Operator Ordering in Quantum Radiative Processes

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Abstract

In this work we reexamine quantum electrodynamics of atomic electrons in the Coulomb gauge in the dipole approximation and calculate the shift of atomic energy levels in the context of Dalibard, Dupont-Roc and Cohen-Tannoudji (DDC) formalism by considering the variation rates of physical observable. We then analyze the physical interpretation of the ordering of operators in the dipole approximation interaction Hamiltonian in terms of field fluctuations and self-reaction of atomic electrons, discussing the arbitrariness in the statistical functions in second order bound-state perturbation theory.

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I. INTRODUCTION

In radiative processes, the ordering problem of atomic and field operators in the interaction Hamiltonian of bound state QED has been raised since the works by Senitzki [1], Ackerhalt et al [2] and others [3]. Behind this discussion is the physical interpretation of atomic radiative effects such as the radiative line shifts in spontaneous emission. Alternative approaches were proposed in order to elucidate important issues concerning such problem. Among them are those based on the complementarity between radiation reaction and vacuum fluctuation effects, which provide a conceptual basis for the physical interpretation of different radiative processes.

In the Dalibard, Dupont-Roc and Cohen-Tannoudji (DDC) formulation, the ordering between the operators of the electromagnetic field, considered as a reservoir ($\mathcal{R}$), and a microscopic atomic system ($\mathcal{S}$) play a fundamental role in the identification of the respective contributions due to the reservoir fluctuation (fr) and the self-reaction (sr) [4] - [6]. They showed that the symmetric ordering gives a true physical meaning to the (fr) and (sr) rates.

In this article we study, in the context of the DDC construct, a more general operator ordering and its physical significance to a given observable variation rate, more specifically, an atomic energy shift.

We use this analysis to establish a formal connection between DDC approach and a closely related treatment, proposed by [6], which is based on the master equation formulation, where the physical motivation relies on the classical theory of damping harmonic oscillator [7]. Finally, we discuss the irrelevance of the ordering to an especific interaction Hamiltonian.

II. THE EFFECTIVE HAMILTONIAN FORMULATION

In the DDC construct the global Hamiltonian for a coupled system $\mathcal{S} + \mathcal{R}$ is, in the dipole approximation, given by

\[ H = H_S + H_R + V, \]
where $H_S$ is the Hamiltonian of the microscopic system $S$, $H_R$ the Hamiltonian of the reservoir $R$ and $V$ the interaction between $S$ and $R$, which we assume to be of the form $V = -gRS$ ($g$ is the coupling constant and $R$ and $S$ are, respectively, Hermitian observable of $R$ and $S$) [4] - [5].

Following [5] we set that the rate of variation for an arbitrary Hermitian observable $G$ of $S$ is given by the Heisenberg equation of motion, and the contribution of the coupling $V$ to this rate can be written as

$$
\left( \frac{dG}{dt} \right)_{\text{coupling}} = -\frac{ig}{\hbar} [R(t)S(t), G(t)] = g\lambda N(t)R(t) + g(1 - \lambda)R(t)N(t),
$$

(2)

where $N(t) = -(i/\hbar)[S(t), G(t)]$ is an Hermitian observable of the microscopic system and $\lambda$ an arbitrary real number. In the above equation we have used the freedom in the ordering of $R(t)$ and $N(t)$, since they commute.

In order to obtain the contributions of reservoir fluctuation (rf) and self-reaction (sr) we perform the following replacement

$$
X(t) = X^f(t) + X^s(t),
$$

(3)

($X = R, S, G$) where $R^f$ (resp. $S^f$ and $G^f$) is the solution, to order 0 in $g$, of the Heisenberg equation of motion for $R$ (resp. $S$ and $G$), corresponding to a free evolution between $t_0$ and $t$, and $R^s(t)$ (resp. $S^s$ and $G^s$) the solution to first order and higher in $g$. Then, substituting (3) in (2) and retaining terms up to second order in $g$, we obtain

$$
\left( \frac{dG}{dt} \right)^{\text{rf}}(t) = -\frac{ig}{\hbar} \{ (1 - \lambda)R^f(t)[S^f(t), G^f(t)] + \lambda[S^f(t), G^f(t)]R^f(t) \} -
\frac{g^2}{\hbar^2} \int_{t_0}^{t} dt'[S^f(t'), [S^f(t), G^f(t)]] \times 
\times ((1 - \lambda)R^f(t')R^f(t) + \lambda R^f(t)R^f(t')),
$$

(4)

$$
\left( \frac{dG}{dt} \right)^{\text{sr}}(t) = -\frac{g^2}{\hbar^2} \int_{t_0}^{t} dt'[R^f(t'), R^f(t)] \times 
\times ((1 - \lambda)S^f(t')[S^f(t), G^f(t)] + \lambda[S^f(t), G^f(t)]S^f(t')).
$$

(5)

Since the rates (4) and (5) contain only free operators, their average value in the reservoir
state \( \sigma_R \) gives\(^1\)

\[
\left\langle \left( \frac{dG}{dt} \right)^{\text{RF}} (t) \right\rangle^{(R)} = -\frac{g'^2}{\hbar^2} \int_{t_0}^{t} dt' C^{(R)}(t, t', \lambda) \left[ S^f(t'), [S^f(t), G^f(t)] \right],
\]

\[
\left\langle \left( \frac{dG}{dt} \right)^{\text{SR}} (t) \right\rangle^{(R)} = -\frac{g'^2}{2\hbar^2} \int_{t_0}^{t} dt' \chi^{(R)}(t, t') \times
\]

\[
\times \{ (1 - \lambda)S^f(t'[S^f(t), G^f(t)] + \lambda[S^f(t), G^f(t)]S^f(t') \},
\]

where we have define \( g' = \sqrt{2}g \) and

\[
C^{(R)}(t, t', \lambda) = \frac{1}{2} \text{Tr}_R[\sigma_R \{ \lambda R^f(t) R^f(t') + (1 - \lambda) R^f(t') R^f(t) \}],
\]

\[
\chi^{(R)}(t, t') = \frac{i}{\hbar} \text{Tr}_R \{ \sigma_R [R^f(t'), R^f(t)] \} \theta(t - t').
\]

The functions \( C^{(R)} \) and \( \chi^{(R)} \) are statistical functions of the reservoir \([8] - [10]\). \( C^{(R)} \) is a kind of correlation function, describing the “dynamics of fluctuations” of \( R \) in the stationary state \( \sigma_R(t_0) \); \( \chi^{(R)} \) is the linear susceptibility of the reservoir, determining the linear response of the averaged observable \( \langle R(t) \rangle \) when the reservoir is acted upon by a perturbation\(^2\).

The above calculation has shown that the freedom in ordering (2) just reflects in the correlation functions given by (8). This result will be explored in section III, where we make a connection with the master equation. In order to get a better understanding of the aforementioned arbitrariness, we will consider the case of an atomic energy shift.

In order to find the energy shifts corresponding to the (rf) and (sr) rates we rewrite (6) and (7) in a convenient form, namely

\[
\left\langle \left( \frac{dG}{dt} \right)^{\text{RF}} (t) \right\rangle^{(R)} = \frac{i}{\hbar} \langle [(H_{\text{eff}}(t))^\text{RF}, G(t)] \rangle_R +
\]

\[
+ \left( -\frac{g'^2}{2\hbar^2} \right) \langle [Y(t, \lambda), [S(t), G(t)]] + [S(t), [Y(t, \lambda), G(t)]] \rangle_R,
\]

\(^1\)Note that the term in the first line of (4) do not contribute to the respective rate since it is linear in the absorption and emission operators of the field.

\(^2\)In (9) \( \theta \) is the Heaviside function, \( \theta(x) = 1 \) if \( x > 0 \), \( \theta(x) = 0 \) if \( x < 0 \).
\[ \left( \frac{dG}{dt} \right)^{\text{sr}}(t) \right)^{(R)} = \frac{i}{\hbar}\langle \{ [H_{\text{eff}}(t)]^{\text{sr}}, G(t) \} \rangle_R + \]
\[ + \left( \frac{-ig'^2}{4\hbar^2} \right) [Z'(t, \lambda)[S(t), G(t)] + [S(t), G(t)]Z''(t, \lambda) - S(t)[Z''(t, \lambda), G(t)] - [Z'(t, \lambda), G(t)]S(t)] \right\rangle_R \]

where
\[ (H_{\text{eff}}(t))^{\text{rf}} = \frac{ig'}{2\hbar}[Y(t, \lambda), S(t)], \]
\[ (H_{\text{eff}}(t))^{\text{sr}} = -\frac{g'}{4}[Z'(t, \lambda)S(t) + S(t)Z''(t, \lambda)] \]

are second order corrections to the Hamiltonian part of \( S \) caused by its interaction with the reservoir and

\[ Y(t, \lambda) = \sum_{ab} q_{ab}(t) \langle a|S|b \rangle \int_0^\infty d\tau C^{(R)}(\tau, \lambda)e^{-i\omega_{ab}\tau}, \]
\[ Z'(t, \lambda) = (1 - \lambda) \sum_{ab} q_{ab}(t) \langle a|S|b \rangle \int_{-\infty}^{\infty} d\tau \chi^{(R)}(\tau)e^{-i\omega_{ab}\tau}, \]
\[ Z''(t, \lambda) = \lambda \sum_{ab} q_{ab}(t) \langle a|S|b \rangle \int_{-\infty}^{\infty} d\tau \chi^{(R)}(\tau)e^{-i\omega_{ab}\tau} \]

with \( q_{ab} \equiv |a\rangle \langle b|, \omega_{ab} = (E_a - E_b)/\hbar \) and \( \tau = t - t' \). Following the same point of view of [5], expression (12) (resp. (13)) describes the part of the evolution due to reservoir fluctuations (resp. due to self-reaction) and which can be described by an effective Hamiltonian. The second line of expression (10) (resp. (11)) describes the non-Hamiltonian part of the evolution of \( G \) caused by the reservoir fluctuation (resp. self-reaction).

**A. The Energy Shifts: Hamiltonian Part**

Corrections (12) and (13) to the Hamiltonian \( H_S \) affect \( S \) through a shifting in its energy eigenstates. Hence, considering a state \( |a\rangle \) (which is an eigenstate of \( H_S \)) we have the following energy shifts

\[ (\delta E_a)^{\text{rf}} = \langle a|(H_{\text{eff}}(t_0))^{\text{rf}}|a\rangle, \]
\[ (\delta E_a)^{\text{sr}} = \langle a|(H_{\text{eff}}(t_0))^{\text{sr}}|a\rangle. \]
Using expression (12), and noting that
\[ Y(t_0) = \int_0^\infty C^{(R)}(\tau, \lambda) S^f(t_0 - \tau) d\tau, \] (19)
expression (17) for \((\delta E_a)^{\text{rf}}\) becomes
\[ (\delta E_a)^{\text{rf}} = -\frac{g^2}{2} \int_{-\infty}^{+\infty} C^{(R)}(\tau, \lambda) \chi^{(S,a)}(\tau) d\tau, \] (20)
where we have introduced a new statistical function, the susceptibility of the system observables
\[ \chi^{(S,a)}(\tau) = i \frac{\hbar}{\lambda} (a[S^f(t_0), S^f(t_0 - \tau)]|a) \theta(\tau). \] (21)

From expression (13) for \((H_{e\text{ff}})^{\text{sr}}\), we can follow the same steps as those from (17) to (20). As a result we obtain
\[ (\delta E_a)^{\text{sr}} = -\frac{g^2}{2} \int_{-\infty}^{+\infty} \chi^{(R)}(\tau) C^{(S,a)}(\tau, \lambda) d\tau, \] (22)
where, again, we have introduced a new statistical function, the “correlation” for the system observable
\[ C^{(S,a)}(\tau, \lambda) = \frac{1}{2} \langle a|\lambda S^f(t_0)S^f(t_0 - \tau) + (1 - \lambda)S^f(t(t_0 - \tau)S^f(t_0)|a \rangle. \] (23)

For future convenience we write (20) and (22) in the frequency space. Using the Parseval’s theorem we have
\[ (\delta E_a)^{\text{rf}} = -\frac{g^2}{2} \int_{-\infty}^{+\infty} d\omega C^{(R)}(\omega, \lambda) \chi^{(S,a)}(\omega), \] (24)
\[ (\delta E_a)^{\text{sr}} = -\frac{g^2}{2} \int_{-\infty}^{+\infty} d\omega \chi^{(R)}(\omega) C^{(S,a)}(\omega, \lambda), \] (25)
where we have used the parity properties of \(C\) and \(\chi\) [5].

Formulas (24) and (25) give us the energy shifts which, \textit{a priori}, depends on \(\lambda\) through the “correlation functions”, expressions (8) and (23). DDC argued that a true physical meaning is obtained by choosing \(\lambda = 1/2\) since then both variation rates, expressions (6)
and (7), become hermitian quantities. As a consequence the energy shifts (24) and (25) will correspond to the (rf) and (sr) effects. It will be shown in section IV that the above assertion becomes meaningless in the case where the interaction Hamiltonian is of the form \( V \sim p \cdot A \).

III. THE MASTER EQUATION APPROACH

In this section, we use the previous results to establish a formal connection with the approach employed in [6], where the physical interpretation of the energy shifts in term (rf) and (sr) are obtained without any reference to operator ordering. In fact, the same energy shifts given by (24) and (25) can also be obtained using a matrix approach based on the evolution equation for the density operator of the global system \( S + R \) in the interaction picture with respect to \( H_S + H_R \). Hence, following [6], the energy shift for a state \( |a\rangle \) of \( S \) caused by its interaction with \( R \) through \( V \) is given by

\[
\Delta a = \frac{1}{\hbar} \mathcal{P} \sum_{\mu,\nu} p_{\mu} \sum_{b} \frac{|\langle \nu, b|V|\mu, a\rangle|^2}{E_{\mu} + E_{a} - E_{\nu} - E_{b}}
\]

(26)

where \( p_{\mu} \) is a distribution of probability corresponding to the reservoir average in the stationary state \( \sigma_R \) and \( |\mu\rangle, |\nu\rangle \) are eigenstates of \( H_R \) with eigenvalue \( E_{\mu}, E_{\nu} \). In (26) \( \mathcal{P} \) denotes the principal value.

It is directly to see that the matrix element \( \langle \mu, a|V|\nu, b\rangle \) in (26) can be factorized in two parts, one relative to \( S \) and another relative to \( R \),

\[
\Delta a = \frac{g^2}{2\hbar^2} \sum_{\mu,\nu} p_{\mu} |\langle \mu|R|\nu\rangle|^2 \left[ \sum_{b} |\langle a|S|b\rangle|^2 \mathcal{P} \frac{1}{\omega_{\mu\nu} + \omega_{ab}} \right].
\]

(27)

In this way, since we know the functional structure of \( C(\omega) \) and \( \chi(\omega) \) [6], namely

\[
C^{(R)}(\omega) = \sum_{\mu,\nu} p_{\mu} \pi |\langle \mu|R|\nu\rangle|^2 \left[ \delta(\omega + \omega_{\mu\nu}) + \delta(\omega - \omega_{\mu\nu}) \right],
\]

(28)

\[
\chi^{(R)}(\omega) = \chi^{i(R)}(\omega) + i\chi^{n(R)}(\omega),
\]

(29)
\[ \chi^{(R)}(\omega) = -\frac{1}{\hbar} \sum_{\mu,\nu} p_{\mu} |\langle \mu | R | \nu \rangle|^2 \left[ \mathcal{P} \frac{1}{\omega_{\mu\nu} + \omega} + \mathcal{P} \frac{1}{\omega_{\mu\nu} - \omega} \right], \] (30)

\[ \chi^{''(R)}(\omega) = \frac{\pi}{\hbar} \sum_{\mu,\nu} p_{\mu} |\langle \mu | R | \nu \rangle|^2 \left[ \delta(\omega_{\mu\nu} + \omega) - \delta(\omega_{\mu\nu} - \omega) \right], \] (31)

and analogous expressions for \( S \) (where only \( p_a = 1 \) is nonzero), we can make a mathematical trick and rewrite the fraction \( 1/(\omega_{\mu\nu} + \omega_{ab}) \) as

\[ \mathcal{P} \frac{1}{\omega_{\mu\nu} + \omega_{ab}} = \frac{1}{2} \int d\omega \times \]
\[ \times \left\{ \left( \mathcal{P} \frac{1}{\omega_{\mu\nu} + \omega} + \mathcal{P} \frac{1}{\omega_{\mu\nu} - \omega} \right) [\lambda \delta(\omega + \omega_{ab}) + (1 - \lambda) \delta(\omega - \omega_{ab})] + \right\} \] (32)

The presence of the \( \lambda \) parameter in the correlation functions of equations (24) and (25) suggests the above construction. This point is crucial because it clarifies the motivation behind the use of (32) in the present context.

Now, substituting (32) into (27) we obtain: \( \Delta_a = \Delta^{rf}_a + \Delta^{sr}_a \), where

\[ \hbar \Delta^{rf}_a = -\frac{g_f^2}{2} \int_{-\infty}^{\infty} d\omega \frac{C^{(R)}(\omega, \lambda)}{2(2\pi)} C^{(S,a)}(\omega), \] (33)

\[ \hbar \Delta^{sr}_a = -\frac{g_f^2}{2} \int_{-\infty}^{\infty} d\omega \frac{C^{(R)}(\omega)}{2(2\pi)} C^{(S,a)}(\omega, \lambda). \] (34)

Chosing \( \lambda = 1/2 \) in the above expression the original formulation given in [6] is recovered and the physical meaning of (27) becomes simple and clear in terms of (rf) and (sr) effects. This are in complete agreement with the results obtained in the last section, expressions (24) and (25). However, in this approach [6] the physical interpretation are borrowed from the classical theory of damping harmonic oscillator, without any reference to the operator ordering in the correlation functions. In fact, for a class of interaction Hamiltonian of the form \( V \sim \mathbf{p} \cdot \mathbf{A} \) the choice of \( \lambda \) in (33) and (34) (or equivalently (24) and (25)) becomes irrelevant as will be shown in the next section.
IV. THE DIPOLE INTERACTION

In order to show the independence of (33) (or (34)) with respect to \( \lambda \) we consider here the same case treated in [6] where the interaction Hamiltonian reduces to the expression

\[
V = - \sum_i \left( \frac{e}{m} p_i \right) A_i(r = 0),
\]

(35)

where \( i = x, y, z \). Comparing the above expression with the definition of \( V \) (\( V = -g \sum_i R_i S_i \)) we see that \( g = 1, R_i = A_i(r = 0) \) (where \( r = 0 \) reflects the long wavelenght approximation we are assuming) and \( S_i = (ep_i/m) \).

In this context, the final expression for the \( x \)-componet of the statistical functions are

\[
\hat{C}_{xx}^R(\omega) = \frac{1}{3\pi \varepsilon_0 c^3} \int_0^{\omega_M} d\omega' \hbar \omega' \left( \langle n(\omega') \rangle + 1/2 \right) \left[ \lambda \delta(\omega' + \omega) + (1 - \lambda) \delta(\omega' - \omega) \right],
\]

(36)

\[
\hat{\chi}_{xx}^R(\omega) = \frac{1}{6\pi^2 \varepsilon_0 c^3} \int_0^{\omega_M} d\omega' \left[ \mathcal{P} \frac{1}{\omega' + \omega} + \mathcal{P} \frac{1}{\omega' - \omega} \right],
\]

(37)

\[
\hat{\chi}_{xx}''^R(\omega) = -\frac{1}{6\pi \varepsilon_0 c^3} \int_0^{\omega_M} d\omega' \omega' \left[ \delta(\omega' + \omega) - \delta(\omega' - \omega) \right],
\]

(38)

and

\[
\hat{C}_{xx}^{\alpha\alpha}(\omega) = \sum_b \frac{e^2}{m^2} |\langle a|p_x|b \rangle|^2 \pi \left[ \lambda \delta(\omega_{ab} + \omega) + (1 - \lambda) \delta(\omega_{ab} - \omega) \right],
\]

(39)

\[
\hat{\chi}_{xx}^{\alpha\alpha}(\omega) = \sum_b \frac{-e^2}{\hbar m^2} |\langle a|p_x|b \rangle|^2 \left[ \mathcal{P} \frac{1}{\omega_{ab} + \omega} + \mathcal{P} \frac{1}{\omega_{ab} - \omega} \right],
\]

(40)

\[
\hat{\chi}_{xx}''^{\alpha\alpha}(\omega) = \sum_b \frac{e^2}{\hbar m^2} |\langle a|p_x|b \rangle|^2 \pi \left[ \delta(\omega_{ab} + \omega) - \delta(\omega_{ab} - \omega) \right],
\]

(41)

where \(|a\rangle\) denotes a given eigenstate of \( H_S \). The above expression are obtained following reference [6] and taking into account identity (32).

In order to obtain the energy shift \( \Delta_{\alpha}^{\text{rf}} \) we substitute (36) and (40) in (33). Remembering that \( g' = \sqrt{2}g = \sqrt{2}, \) we obtain

\[
\hbar \Delta_{\alpha}^{\text{rf}} = - \int_{\infty}^{\infty} \frac{d\omega}{2\pi} \left\{ -\frac{1}{\hbar} \sum_b \frac{e^2}{m^2} |\langle a|p_x|b \rangle|^2 \left[ \mathcal{P} \frac{1}{\omega_{ab} + \omega} + \mathcal{P} \frac{1}{\omega_{ab} - \omega} \right] \right\} \times \left\{ \frac{1}{3\pi \varepsilon_0 c^3} \int_0^{\omega_M} d\omega' \hbar \omega' \left( \langle n(\omega') \rangle + 1/2 \right) \left[ \lambda \delta(\omega' + \omega) + (1 - \lambda) \delta(\omega' - \omega) \right] \right\}.
\]

(42)
Performing the integral over $\omega$, it is straightforward to see that the contribution from $\lambda$ and that from $-\lambda$ will cancel each other remaining just the contribution from the independent term, namely,

$$\hbar \Delta_{\text{rf}}^{a} = \frac{e^2}{6\pi\varepsilon_0 m^2 c^3} \sum_{b} |\langle a | p | b \rangle|^2 \int_{0}^{\omega_{M}} d\omega' \omega' \left( \langle n(\omega') \rangle + 1/2 \right) \left[ P_{\omega_{ab} + \omega'} + P_{\omega_{ab} - \omega'} \right].$$

(43)

where the analogous contribution coming from the $y$ and $z$ components have already been added. As we have seen, the above construct clearly shows the independence of (33) with respect to $\lambda$. In the original formulation given in [6] the above expression may indeed be divided in two parts: one proportional to the factor 1/2 and associated to the Lamb-Retherford energy shift and another proportional to photon number and associated to the AC Stark effect.

It must be also noted that the choice $\lambda = 1/2$ made in the operatorial approach may be seen as a fixing parameter which ultimately gives an hermitian character to the contributions (6) and (7) allowing a fictitious physical meaning for (24) and (25) in terms of (rf) and (sr) effects. However, as remarked by Milonni [11], a true physical meaning is completely arbitrary since the observable variation rates are, in fact, unaccessible for the experiment.

V. CONCLUDING REMARKS

In this work we have applied to the original formulation of DDC construct a more general ordering between the atomic and electromagnetic field (reservoir) operators and calculated the energy shift due to the effective Hamiltonian part. The result showed that the freedom in ordering expression (2) reflects in the energy shifts (24) and (25) through the $\lambda$’s appearance in the correlation functions.

We have also established a formal connection between the Effective Hamiltonian approach and that based on the master equation theory. This connection was guided by the results concerning the general ordering (2) and consequently made explicit the same kind of arbitrariness in the physical interpretation of the related variation rates (i.e., the en-
ergy shifts (33) and (34)) of section III. In the sequence, it was also discussed that, for an interaction Hamiltonian of the form

\[ V = -\sum_i \left( \frac{e}{m} p_i \right) A_i(\mathbf{r} = \mathbf{0}), \]  

(44)

the ordering (2) (or (4) and (5)) becomes meaningless and its role on the final result (expression (43)) turn out to be irrelevant. As a consequence the Lamb-Retherford shift and the AC Stark effect may be obtained without any particular ordering [12].

Another interesting result relies on the fact that our procedure still permit us to fix \textit{a posteriori} a suitable ordering which keeps its (rf) and (sr) interpretation, as can be seen by looking directly to expressions (33) and (34). However, since the Hermicity of (33) and (34) holds only for \( \lambda = 1/2 \), such interpretation becomes artificial.

Once we get a better understanding on the arbitrariness in the operator ordering in DDC construct, we expect to find a direct connection with the works by Senitzki, Ackerhalt and others. The main idea is to construct a similar structure in Fock space and analyze its connection with all possible physical interpretations.

Another interesting application of the present formalism is a possible generalization of the operator ordering in the spirit of q-deformed operator algebras [13], subject of a forthcoming work.

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