependence on $\rho_g(\mathbf{k})$ is arguable. Whether a refined means of including the random intercollision time exists, we do not know yet. We expect the following features of the new QLBE. In very dilute gas, we can take $\tau \rightarrow \infty$ and $\delta_\tau \rightarrow \delta$, the off-diagonal elements of $\hat{\rho}$ become completely suppressed and our QLBE recovers the standard LBE for the diagonal elements of the density matrix. If the gas is less dilute, the LBE remains valid for the diagonal part of $\hat{\rho}$ while the off-diagonal part reappears and then τ becomes the relevant quantity for it. We are going to illustrate these features.

C. Diffusion limit

A common test of QLBE is Dekker's equation [17] containing the phenomenological constants η , D_{pp} , and D_{xx} of friction, momentum and position diffusion (i.e., position and momentum decoherence), respectively,

$$\begin{aligned} \frac{d\hat{\rho}}{dt} = & -i[\hat{H}, \hat{\rho}] - D_{pp}[\hat{\mathbf{X}}, [\hat{\mathbf{X}}, \hat{\rho}]] - D_{xx}[\hat{\mathbf{P}}, [\hat{\mathbf{P}}, \hat{\rho}]] \\ & - i\frac{\eta}{2M}[\hat{\mathbf{X}}, \{\hat{\mathbf{P}}, \hat{\rho}\}]. \end{aligned} \quad (17)$$

For a heavy Brownian particle ($M \gg m$), in the diffusion limit (cf., e.g., [9,15]), this equation is recovered by all QLBEs. They yield the same (classical) momentum diffusion constant D_{pp} and friction constant η

$$\begin{aligned} \beta\eta = D_{pp} = & \frac{1}{6} \frac{n_g}{m} \int \int d\mathbf{Q} d\mathbf{k}_\perp \rho_g\left(\mathbf{k}_\perp + \frac{1}{2}\mathbf{Q}\right) \\ & \times \left| f\left(\mathbf{k}_\perp - \frac{1}{2}\mathbf{Q}, \mathbf{k}_\perp + \frac{1}{2}\mathbf{Q}\right) \right|^2. \end{aligned} \quad (18)$$

Different QLBEs do not agree upon the value of the position diffusion (momentum decoherence) constant D_{xx} . Our QLBE (15) provides a τ -dependent D_{xx} . We are going to derive it.

Since the Dekker equation (17) corresponds to the second order Taylor expansion of the QLBE (15) in the operators $\hat{\mathbf{X}}, \hat{\mathbf{P}}$, we shall compare the respective coefficients of the term $\hat{\mathbf{P}}\hat{\rho}\hat{\mathbf{P}}$ in the Dekker equation and in the QLBE. In the latter, the leading term comes from the expansion of the $\delta_\tau(\hat{E}_{fi}^*)$ factors around $\hat{\mathbf{P}}=0$. To calculate their argument for $m/M \rightarrow \infty$, recall Eq. (10)

$$\hat{E}_{fi}^* = \frac{\mathbf{Q}}{m} \left(\mathbf{k} - \frac{\mathbf{Q}}{2} \right) - \frac{1}{M} \mathbf{Q} \hat{\mathbf{P}} \equiv E_{fi} - \frac{1}{M} \mathbf{Q} \hat{\mathbf{P}}. \quad (19)$$

Therefore, we get

$$\begin{aligned} 2D_{xx} \hat{\mathbf{P}} \hat{\rho} \hat{\mathbf{P}} = & \frac{1}{M^2} \frac{2\pi n_g}{\tau m^2} \int \int d\mathbf{Q} d\mathbf{k} \rho_g(\mathbf{k}) \\ & \times |f(\mathbf{k} - \mathbf{Q}, \mathbf{k})|^2 [\delta'_\tau(E_{fi})]^2 \mathbf{Q} \hat{\mathbf{P}} \hat{\rho} \hat{\mathbf{P}} \mathbf{Q}, \end{aligned} \quad (20)$$

where we set $\hat{\mathbf{P}}=0$ in the arguments of f , assuming a slowly varying f . Now, we change the integration variable according to $dk_\parallel = (m/Q) dE_{fi}$ and perform the integral over E_{fi} . We can ignore the variation of $\rho(\mathbf{k})$ and $f(\mathbf{k} - \mathbf{Q}, \mathbf{k})$ with the variation of k_\parallel , which can only take values from a narrow vicinity of $Q/2$ because of the narrow support of the δ'_τ . Hence, we substitute $\mathbf{k}_\parallel \approx \mathbf{Q}/2$ in ρ_g and f , then we perform the integral

$$\int [\delta'_\tau(E_{fi})]^2 dk_\parallel = \frac{m}{Q} \int [\delta'_\tau(E_{fi})]^2 dE_{fi} = \frac{m}{Q} \frac{\tau^3}{24\pi}. \quad (21)$$

The resulting equation reads

$$\begin{aligned} 2D_{xx} \hat{\mathbf{P}} \hat{\rho} \hat{\mathbf{P}} = & \frac{\tau^3}{24\pi M^2} \frac{1}{\tau m} \int \int d\mathbf{Q} d\mathbf{k}_\perp \rho_g\left(\mathbf{k}_\perp + \frac{1}{2}\mathbf{Q}\right) \\ & \times \left| f\left(\mathbf{k}_\perp - \frac{1}{2}\mathbf{Q}, \mathbf{k}_\perp + \frac{1}{2}\mathbf{Q}\right) \right|^2 \frac{1}{Q} \mathbf{Q} \hat{\mathbf{P}} \hat{\rho} \hat{\mathbf{P}} \mathbf{Q}, \end{aligned} \quad (22)$$

where rotational symmetry allows for substituting $\mathbf{Q} \hat{\mathbf{P}} \hat{\rho} \hat{\mathbf{P}} \mathbf{Q}$ by $(Q^2/3) \hat{\mathbf{P}} \hat{\rho} \hat{\mathbf{P}}$. We get finally that the quantum position diffusion (momentum decoherence) is proportional to the classical momentum diffusion (18)

$$D_{xx} = \frac{1}{12} \left(\frac{\tau}{M} \right)^2 D_{pp}. \quad (23)$$

This result is in agreement with the conclusion of our Gedanken experiment: the longer the intercollision time, the stronger the momentum decoherence is. According to a universal constraint [3,17] of complete positivity of the Dekker equation (17), the inequality $D_{xx} \geq (\beta/4M)^2 D_{pp}$ must be satisfied, which puts a lower bound on position diffusion at fixed friction η . The QLBE introduced in Ref. [11] predicts this minimum position diffusion. All other previous QLBEs [9–16] predict similar position diffusion suggesting that it is irrelevant at high temperatures. Our result is different: position diffusion (momentum decoherence) goes with τ^2 , not with β^2 . The above complete positivity condition gives the constraint $\tau k_B T \geq \sqrt{3} \hbar/2$ for τ , where T is the temperature; we restored the standard physical dimensions. Substituting Eq. (12) and introducing the thermal de Broglie wavelength λ_T of the molecules, we get

$$\lambda_T \sigma n_g \leq \sqrt{2\pi/3}, \quad (24)$$

suggesting that our QLBE works until a volume $\lambda_T \sigma$ will contain more than cca. 1 molecule on average. This may happen when the density n_g is too high, or the temperature is too low, or the cross section (e.g., the size of the particle) is too big. But otherwise, there is an ample regime for our

particle to develop a localized wave function subject to our QLBE.

II. SUMMARY AND OUTLOOK

Quantum dynamics of a particle in interaction with a dilute gas is definitely tractable by the collision model. We pointed out—in agreement with Ref. [16]—that the coherence between different momentum eigenstates is heavily suppressed (cf. related remarks in [18]) and the very persistence of any localized coherent dynamics is only due to the uncompleted quantum collisions, i.e., to the finite intercollision time τ . This brings a dramatic change in our understanding the quantum behavior of the test particle. Previously, momentum decoherence (i.e., position diffusion) was considered a weak quantum effect irrelevant at high temperatures. Now, we must claim that such weak momentum decoherence predicted by the previous QLBEs was an artifact. Momentum decoherence is a dominating effect. In very dilute gas, at

very small scattering rates, the true dynamics is just the classical diffusion (cf. LBE) through the momentum eigenstates. This means, in particular, that a gradual momentum decoherence under controlled conditions may become testable, e.g., in fullerene [8] or in nano-object interference experiments [19].

There is no guarantee that momentum decoherence has a universal Markovian description. The proposed QLBE captures the influence of finite intercollision time τ within the concept of independent collisions while the mechanism is beyond it: the second collision literally interferes with the first one. Might then we not think of a Lindblad structure \mathcal{L} that is linear functional of $\rho(\mathbf{k})\rho(\mathbf{k}')$? In any case, our QLBE with the finite collision time represents a progressive phenomenology compared to the earlier QLBEs.

ACKNOWLEDGMENTS

This work was supported by the Hungarian OTKA under Grants No. 49384 and No. 75129.

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