Retaining space and time coherence in radiative transfer models

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(Received 18 November 2014; published 15 May 2015)

A recent model for radiative transfer that accounts for spatial coherence is extended in such a way as to retain temporal coherence. The method employs Bogoliubov-Born-Green-Kirkwood-Yvon hierarchy techniques. Both spatial and temporal coherence are shown to affect the formation of atomic line spectra. Calculations of Lyman α radiation transport in optically thick divertor plasma conditions are reported as an illustration of the model. A possible extension of the formalism to dense media involving correlations between atoms is discussed in an appendix. A link to partial frequency redistribution modeling is also discussed.

DOI: 10.1103/PhysRevE.91.053103 PACS number(s): 52.25.Os, 05.60.Gg, 42.25.Kb, 32.70.Jz

I. INTRODUCTION

Radiation transport models are involved in optically thick gases and plasmas: applications include astrophysics [1], laboratory experiments (e.g., inertial and magnetic fusion facilities [2,3]), gas discharge lamps [4], lasers [5], and biomedical physics [6,7]. As a rule, the radiation field is viewed as a set of particles (photons) evolving along straight lines and interacting locally with massive particles (e.g., atoms) through emission, absorption, and scattering processes. This particle picture is suitable for an interpretation in terms of classical physics (through Newtonian mechanics and geometrical optics), but it may be inaccurate in regimes such that the radiation coherence length $\lambda_c \sim c/\Delta\omega \equiv \hbar/\Delta p$ (with $\Delta\omega = c\Delta p/\hbar$ being the radiation's characteristic spectral band [8]) is comparable to the characteristic gradient length scales.

A generalization of the radiative transfer equation that accounts for spatial coherence has been derived recently, using a master equation for the quantized electromagnetic field and adapting it to the one-photon Wigner function [9]. The resulting transport equation has nonlocal source and loss terms involving a phase-space volume $(\Delta x\Delta p)^3$ of the order of $\hbar^3$. For radiation with a small spectral band as in atomic lines, the delocalization in space can be important and it would be observable, in principle, according to calculations that have been performed in [10]. An experimental test of the theory using a laser source has also been suggested in [10]. Because a laser light has a narrow spectrum, the coherence length can be very large (up to several tens of centimeters [5]) and comparable to the cavity size, so that a measurement of the output power could serve as a benchmark.

An issue that remains to be clarified concerns the role of temporal coherence. The coherence time $1/\Delta\omega$ implies a "coarse-graining" scale under which physical processes are ambiguous, in the same fashion as does the coherence length. This ambiguity is associated with the Fourier time-frequency (or Heisenberg time-energy) uncertainty relation, and it must be retained consistently with the spatial delocalization in a realistic calculation.

The purpose of this article is to provide a method to account for both spatial and temporal coherence in radiation transport models and to investigate the effects of temporal coherence on the formation of spectral lines. The method is inspired from plasma kinetic theory and employs an adaptation of the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy to photons and atoms. The article is organized as follows: Sec. II gives a summary of the radiation transport model accounting for spatial coherence and previously developed in [10], the BBGKY hierarchy technique is presented and discussed in Sec. III, and a closure model for coherent radiation is presented in Sec. IV. Calculations of the transport of hydrogen Lyman photons in optically thick divertor plasma conditions are reported as an illustration of the model in Sec. V. Two appendixes are devoted to further extensions of the formalism and application to partial frequency redistribution modeling.

II. RADIATIVE TRANSFER EQUATION WITH SPATIAL COHERENCE

The derivation of a photon transport equation retaining radiation coherence requires a careful consideration of the coupled photon-atom dynamics. In [10], a first-principles approach based on QED master equations has been used and adapted to the quantum phase-space formalism, in such a way as to capture the particle (photon) picture involved in radiative transfer textbooks. We give hereafter a summary of the theoretical background.

The system of interest is a gas of $\mathcal{N}$ atoms or multicharged ions with discrete energy levels, immersed in a plasma and emitting and absorbing radiation in spectral lines. It is assumed that the radiation has a narrow band in the sense that spectral lines are well resolved; a criterion for that is provided by the condition $\Delta\omega/\omega_0 \ll 1$, where ω_0 is the position in frequency of a spectral line and $\Delta\omega$ is the characteristic width. The radiation is described within the second quantization, using the standard discretization procedure for modes [11]. The density operator ρ of the total (atoms + plasma + radiation) system obeys the Liouville-von Neumann equation

$$\frac{d\rho}{dt} + \frac{i}{\hbar}[H,\rho] = 0, \tag{1}$$

and the Hamiltonian reads

$$H = H_R + H_A + V \equiv H_R + \sum_{a=1}^{\mathcal{N}}[H_A(a) + V(a)]. \tag{2}$$

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H_R refers to the evolution of the free radiation field, $H_A(a)$ denotes the evolution of the atom a under the influence of the plasma but assuming no radiation is present, and $V(a)$ is the dipole interaction term (long-wavelength approximation). Explicitly, this term reads

$$V = \sum_a -\mathbf{d}_a \cdot \mathbf{E}(\mathbf{r}_a), \tag{3}$$

where $\mathbf{d}_a$, $\mathbf{r}_a$ are the dipole moment and the position operator of the center of mass of the atom a, and $\mathbf{E}$ is the quantized transverse electric field:

$$\mathbf{E}(\mathbf{r}) = \sum_j i\sqrt{\frac{\hbar\omega_j}{2\varepsilon_0 L^3}}(a_j e^{i\mathbf{k}_j\cdot\mathbf{r}} - a_j^\dagger e^{-i\mathbf{k}_j\cdot\mathbf{r}})\boldsymbol{\varepsilon}_j. \tag{4}$$

Here, the sum is done over the discretized radiation modes $(\mathbf{k}_j,\boldsymbol{\varepsilon}_j) \equiv j$ ($\mathbf{k}_j$ is the wave vector and $\boldsymbol{\varepsilon}_j$ is the polarization vector), a_j and $a_j^\dagger$ are the annihilation and creation operators, L^3 is the quantization volume, and $\omega_j = |\mathbf{k}_j|c$. In Eq. (2), it is implied that the free charged particles (the "plasma") do not interact with the radiation, i.e., we omit the bremsstrahlung and its inverse in our treatment. It is also implied that the atoms do not collide between each other.

The Liouville-von Neumann equation (1) is general but not practical for calculations since it involves a huge Hilbert space. A common approach consists in writing down master equations for reduced density operators obtained from partial trace; examples of such operators include the radiation field density operator $\rho_R = \mathrm{Tr}_A(\rho)$ and the atomic density operator $\rho_A = \mathrm{Tr}_R(\rho)$ where $\mathrm{Tr}_A(\dots)$ and $\mathrm{Tr}_R(\dots)$ refer to trace with respect to the atoms' and the radiation's Hilbert space, respectively. The master equation used in [10] for the radiation field results from a series of algebraic operations done on the Liouville-von Neumann equation within the weak-coupling approximation, following previous works [12,13]. It reads

$$\frac{d\rho_R}{dt}(t) + \frac{i}{\hbar}[H_R,\rho_R(t)] = -\frac{1}{\hbar^2}\int_0^\infty d\tau \mathrm{Tr}_A[V,[e^{-iH_0\tau/\hbar}Ve^{iH_0\tau/\hbar},\rho_A(t)\rho_R(t)]], \tag{5}$$

where $H_0 = H_A + H_R$. This equation can be interpreted as a balance relation assuming the photon-atom interaction processes are short-time events well separated in time. These constraints ensure that the integral's upper bound can be set to infinity, while the density operator can be factorized and evaluated at time t (Markov approximation, e.g., [14,15]). In the right-hand side, it is sufficient to keep only terms bilinear in a_j, $a_j^\dagger$ because they are resonant (rotating-wave approximation).

A phase-space adaptation of Eq. (5) that makes a link to the radiative transfer equation is obtained by forming an evolution equation for the coherence function $N_{jj'}(t) = \mathrm{Tr}_R[a_j^\dagger a_{j'}\rho_R(t)]$ and "Wigner" transforming it, using the relation

$$W(\mathbf{r},\mathbf{p},t) = \left(\frac{2}{\hbar L}\right)^3 \sum_{jj'} \delta_{\boldsymbol{\varepsilon}_j\boldsymbol{\varepsilon}_{j'}}\delta\left(\mathbf{k}_j + \mathbf{k}'_j - \frac{2\mathbf{p}}{\hbar}\right) \times N_{jj'}(t)e^{-i\mathbf{k}_{jj'}\cdot\mathbf{r}}, \tag{6}$$

and its reciprocal

$$N_{jj'}(t) = \left(\frac{2\pi\hbar}{L}\right)^3 \frac{\delta_{\boldsymbol{\varepsilon}_j\boldsymbol{\varepsilon}_{j'}}}{2}\int d^3r e^{i\mathbf{k}_{jj'}\cdot\mathbf{r}} W\left(\mathbf{r},\frac{\hbar\mathbf{k}_j}{2} + \frac{\hbar\mathbf{k}_{j'}}{2},t\right), \tag{7}$$

with $W(\mathbf{r},\mathbf{p},t)$ being the one-photon Wigner function. Here by definition $\mathbf{k}_{jj'} = \mathbf{k}_j - \mathbf{k}_{j'}$. The evolution equation for $N_{jj'}$ has the structure of a first-order matrix differential equation

$$\frac{dN_{jj'}}{dt} - i(\omega_j - \omega_{j'})N_{jj'} = \gamma_{2jj'} - \sum_{j''}(\Gamma_{jj''}N_{j''j'} + \Gamma^*_{j'j''}N_{jj''}). \tag{8}$$

The first term of the right-hand side is a source corresponding to spontaneous emission and the second term accounts for absorption and stimulated emission and can be interpreted as a loss if the medium is not amplifying. The rates can be written in terms of two half-Fourier transforms of correlation functions denoted $\Gamma_{1jj'}$ and $\Gamma_{2jj'}$; explicitly

$$\gamma_{2jj'} = \Gamma_{2jj'} + \Gamma^*_{2j'j}, \tag{9}$$

$$\Gamma_{jj'} = \Gamma^*_{1jj'} - \Gamma_{2jj'}, \tag{10}$$

with

$$\Gamma_{1jj'} = \frac{\sqrt{\omega_j\omega_{j'}}}{2\varepsilon_0\hbar L^3}\mathrm{Tr}_A\sum_{a,a'=1}^{\mathcal{N}}\int_0^\infty d\tau D_{aj}e^{-iH_A\tau/\hbar} \times D^\dagger_{a'j'}e^{iH_A\tau/\hbar}e^{i\omega_{j'}\tau}\rho_A(t), \tag{11}$$

$$\Gamma_{2jj'} = \frac{\sqrt{\omega_j\omega_{j'}}}{2\varepsilon_0\hbar L^3}\mathrm{Tr}_A\sum_{a,a'=1}^{\mathcal{N}}\int_0^\infty d\tau D^\dagger_{aj}e^{-iH_A\tau/\hbar} \times D_{a'j'}e^{iH_A\tau/\hbar}e^{-i\omega_{j'}\tau}\rho_A(t), \tag{12}$$

and where the notation $D_{aj} = \mathbf{d}_a\cdot\boldsymbol{\varepsilon}_j\exp(-i\mathbf{k}_j\cdot\mathbf{r}_a)$ has been used. A practical simplification consists in assuming the atoms independent of each other and yields the formal substitution $\sum_{a,a'=1}^{\mathcal{N}} \to \mathcal{N}\sum_{a,a'=1}^{\mathcal{N}}\delta_{aa_0}\delta_{a'a_0}$ with a_0 referring to any atom. A summation of Eq. (8) over the radiation modes following Eq. (6), together with the reciprocal relation (7), yields the following closed transport equation for the Wigner function:

$$\frac{\partial W}{\partial t} + c\{W,|\mathbf{p}|\}_M = S - \mathcal{L}[W], \tag{13}$$

where $\{,\}_M$ denotes the Moyal bracket

$$\{W,|\mathbf{p}|\}_M = \frac{1}{i\hbar}(W\star|\mathbf{p}| - |\mathbf{p}|\star W), \tag{14}$$

and where the source and loss terms are given by

$$S(\mathbf{r},\mathbf{p},t) = \frac{1}{\hbar|\mathbf{p}|^3}\lim_{\substack{\mathbf{r}'\to\mathbf{r}\\ \mathbf{p}'\to\mathbf{p}}}\mathrm{Re}(\eta_c\star\Delta^*_{\mathbf{r}',\mathbf{p}'})(\mathbf{r},\mathbf{p},t), \tag{15}$$

$$\mathcal{L}[W](\mathbf{r},\mathbf{p},t) = c\lim_{\substack{\mathbf{r}'\to\mathbf{r}\\ \mathbf{p}'\to\mathbf{p}}}\mathrm{Re}[(W\Delta^*_{\mathbf{r}',\mathbf{p}'})\star\chi_c](\mathbf{r},\mathbf{p},t), \tag{16}$$

with $\Delta_{\mathbf{r}',\mathbf{p}'} = (\pi\hbar)^3(\delta_{\mathbf{r}'}\star\delta_{\mathbf{p}'})$. The Moyal star product $\star$ implies nonlocal terms owing to radiation coherence. Equation (13)

reduces to the common radiative transfer equation given in textbooks [1,5,6,16–18] at the large spectral band limit (incoherent radiation). The quantities η_c and χ_c in Eqs. (15) and (16) are complex generalizations of the emission and absorption coefficients used in the standard radiative transfer equation, and they are related to the complex refractive index used in classical electromagnetism [19]. Scattering is implied and described within the complete redistribution approximation.

The transport equation (13) presents a challenging computational issue due to the six-dimensional integrals implied in the Moyal products. It has been solved in specific cases in [9,10,20], using simplifications (e.g., slab geometry with localized boundary conditions) to illustrate the possible inaccuracy of the standard radiative transfer theory at regimes with significant coherence length. It should be emphasized that such simplifications are very specific to the problem under consideration and require a systematic analysis. The integrals in Eqs. (15) and (16) involve strongly oscillating kernels, with no straightforward discretization scheme, and the Wigner function may exhibit significant "wave" features, sufficiently so that the use of localized boundary conditions can be questionable, e.g., [21]. An adaptation of Boltzmann kinetic Monte Carlo solvers is also not straightforward, because neither the kernels nor the Wigner function are true probability density functions. The Monte Carlo method is suitable for the weakly coherent radiation limit, where only terms of the first order in $\hbar$ of the Moyal series are required (such a procedure has already been used, e.g., in [22]). The main advantage of the quantum phase-space approach is to provide a transparent link between the particle picture utilized in radiative transfer textbooks and the first-principles QED framework. It should be noted that the transport equation (13) contains the same amount of information as Eq. (8), while being more complicated. It would be relevant to consider Eq. (8) for numerical calculations. In the stationary regime ($d/dt \equiv 0$), this equation is of the Lyapunov-type and can be solved using algorithms already available in the literature [23,24].

III. RETAINING TEMPORAL COHERENCE

The transport equation presented in the previous section, either in the form (8) or (13), is a relation local in time, implying the photon-atom interaction processes are short-time events whose duration is negligible with respect to a relevant relaxation time scale T_R. This assumption is questionable if the radiation is coherent. The temporal coherence involves a time scale ($\Delta\omega^{-1}$) under which the physical processes are ambiguous, in the same fashion as does the spatial coherence. A critical assumption made in the derivation of Eq. (8) [or (13)] concerns the weak-coupling approximation implied in the master equation (5). This approximation rests on a separation in time of the photon-atom interaction processes and utilizes the ordering $\Delta\omega T_R \gg 1$ (e.g., [14]). If this ordering is not satisfied, the distinction between two consecutive processes, say, a photon absorption at time t_1 and a subsequent emission at time t_2, becomes ambiguous. A generalization of the master equation (5) accounting for temporal coherence should involve the following, more general, right-hand side

$$\text{r.h.s.}' = -\frac{1}{\hbar^2}\int_0^t d\tau \,\mathrm{Tr}_A[V,[e^{-iH_0\tau/\hbar}Ve^{iH_0\tau/\hbar},e^{-iH_0\tau/\hbar} \times \rho(t-\tau)e^{iH_0\tau/\hbar}]], \tag{17}$$

together with an appropriate closure relation, an alternative to the relation $e^{-iH_0\tau/\hbar}\rho(t-\tau)e^{iH_0\tau/\hbar} \simeq \rho_A(t)\rho_R(t)$ implied in Eq. (5).

Theoretical works have already been carried out in such a way as to get such a relation for the description of photon scattering processes, in the framework of redistribution function modeling [13]. We develop here a method based on the BBGKY hierarchy and inspired from quantum kinetic theory (e.g., [25–29]). The quantities of interest are the reduced p-atom density operators defined as $F_p(1\ldots p) = \mathrm{Tr}_{p+1\ldots\mathcal{N}}(\rho)$. They obey the following hierarchy of equations (t is not written explicitly)

$$\left\{\frac{d}{dt} + i\left[\hat{L}_R + \sum_{a=1}^{p}[\hat{L}_A(a) + \hat{V}(a)]\right]\right\} F_p(1\ldots p) = -i(\mathcal{N}-p)\mathrm{Tr}_{p+1}[\hat{V}(p+1)F_{p+1}(1\ldots p+1)]. \tag{18}$$

Here, the hat $\hat{}$ denotes Liouville superoperators defined in terms of commutators: $\hat{L}_R X \equiv [H_R, X]/\hbar$, $\hat{L}_A X \equiv [H_A, X]/\hbar$, and $\hat{V}X \equiv [V,X]/\hbar$ for any operator X. The hierarchy equation (18) with $0 \leqslant p \leqslant \mathcal{N}$ is equivalent to the Liouville-von Neumann equation (1). The radiation density operator ρ_R is identical to F_0. As in classical kinetic theory, it is practical to introduce a cluster expansion involving the correlations

$$\begin{aligned}
F_1(1) &= \bar{F}_1(1)F_0 + G_1(1),\\
F_2(1,2) &= \bar{F}_2(1,2)F_0 + \bar{F}_1(1)G_1(2) + \bar{F}_1(2)G_1(1) + G_2(1,2),\\
F_3(1,2,3) &= \bar{F}_3(1,2,3)F_0 + \bar{F}_2(1,2)G_1(3) + \bar{F}_2(1,3)G_1(2)\\
&\quad + \bar{F}_2(2,3)G_1(1), + \bar{F}_1(1)G_2(2,3) + \bar{F}_1(2)G_2(1,3)\\
&\quad + \bar{F}_1(3)G_2(1,2) + G_3(1,2,3), \ldots
\end{aligned} \tag{19}$$

Here, $\bar{F}_1(1)$, $\bar{F}_2(a,b)$, etc. are joint atomic distribution functions (operators) traced over the radiation field, i.e., $\bar{F}_p(1\ldots p) = \mathrm{Tr}_R F_p(1\ldots p)$, and the G's denote correlations between the atoms and the radiation field. The reduced density operator ρ_A defined in the previous section is identical to $\bar{F}_{\mathcal{N}}$.

Equation (18) with $0 \leqslant p \leqslant \mathcal{N}$ is equivalent to a hierarchy of equations for the correlations, which is suitable for the elaboration of a closure relation. It is instructive to examine the first equations of this hierarchy:

$$\left\{\frac{d}{dt} + i[\hat{L}_A(1) + \hat{\bar{V}}(1)]\right\}\bar{F}_1(1) = -i\mathrm{Tr}_R[\hat{V}(1)G_1(1)], \tag{20}$$

$$\left[\frac{d}{dt} + i(\hat{L}_R + \hat{\bar{V}}_R)\right]F_0 = -i\mathcal{N}\mathrm{Tr}_1[\hat{V}(1)G_1(1)], \tag{21}$$

$$\begin{aligned}
&\left\{\frac{d}{dt} + i[\hat{L}_R + \hat{L}_A(1) + \hat{V}(1) + \hat{\bar{V}}_R]\right\}G_1(1)\\
&\quad -iF_0\mathrm{Tr}_R[\hat{V}(1)G_1(1)]\\
&= -i\delta\hat{V}(1)[\bar{F}_1(1)F_0] - i\mathcal{N}\mathrm{Tr}_2\{\hat{V}(2)[\bar{G}_2(1,2)\bar{F}_0 + G_2(1,2)]\}.
\end{aligned} \tag{22}$$

The derivation involves tedious algebraic manipulations with traces and commutators and has not been detailed here. The first equation describes the evolution of the one-atom density operator under the influence of interactions with the radiation field and with the plasma (recall that the plasma is also accounted for in $\hat{L}_A$ and in $\bar{F}_1$). The superoperator $\hat{\bar{V}}(1)$ is associated with the interaction Hamiltonian $V(1)$ averaged over the radiation field, $\bar{V}(1) = \mathrm{Tr}_R[V(1)F_0]$ (Hartree-Fock contribution). It involves the mean electric field and can be important in amplifying media, e.g., in a laser cavity. It is identically zero if the radiation field is in a well-defined Fock state (totally incoherent limit). Note the analogy with the mean field involved in the classical Vlasov equation. In the atomic physics framework, it can also be viewed as a "dressing" correction to the energy levels. The right-hand side is a source + loss term induced by radiation-atom correlations and can be assimilated to the usual spontaneous or stimulated emission and absorption processes. Equation (21) describes the evolution of the radiation field and is equivalent to the master equation (5) with the r.h.s. (17). The superoperator $\hat{\bar{V}}_R$ is associated with the operator $\mathcal{N}\mathrm{Tr}_1[V(1)\bar{F}_1(1)] \equiv \bar{V}_R$ and involves the mean dipole resulting from the $\mathcal{N}$ atoms. It can be important if there are atomic coherences (nondiagonal matrix elements of the atomic density operator $\bar{F}_1$). Finally, Eq. (22) describes the evolution of the correlation between the radiation field and atom 1; $\delta V(1) = V(1) - \bar{V}(1)$ [$\delta\hat{V}(1) = \hat{V}(1) - \hat{\bar{V}}(1)$ in the Liouville space] is short-hand notation and denotes the deviation to the mean interaction between the radiation field and atom 1; $\bar{G}_2(1,2) = \bar{F}_2(1,2) - \bar{F}_1(1)\bar{F}_1(2)$ is the correlation between atom 1 and atom 2.

Equations (20)–(22) are coupled to each other and do not form a closed set of equations due to the presence of the correlations G_2 and $\bar{G}_2$. In general, the correlations G_1, G_2, etc., can be viewed as quantities of first order, second order, etc., in δV, which suggests a perturbative scheme for the elaboration of a closure relation. The weak-coupling approximation corresponds to assuming $G_2 \equiv 0$ and $\bar{G}_2 \equiv 0$, together with $\bar{V}_1 \equiv 0$, and neglecting terms of the second order in δV in Eq. (22); the G_1 correlation obtained in this framework reads

$$\begin{aligned} G_1^{(1)}(1,t) &= -i\int_0^t d\tau e^{-i[\hat{L}_R+\hat{L}_A(1)]\tau}\hat{V}(1)[\bar{F}_1(1,t-\tau)F_0(t-\tau)] \\ &\simeq -i\int_0^t d\tau e^{-i[\hat{L}_R+\hat{L}_A(1)]\tau}\hat{V}(1)e^{i[\hat{L}_R+\hat{L}_A(1)]\tau}[\bar{F}_1(1,t)F_0(t)] \\ &\simeq -i\int_0^\infty d\tau e^{-i[\hat{L}_R+\hat{L}_A(1)]\tau}\hat{V}(1)e^{i[\hat{L}_R+\hat{L}_A(1)]\tau}[\bar{F}_1(1,t)F_0(t)], \end{aligned} \tag{23}$$

indicating the factorization $e^{-iH_0\tau/\hbar}\rho(t-\tau)e^{iH_0\tau/\hbar} \simeq \rho_A(t)\rho_R(t)$ implied in Eq. (5) and discussed above. The $t \to \infty$ limit in the integral's upper bound denotes an asymptotic description, as done in the elaboration of classical master equations (e.g., [30]).

IV. CLOSURE MODEL FOR COHERENT RADIATION

We propose hereafter an extension of the weak-coupling approximation designed in such a way as to account for finite time coherence. We still neglect second-order correlations and mean fields but relax the approximation $\bar{F}_1(1,t-\tau)F_0(t-\tau) \simeq \exp\{i[\hat{L}_R + \hat{L}_A(1)]\tau\}[\bar{F}_1(1,t)F_0(t)]$ done in Eq. (23). In this framework, the radiation and one-atom density operators F_0 and $\bar{F}_1(1)$ obey a closed set of equations nonlocal in time

$$\left[\frac{d}{dt} + i\hat{L}_A(1)\right]\bar{F}_1(1,t) = -\mathrm{Tr}_R\int_0^t d\tau\hat{V}(1)e^{-i[\hat{L}_R+\hat{L}_A(1)]\tau}\hat{V}(1)[\bar{F}_1(1,t-\tau)F_0(t-\tau)], \tag{24}$$

$$\left(\frac{d}{dt} + i\hat{L}_R\right)F_0(t) = -\mathcal{N}\mathrm{Tr}_1\int_0^t d\tau\hat{V}(1)e^{-i[\hat{L}_R+\hat{L}_A(1)]\tau}\hat{V}(1)[\bar{F}_1(1,t-\tau)F_0(t-\tau)]. \tag{25}$$

It is possible to write down an evolution equation for the coherence function $N_{jj'}$ introduced in Sec. II, starting from Eq. (25) and using the commutation properties of the creation and annihilation operators. The resulting equation has a structure similar to Eq. (8) but it involves source and loss terms nonlocal in time:

$$\left(\frac{d}{dt} - i\omega_{jj'}\right)N_{jj'}(t) = \int_0^t d\tau\bar{\gamma}_{2jj'}(t,\tau) - \sum_{j''}\int_0^t d\tau[\bar{\Gamma}_{jj''}(t,\tau)e^{i\omega_{j''j'}\tau}N_{j''j'}(t-\tau) + \bar{\Gamma}^*_{j'j''}(t,\tau)e^{i\omega_{jj''}\tau}N_{jj''}(t-\tau)]. \tag{26}$$

Here, the short-hand notation $\omega_j - \omega_{j'} \equiv \omega_{jj'}$ has been introduced and the rates present in the integrals are defined by $\bar{\gamma}_{2jj'} = \bar{\Gamma}_{2jj'} + \bar{\Gamma}^*_{2j'j}$, $\bar{\Gamma}_{jj'} = \bar{\Gamma}^*_{1jj'} - \bar{\Gamma}_{2jj'}$, with

$$\bar{\Gamma}_{1jj'}(t,\tau) = \frac{\mathcal{N}\sqrt{\omega_j\omega_{j'}}}{2\varepsilon_0\hbar L^3}\mathrm{Tr}_1[e^{iH_A(1)\tau/\hbar}D_j e^{-iH_A(1)\tau/\hbar}D^\dagger_{j'}\bar{F}_1(1,t-\tau)]e^{i\omega_{j'}\tau}, \tag{27}$$

$$\bar{\Gamma}_{2jj'}(t,\tau) = \frac{\mathcal{N}\sqrt{\omega_j\omega_{j'}}}{2\varepsilon_0\hbar L^3}\mathrm{Tr}_1[e^{iH_A(1)\tau/\hbar}D^\dagger_j e^{-iH_A(1)\tau/\hbar}D_{j'}\bar{F}_1(1,t-\tau)]e^{-i\omega_{j'}\tau}. \tag{28}$$

The operator $D_j = \mathbf{d}\cdot\boldsymbol{\varepsilon}_j\exp(-i\mathbf{k}_j\cdot\mathbf{r})$ refers to the dipole moment $\mathbf{d}$ and the position $\mathbf{r}$ of atom 1, i.e., $D_j \equiv D_{aj}$ with $a = 1$. The source and loss terms in Eq. (26) are nonlocal in time due to the finite duration of the photon-atom interaction processes. They become local at the limit $\Delta\omega^{-1} \to 0$.

A phase-space picture can be set up using the same procedure as in Sec. II, i.e., introducing the one-photon Wigner function and interpreting the trace in Eqs. (27) and (28) as an average involving the atomic phase-space density. The resulting evolution equation for the photon Wigner function has the same structure as Eq. (13) but involves source and loss terms nonlocal in time:

$$S(\mathbf{r},\mathbf{p},t) = \frac{1}{\pi^3\hbar^4 p^3}\mathrm{Re}\int d^3r'\int d^3p'\int_0^t d\tau\,\bar{\eta}_c(\mathbf{r}',\mathbf{p}',t,\tau)e^{-2i(\mathbf{r}-\mathbf{r}')\cdot(\mathbf{p}-\mathbf{p}')/\hbar}, \tag{29}$$

$$\mathcal{L}[W](\mathbf{r},\mathbf{p},t) = \int d^3r'\int d^3p'\int_0^t d\tau\,\bar{K}(\mathbf{r},\mathbf{p},\mathbf{r}',\mathbf{p}',t,\tau)W(\mathbf{r}',\mathbf{p}',t-\tau), \tag{30}$$

$$\bar{K}(\mathbf{r},\mathbf{p},\mathbf{r}',\mathbf{p}',t,\tau) = \frac{c}{(\pi\hbar)^6}\mathrm{Re}\int d^3r''\int d^3p''\,\bar{\chi}_c(\mathbf{r}'',\mathbf{p}'',t,\tau)e^{2i[(\mathbf{r}-\mathbf{r}')\cdot(\mathbf{p}'-\mathbf{p}'')-(\mathbf{r}-\mathbf{r}'')\cdot(\mathbf{p}-\mathbf{p}')]/\hbar}\times e^{ic\tau(p''-|2\mathbf{p}'-\mathbf{p}''|)/\hbar}. \tag{31}$$

Here, by definition,

$$\bar{\chi}_c = \bar{\chi}_{\mathrm{c,abs}} - \bar{\chi}_{\mathrm{c,em}}, \tag{32}$$

$$\bar{\eta}_c = \frac{A_{eg}}{B_{eg}}\bar{\chi}_{\mathrm{c,em}}, \tag{33}$$

$$\bar{\chi}_{\mathrm{c,abs}}(\mathbf{r},\mathbf{p},t,\tau) = \int d^3v\,\tilde{\chi}_{\mathrm{c,abs}}(\mathbf{r},\mathbf{p},\mathbf{v},t,\tau), \tag{34}$$

$$\bar{\chi}_{\mathrm{c,em}}(\mathbf{r},\mathbf{p},t,\tau) = \int d^3v\,\tilde{\chi}_{\mathrm{c,em}}(\mathbf{r},\mathbf{p},\mathbf{v},t,\tau), \tag{35}$$

$$\tilde{\chi}_{\mathrm{c,abs}}(\mathbf{r},\mathbf{p},\mathbf{v},t,\tau) = f_g(\mathbf{r}-\mathbf{v}\tau,\mathbf{v},t-\tau)\frac{\hbar\omega_0 B_{ge}}{4\pi^2}C(\tau)e^{-i(pc-\mathbf{p}\cdot\mathbf{v})\tau/\hbar}, \tag{36}$$

$$\tilde{\chi}_{\mathrm{c,em}}(\mathbf{r},\mathbf{p},\mathbf{v},t,\tau) = f_e(\mathbf{r}-\mathbf{v}\tau,\mathbf{v},t-\tau)\frac{\hbar\omega_0 B_{eg}}{4\pi^2}C(\tau)e^{-i(pc-\mathbf{p}\cdot\mathbf{v})\tau/\hbar}. \tag{37}$$

The subscripts g, e refer to the lower and upper levels of the transition; A_{eg}, B_{eg}, and B_{ge} are the Einstein coefficients; $C(\tau)$ is the autocorrelation function of the atomic dipole projected onto the polarization plane in reduced units; and f_g and f_e are the phase-space densities of atoms in the lower and upper levels normalized to the total number of atoms [i.e., $\int d^3r d^3v(f_g+f_e) = \mathcal{N}$].

The atoms' phase-space densities are formally defined as $f_{g,e}(\mathbf{r}',\mathbf{v}',t) = \mathcal{N}\mathrm{Tr}_1[P_{g,e}\delta(\mathbf{r}'-\mathbf{r})\delta(\mathbf{v}'-\mathbf{v})F_1(1,t)]$, where P_g and P_e are projectors onto the lower and upper levels, $\mathbf{r}$ and $\mathbf{v}$ are the position and velocity operator of the atom's center of mass, and $\mathbf{r}'$ and $\mathbf{v}'$ stand here for scalar quantities. A Wigner transform can be used instead of the delta functions if the center-of-mass thermal de Broglie length is comparable to another relevant length scale. Applying the trace $\mathcal{N}\mathrm{Tr}_1[P_{g,e}\delta(\mathbf{r}'-\mathbf{r})\delta(\mathbf{v}'-\mathbf{v})\dots]$ to Eq. (24) yields a set of evolution equations for f_g and f_e

$$\frac{\partial f_{g,e}}{\partial t}(\mathbf{r},\mathbf{v},t) + \mathbf{v}\cdot\frac{\partial f_{g,e}}{\partial \mathbf{r}}(\mathbf{r},\mathbf{v},t) = \pm A_{eg}f_e(\mathbf{r},\mathbf{v},t) \mp \frac{c}{(\pi\hbar)^3}\mathrm{Re}\int d^3r'\int d^3p'\int d^3p''\int_0^t d\tau\,\tilde{\chi}_c(\mathbf{r},\mathbf{p}'',\mathbf{v},t,\tau)$$
$$\times e^{2i[(\mathbf{r}-\mathbf{r}')\cdot(\mathbf{p}'-\mathbf{p}'')]/\hbar}\,e^{ic\tau(p''-|2\mathbf{p}'-\mathbf{p}''|)/\hbar}\,W(\mathbf{r}',\mathbf{p}',t-\tau). \tag{38}$$

The upper (lower) sign refers to the evolution of f_g (f_e). The plus or minus sign indicates that the total number of atoms $\mathcal{N}$ is conserved. Equation (38) holds in the limiting case where interactions with the plasma (collisions) are negligible. A generalization that accounts for collisional excitation and deexcitation can be set up through additional source and loss terms involving rates; ionization and recombination processes can be retained in the same way. It is worth noting that the density of atoms in the ground level $N_g = \int d^3v f_g$ obeys the same evolution equation as the photon density $N = \int d^3p W$ if collisions and mean (fluid) velocity are neglected.

V. APPLICATION

We have applied the model to the absorption of a large spectral band radiation pencil propagating in a hydrogen gas at conditions of optically thick magnetic fusion plasmas [3,31–35]. The first transition (Lyman α, $2\to 1$) is discussed here. The time evolution of the mean photon number $N_j(t) = \mathrm{Tr}_R[a_j^\dagger a_j \rho_R(t)]$ has been calculated from the master equation (26). Only diagonal matrix elements in the radiation's Fock space have been retained. For simplicity, we have assumed a homogeneous medium with space- and time-independent atomic parameters, and emission processes have not been retained. In this framework, the mean photon number obeys an integro-differential equation with a convolution term

$$\frac{dN_j}{dt} = -c\int_0^t d\tau\,\bar{\chi}(\omega_j,\tau)N_j(t-\tau). \tag{39}$$

The kernel is given by

$$\bar{\chi}(\omega_j,\tau) = N_g\frac{\hbar\omega_0 B_{ge}}{4\pi^2}\mathrm{Re}[C_D(\tau)e^{-i\omega_j\tau}], \tag{40}$$

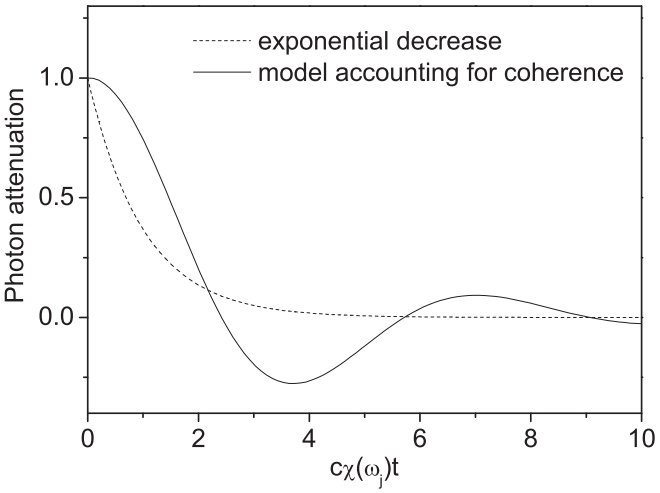

FIG. 1. Photon attenuation $N_j(t)/N_j(t=0)$ at the mode corresponding to the central frequency of Lyman α, at optically thick divertor plasma conditions (see text). The time is set in units of the photon time of flight, estimated as $1/c\chi(\omega_j)$ and of the order of 10^{-12} s here. The photon attenuation deviates from a pure exponential decrease due to coherence. Negative values are a feature of the ambiguity in the definition of a photon number at times smaller than the coherence time.

where $C_D(\tau) = C(\tau)\exp(-\Delta\omega_D^2\tau^2/4)$ accounts for the Doppler broadening [$\Delta\omega_D \equiv \omega_0 v_0/c$ stands for the Doppler width associated with the thermal velocity $v_0 = (2T_{\rm at}/m_{\rm at})^{1/2}$]. A formal solution of Eq. (39) can be obtained from Laplace transform techniques. The photon attenuation at time t is given by the following relation:

$$\frac{N_j(t)}{N_j(t=0)} = \frac{1}{2i\pi}\int_{\mathcal{C}} \frac{ds e^{st}}{s + \frac{c}{2}[\chi_c(\omega_j - is) + \chi_c^*(\omega_j - is^*)]}. \tag{41}$$

Here $\mathcal{C}$ is the vertical Bromwich contour and χ_c is the complex extinction coefficient introduced in Sec. II. The integral (41) has been evaluated numerically. Figure 1 shows the photon attenuation at the line center ($\omega_j = \omega_0$) assuming a density of absorbers of 2×10^{14} cm^{-3}, with the temperature $T_{\rm at} = 1$ eV. Only Doppler broadening has been retained. At these conditions, the coherence time is of the same order as the photon time of flight [estimated as $1/c\chi(\omega_j)$; it provides a characteristic relaxation time]. As can be seen, the attenuation curve is distorted and exhibits oscillations when coherence is retained. Negative values are a feature of the ambiguity in the definition of a photon number at times smaller than the coherence time, in the same way as an ambiguity occurs in the localization of a wave packet at a spatial scale shorter than the coherence length. This result is in agreement with heuristic arguments involving the Fourier time-frequency uncertainty relation. It suggests that time-dependent radiation transport models can be inaccurate if the coherence time is comparable to or larger than a relevant relaxation time in the problem under consideration. It is instructive to examine an ideal case where the dipole autocorrelation function is an exponential (e.g., due to collisions) and Doppler broadening is not retained. The integral (41) can be calculated explicitly:

$$\frac{N_j(t)}{N_j(t=0)} = \frac{1+\sqrt{1-4c\chi_0/\gamma}}{2\sqrt{1-4c\chi_0/\gamma}} e^{-\gamma(1-\sqrt{1-4c\chi_0/\gamma})t/2} - \frac{1-\sqrt{1-4c\chi_0/\gamma}}{2\sqrt{1-4c\chi_0/\gamma}} e^{-\gamma(1+\sqrt{1-4c\chi_0/\gamma})t/2}. \tag{42}$$

The dimensionless parameter $c\chi_0/\gamma$ denotes the ratio between the coherence time $1/\gamma$ and the photon time of flight $1/c\chi_0$ [with $\chi_0 \equiv \chi(\omega_0)$]. As can be seen, the photon attenuation deviates from the pure exponential decrease $\exp(-c\chi_0 t)$ due to the radiation coherence. The presence of an imaginary part in the exponentials as soon as $c\chi_0/\gamma$ exceeds $1/4$ indicates damped oscillations. This result is qualitatively in agreement with the case where Doppler broadening is considered. At the limit of strong coherence ($c\chi_0/\gamma \to \infty$), the photon attenuation behaves as

$$\frac{N_j(t)}{N_j(t=0)} \sim e^{-\gamma t/2}\cos\left(\sqrt{\frac{c\chi_0}{\gamma}}\gamma t\right), \tag{43}$$

which indicates damped oscillations with a damping constant of the order of the inverse coherence time.

VI. CONCLUSION

In this work, we have developed an extension of the radiative transfer theory suitable for the modeling of coherent radiation with narrow-band spectrum. The coherence length and time are interpreted as space and time scales characteristic to photon-atom interaction processes. In the large spectral band limit, these quantities are formally treated as arbitrarily small parameters and the interactions are local. If a relevant relaxation time or gradient length is sufficiently small so as to be comparable to the coherence time or length, the locality assumption becomes invalid. The nonlocality owing to spatial coherence can be addressed through a quantum phase-space description of photons involving Wigner functions. The temporal coherence directly affects the photon-atom master equations and requires a reconsideration of basic assumptions such as Markovianity. With an adaptation of the BBGKY hierarchy, we have shown that temporal coherence yields an ambiguity in the definition of a characteristic duration of photon-atom interactions.

An application to magnetic fusion plasma conditions has indicated an alteration of the formation of spectral lines, suggesting a possible inaccuracy in standard radiative transfer models. These results are still qualitative and require further examination with confrontations to experiments. A possible extension of the work could involve the analysis of atomic lines observed in discharge lamps. Specific experiments carried out on excimer light sources and dielectric barrier discharges have indicated very clean spectra of the Lyman α line [36–39] that would serve as a test of the model. Another domain of application includes the characterization of ultrashort laser pulses and their interaction with an absorbing or amplifying medium. Recent investigations have indicated an interplay between the pulse duration and the probability of radiation absorption [40]. This issue could be addressed using the radiation transport model presented here. On the theoretical side, an extension of the work should concern the link between

the present model and alternative first-principles approaches to radiative transfer that involve the quantized Maxwell equations and related atomic operators in the Heisenberg picture (e.g., [41,42]). Such approaches are convenient for an interpretation in terms of classical electromagnetism. Recent works have also shown their applicability in problems involving two-photon correlations (see [43]).

APPENDIX A: BEYOND THE WEAK-COUPLING LIMIT

We present hereafter an extension of the closure relation $G_1(1,t) = -i\int_0^t d\tau \exp\{-i[\hat{L}_R + \hat{L}_A(1)]\tau\}\hat{V}(1)[\bar{F}_1(1,t-\tau)F_0(t-\tau)]$ performed in Sec. IV, which is designed to account for correlations between atoms. The method involves a finite expression for G_2 and is inspired from renormalization techniques in quantum kinetic theory [26,27]. It is also inspired from previous works in plasma spectroscopy (Stark line shape) modeling [44–47]. We assume that quantities proportional to $\mathcal{N}\delta V$ can be of an arbitrary order of magnitude, using that $\mathcal{N}$ is large, while the perturbation δV is still considered as a small parameter (in the terminology of [45], the model retains the cumulative effect of weak individual interactions). The evolution of the G_1 correlation at the first order in δV is given by the following equation:

$$\left\{\frac{d}{dt} + i\left[\hat{L}_R + \hat{L}_A(1)\right]\right\} G_1(1) = -i\hat{V}(1)[\bar{F}_1(1)F_0] - i\mathcal{N}\mathrm{Tr}_2[\hat{V}(2)G_2(1,2)]. \quad \text{(A1)}$$

Here, the Hartree-Fock contributions $\hat{\bar{V}}_R$ and $\hat{\bar{V}}(1)$ have not been retained and the substitution $\delta\hat{V} \equiv \hat{V}$ has been done for the sake of simplicity. The $\bar{G}_2$ correlation is also neglected. The contribution of G_2 is described using the following model:

$$i\mathcal{N}\mathrm{Tr}_2[\hat{V}(2)G_2(1,2)] \equiv \hat{K}G_1(1) + [\hat{K}G_1(1)]^\dagger, \quad \text{(A2)}$$

where $\hat{K}$ is a linear superoperator nonlocal in time (i.e., involving a time integral) and interpreted as a self-energy contribution to the Hamiltonian H_R. It is defined implicitly by identification with self-energy terms in Eq. (21). To determine an explicit definition we first write the formal solution of Eq. (A1) as

$$G_1(1,t) = -\frac{i}{\hbar}\int_0^t dt' Q_1(1,t,t')[V(1),\bar{F}_1(1,t')F_0(t')] \times Q_1^\dagger(1,t,t'). \quad \text{(A3)}$$

The operator $Q_1(1,t,t')$ is a propagator defined by

$$\left\{\frac{\partial}{\partial t} + \frac{i}{\hbar}[H_R + H_A(1)] + \hat{K}\right\} Q_1(1,t,t') = \delta(t-t'). \quad \text{(A4)}$$

Inserting the formal solution (A3) in the right-hand side of Eq. (21) yields four terms resulting from the double commutator

$$\left(\frac{d}{dt} + i\hat{L}_R\right) F_0(t) = -\frac{\mathcal{N}}{\hbar^2}\mathrm{Tr}_1\int_0^t dt' V(1)Q_1(1,t,t')V(1)\bar{F}_1(1,t') \times F_0(t')Q_1^\dagger(1,t,t') + \frac{\mathcal{N}}{\hbar^2}\mathrm{Tr}_1\int_0^t dt' V(1)Q_1(1,t,t') \times \bar{F}_1(1,t')F_0(t')V(1)Q_1^\dagger(1,t,t') + \text{H.c.} \quad \text{(A5)}$$

H.c. stands for the Hermitian conjugate. The first term of the right-hand side and its conjugate can be combined with the Hamiltonian H_R (implied in the Liouvillian $\hat{L}_R$) and interpreted as self-energy contributions, which provides a definition for $\hat{K}$

$$\hat{K}F_0(t) = \frac{\mathcal{N}}{\hbar^2}\mathrm{Tr}_1\int_0^t dt' V(1)Q_1(1,t,t') \times V(1)\bar{F}_1(1,t')F_0(t')Q_1^\dagger(1,t,t'). \quad \text{(A6)}$$

An approximation suitable for calculations consists in assuming the $\hat{K}$ superoperator local in time and assimilating it to an operator, $\hat{K} \equiv K$. This can be done through the substitution $\bar{F}_1(1,t')F_0(t')Q_1^\dagger(1,t,t') \to \exp\{i[H_R + H_A(1)](t-t')/\hbar\}\bar{F}_1(1,t)F_0(t)$ in Eq. (A6). The resulting K operator is defined as

$$K(t) = \frac{\mathcal{N}}{\hbar^2}\mathrm{Tr}_1\int_0^t dt' V(1)Q_1(1,t,t') \times V(1)e^{\frac{i}{\hbar}[H_R+H_A(1)](t-t')}\bar{F}_1(1,t), \quad \text{(A7)}$$

and can be further simplified as

$$K(t) \simeq \frac{\mathcal{N}}{\hbar^2}\mathrm{Tr}_1\int_0^\infty d\tau V(1)e^{\{-\frac{i}{\hbar}[H_R+H_A(1)]-K(t)\}\tau} \times V(1)e^{\frac{i}{\hbar}[H_R+H_A(1)]\tau}\bar{F}_1(1,t), \quad \text{(A8)}$$

using the approximation $Q_1(1,t,t') \simeq \exp(\{-i[H_R + H_A(1)]/\hbar - K(t)\}(t-t'))$ and taking the $t \to \infty$ limit in the integral's upper bound. Equation (A8) is not a closed expression for K due to the presence of this term in the exponential. In practice, a calculation can be done by iterations, using Eq. (A8) with $K \equiv 0$ in the right-hand side as an initialization.

The master equation resulting from this model has the same formal structure as Eq. (5) with the substitution $[e^{-iH_0\tau/\hbar}Ve^{iH_0\tau/\hbar}, \rho_A\rho_R] \to e^{-i(H_0/\hbar - iK)\tau}Ve^{iH_0\tau/\hbar}\rho_A\rho_R -$ H.c. Physically, the presence of K in the exponential denotes an interruption of emitted or absorbed radiation wave packets owing to the interactions with atoms located at a distance of the order of the photon mean free path. It can be significant if the radiation is coherent. Equation (A8) presents similarities with the result of the so-called resonance broadening theory used for plasma turbulence (e.g., in [48], see the analogy with the implicit definition of the diffusion coefficient).

APPENDIX B: PARTIAL FREQUENCY REDISTRIBUTION

The formalism presented above can be adapted to problems involving radiation frequency redistribution. We give here an illustration in an ideal case. A set of two-level atoms with constant and homogeneous phase-space density for the ground state [i.e., $f_g(\mathbf{r},\mathbf{v},t) \equiv f_g(\mathbf{v})$] is considered. In this framework, we can write a closed set of equations for the photon spectral density $N(\omega,\mathbf{n},t) = \int d^3r\, W(\mathbf{r},\mathbf{p},t)\hbar p^2/c$

[unit: $(\text{rad/s})^{-1}\text{sr}^{-1}$; here, by convention $\mathbf{p} = \hbar\omega\mathbf{n}/c$] and the space-integrated atomic phase-space density $F_e(\mathbf{v},t) = \int d^3r f_e(\mathbf{r},\mathbf{v},t)$:

$$\frac{\partial N}{\partial t}(\omega,\mathbf{n},t) = \frac{1}{\hbar\omega_0}\int_0^t d\tau\,\bar{\eta}(\omega,t,\tau) - c\int_0^t d\tau\,\bar{\chi}(\omega,\tau)N(\omega,\mathbf{n},t-\tau), \quad \text{(B1)}$$

$$\frac{\partial F_e}{\partial t}(\mathbf{v},t) = -(A_{eg}+C_{eg})F_e(\mathbf{v},t) + c\int d\omega'\int d\Omega' \times\int_0^t d\tau\,\tilde{\chi}(\omega',\mathbf{n}',\mathbf{v},\tau)N(\omega',\mathbf{n}',t-\tau) + C_{ge}F_g(\mathbf{v}). \quad \text{(B2)}$$

Equation (B1) is obtained from space integration of the quantum radiative transfer equation (13) with the source and loss terms (29) and (30), and the terms $\bar{\eta}$ and $\bar{\chi}$ are defined by

$$\bar{\eta}(\omega,t,\tau) = \int d^3v F_e(\mathbf{v},t-\tau)\frac{\hbar\omega_0 A_{eg}}{4\pi^2}\text{Re}[C(\tau)e^{-i\omega(1-\mathbf{n}\cdot\mathbf{v}/c)\tau}], \quad \text{(B3)}$$

$$\bar{\chi}(\omega,\tau) = \int d^3v\,\tilde{\chi}(\omega,\mathbf{n},\mathbf{v},\tau), \quad \text{(B4)}$$

with

$$\tilde{\chi}(\omega,\mathbf{n},\mathbf{v},\tau) = f_g(\mathbf{v})\frac{\hbar\omega_0 B_{ge}}{4\pi^2}\text{Re}[C(\tau)e^{-i\omega(1-\mathbf{n}\cdot\mathbf{v}/c)\tau}]. \quad \text{(B5)}$$

Equation (B2) stems from space integration of the evolution equation for the atoms in the upper state, Eq. (38). The C coefficients denote collision rates. For the sake of simplicity, we neglect their dependence on velocity. We also assume that the population of the excited level is much smaller than that of the ground state ($N_e \ll N_g$), so that stimulated emission is negligible.

A reformulation of the photon transport equation (B1) with explicit mention to scattering can be obtained by solving formally Eq. (B2), separating the contributions of photoexcitation and collisional excitation, and inserting the solution in the source term (B3). If we neglect retardation effects on the photon density [viz., formally setting $N(\omega',\mathbf{n}',t-\tau) \simeq N(\omega',\mathbf{n}',t)$ in the integral] and if we take the large time limit, the resulting photon transport equation becomes similar to models involving partial frequency redistribution (e.g., [1])

$$\frac{\partial N}{\partial t}(\omega,\mathbf{n},t) = -c\chi(\omega)N(\omega,\mathbf{n},t) + \frac{\mathcal{N}_g C_{ge}}{A_{eg}+C_{eg}}\frac{A_{eg}}{4\pi}\phi(\omega) + \frac{A_{eg}}{A_{eg}+C_{eg}}\frac{cN_g\hbar\omega_0 B_{ge}}{4\pi}\int d\omega' \times\int\frac{d\Omega'}{4\pi}R(\omega,\mathbf{n},\omega',\mathbf{n}')N(\omega',\mathbf{n}',t). \quad \text{(B6)}$$

The redistribution function $R(\omega,\mathbf{n},\omega',\mathbf{n}')$ is defined here by

$$R(\omega,\mathbf{n},\omega',\mathbf{n}') = \frac{1}{\pi^2}\int d^3v F_g(\mathbf{v})\text{Re}\int_0^\infty d\tau C(\tau)e^{-i\omega(1-\mathbf{n}\cdot\mathbf{v}/c)\tau} \times\text{Re}\int_0^\infty d\tau' C(\tau')e^{-i\omega'(1-\mathbf{n}'\cdot\mathbf{v}/c)\tau'}. \quad \text{(B7)}$$

In the case where only Doppler broadening is retained, the time integrals are proportional to δ functions and the redistribution function reduces to the R_I function introduced by Hummer [49].

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