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Time-Dependent Behavior of a Class of Nonequilibrium Radiating Systems*

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A kinetic-theoretic approach is employed to analyze the temporal behavior of simple radiating systems that are, generally, not at equilibrium. The system models considered herein are simple in the sense that streaming of radiation and particles (hydrogenic atoms) is neglected, relativistic effects are not considered, and bound-bound radiative transitions are taken to be the dominant interaction process. Of major concern is the manner in which physically interpretable factors bear on the approach of mean (singlet) densities of atoms in various states and of photons to their steady-state values. Additionally, the present work is of sufficient generality, statistically, to allow the retention of higher-order densities. That is, mean square (doublet) densities for the atoms in their various states and for the radiation (this includes joint doublet densities between species) are explicitly included. These additional terms comprise the vehicle that statistically couples the various distributions in a system to the next-higher order (than the first). The cases for which solutions have been obtained are parametrized by radiation linewidths, by statistical weights of atomic levels, and, of course, by initial conditions on the atomic and radiation densities. Major trends and characteristic results are noted and mechanistically discussed; and elemental limits on the application and interpretation of the kinetic theory, *per se*, for this class of radiating systems are indicated.

I. INTRODUCTION

The primary object of the present work is to examine the temporal evolution of a class of radiating systems that are, in general, not at equilibrium. To study the approach to equilibrium – that is, the approach to a steady state for the mean (singlet) densities of atoms in various states and of the radiation – appropriate sets of kinetic equations are developed and solved. In addition to writing and solving a somewhat conventional set of kinetic equations for the singlet densities mentioned above, it is also a straightforward matter to extend the treatment to include second-order (doublet) densities, the relevance being that the doublet densities for particles and photons[1] can be viewed as statistical coupling terms. These additional terms explicitly relate (to the next-higher statistical order) the manner in which the various particle and radiation distributions jointly fluctuate owing to the interaction processes that obtain in a given system. Accordingly, solutions can be obtained not only for the temporal evolution of the singlet densities but also for the temporal evolution of atomic and radiation fluctuations in systems that are not at equilibrium.

To focus primary attention upon time-dependent behavior and upon elemental radiative mechanisms that underlie departures from thermodynamic equilibrium, the system models in the present work have purposely been made simple. They are simple in the sense that relativistic effects are neglected, and the system is assumed to be sufficiently large and uniform so that losses at the boundaries and streaming of particles and photons may be neglected. Bound-bound transitions of two-level hydrogen atoms are taken to be the dominant interaction process. On the other hand, the kinetic equations themselves are deduced with sufficient generality to allow the retention of effects due to the finite lifetimes of emitting and absorbing atomic states. Indeed, finite lifetimes will be found to assume a central importance with regard to the dynamic evolution of a given system, inasmuch as linewidth considerations can be seen to be an integral element of the internal consistency of the kinetic theory, *per se*. Statistical weights associated with atomic states are also included in general terms, and their pertinence to dynamic features as well as to final distributions at steady state is assessed in specific cases. Thus present consideration is simply directed to a set of models that is both explicitly solvable and yet possessed of some practical and theoretical utility. This set of collisionless models will apply in a practical sense to rare systems (such as regions of interstellar space where particle densities may be as low as 10^2–10^3 per cm^3), because in this regime radiative rates are much greater than collisional rates. As particle densities increase to the order of, say, 10^{12}–10^{16} per cm^3(corresponding to some laboratory plasmas and to some regions of some stellar interiors), it will be seen that the purely radiative behavior will depart from that deduced for rare systems. It is, of course, recognized that, in this regime of higher particle densities, the role of collisional processes becomes increasingly important. But the characteristic behavior of simple collision-dominated systems can be readily determined, and it is recognized that the radiation simply assumes a Planck distribution at the initial temperature of the atoms (which, of course, have their levels populated in accordance with Boltzmann's law[2]). A collisionless model then becomes of interest in a theoretical sense for non-rare systems, since radiative processes constitute

an elemental means by which the simple Planckian behavior above may be redistributed to other temperatures, temporally and asymptotically. But, in these more general instances, the temperature at any given time is merely a defined quantity whose value depends upon system parameters such as linewidths, statistical weights, and particle and radiation densities. Thus the present program is primarily devoted to the very basic problem of determing the evolution of a nonequilibrium system by explicit means – i.e., by solving appropriate sets of coupled kinetic equations *without a priori* assumption regarding the form of any of the distribution functions subsequent to time zero.

The format for this initial program is to develop the requisite kinetic equations (to second statistical order) in Sec. II. Section III reviews examples leading to simple Planckian behavior, and Sec. IV deals with systems that depart from the simple behavior of Sec. III. Results for specific examples are obtained in Sec. IV, both with the terms of higher statistical order retained and with them neglected.

II. THE KINETIC EQUATIONS

The method by which the forthcoming kinetic equations are deduced has been presented in some detail in numerous contexts[3,4] by now. Therefore only the very essential elements of the formulation will be reiterated. Perhaps the most elemental point to be made is that we are dealing with systems in which a number of quantum aspects are manifestly evident. The fact that particles in various states, as well as photons, are being created and destroyed suggests the utility of a second-quantized formulation – since the observables of our theory are expectation values of numbers (or products of numbers) of particles in various states, and/or photons, at a given time, t. The quantum-statistical axioms are well-suited to this description and are accordingly invoked. The temporal evolution of the observables is obtained from the Liouville equation

$$\partial\rho(t)/\partial t = (i/\hbar)[\rho,H] \quad , \tag{1}$$

where $\rho(t)$ is the density operator for a system of interest, and H is the Hamiltonian of the system. Furthermore atomic states most often have finite lifetimes, which are, in turn, manifest in corresponding nonzero radiation linewidths. Thus, in view of uncertainty requirements, the momentum uncertainty related to a nonzero radiation linewidth leads to a minimum value for the dimensions (in configuration space) of a fundamental phase cell. In radiative systems, these dimensions can be of macroscopic size. The analytic accomodation of the phase cell concept into a kinetic-theoretic formulation is, therefore, not only an important matter of principle, but is indeed of considerable practical significance as well, particularly when fluctuations are included. The singlet density for particles or photons of kind A is therefore defined as

$$F_1^A(\vec{X},\vec{K},a,t) \equiv [1/(2\pi)^3]\,\mathrm{Tr}\hat{N}^A(\vec{X},K,a)\rho(t) \quad , \tag{2}$$

where the coordinates $(\vec{X},\vec{K})$ are coarse-grained coordinates in that they merely locate the center of a phase cell of volume $(2\pi)^3$ in the six-dimensional phase space (μ space). If a particle or photon is located anywhere within a phase cell, its coordinates are designated simply by the location of the center of the cell. The label a is used to designate the quantum numbers that complete the description of the particle's or photon's state. The number operator $\hat{N}^A(\vec{X},\vec{K},a)$ is defined by

$$\hat{N}^A(\vec{X},\vec{K},a) \equiv \alpha_A{}^\dagger(\vec{X},\vec{K},a)\alpha_A(\vec{X},\vec{K},a), \tag{3}$$

where the creation and annihilation operators, $\alpha_A{}^\dagger(\vec{X},\vec{K},a)$ and $\alpha_A(\vec{X},\vec{K},a)$, respectively, obey the commutation or anticommutation relationships appropriate to the A-type particles or photons. In a diagonalizing representation, the eigenvalues of $\hat{N}^A(\vec{X},\vec{K},a)$ are the possible numbers of A-type particles or photons in the cell about $(\vec{X},\vec{K})$ with quantum labels a. The doublet density for particles of kinds A and B is defined by

$$F_2^{AB}(\vec{X},\vec{K},a;\vec{X}',\vec{K}',b;t) \equiv [1/(2\pi)^6]\,\mathrm{Tr}\hat{N}^A(\vec{X},\vec{K},a)\hat{N}^B(\vec{X}',\vec{K}',b)\rho(t) \quad . \tag{4}$$

The temporal evolution for a singlet density is given by the finite difference expression,

$$[F_1^A(\vec{X},\vec{K},a,t+\tau) - F_1^A(\vec{X},\vec{K},a,t)]/\tau = [1/(2\pi)^3]\,\mathrm{Tr}\hat{N}^A(\vec{X},\vec{K},a)[\rho(t+\tau)-\rho(t)]/\tau \quad . \tag{5}$$

This expression approaches $\partial F_1^A(\vec{X},\vec{K},a,t)/\partial t$ for sufficiently small τ, such that densities do not vary too rapidly over the interval. On the other hand, τ has a lower limit, on the order of effective interaction times or lifetimes of states, in that densities are not meaningfully defined at two times closer together than these characteristic times. The temporal evolution of doublet densities is then expressed as

$$[F_2^{AB}(\vec{X},\vec{K},a;\vec{X}',\vec{K}',b;t+\tau) - F_2^{AB}(\vec{X},\vec{K},a;\vec{X}',\vec{K}',b;t)]/\tau = [1/(2\pi)^6]\,\mathrm{Tr}\hat{N}^A(\vec{X},\vec{K},a)\hat{N}^B(\vec{X}',\vec{K}',b)[\rho(t+\tau)-\rho(t)]/\tau. \tag{6}$$

From here it is a straightforward, but somewhat tedious, task to deduce the requisite kinetic equations. Proceeding as did Gelinas and Osborn,[3-5] space- and time-dependent kinetic equations are obtained. In order to reduce to a strictly time-dependent description, all streaming terms in all of the kinetic equations are neglected, and singlet and doublet densities are spatially averaged over the volume of the system, denoted by $\mathcal{V}_S$, as

$$F_1^A(\vec{K}, a, t) \equiv [1/\mathcal{V}_S] \int_{\mathcal{V}_S} d^3X \, F_1^A(\vec{X}, \vec{K}, a, t) \tag{7}$$

$$\text{and} \quad F_2^{AB}(\vec{K}, a; \vec{K}', b; t) \equiv [1/(\mathcal{V}_S)^2] \int_{\mathcal{V}_S} d^3X \int_{\mathcal{V}_S} d^3X' F_2^{AB}(\vec{X}, \vec{K}, a; \vec{X}', \vec{K}', b; t), \tag{8}$$

where the notation of the space-averaged (or "system-averaged") quantities is accomplished by simply dropping the spatial variables from the arguments. The kinetic equations can then be written in the general form (for A-type particles and/or photons)

$$\frac{dF_1^A(\vec{K}, a, t, \tau)}{dt} = \frac{1}{(2\pi)^3 \mathcal{V}_S} \int_{\mathcal{V}_S} d^3X \sum_{n,n'} W_{n'n}(\tau) [N_{n'n'}^A(\vec{X}, \vec{K}, a) - N_{nn}^A(\vec{X}, \vec{K}, a)] \rho_{nn}(t), \tag{9}$$

$$\text{and} \quad \frac{dF_2^{AB}(\vec{K}, a; \vec{K}', b; t, \tau)}{dt} = \frac{1}{(2\pi)^6 \mathcal{V}_S^2} \int_{\mathcal{V}_S} d^3X \int_{\mathcal{V}_S} d^3X' \sum_{n,n'} W_{n'n}(\tau)$$

$$\times [N_{n'n'}^A(\vec{X}, \vec{K}, a) N_{n'n'}^B(\vec{X}', \vec{K}', b) - N_{nn}^A(\vec{X}, \vec{K}, a) N_{nn}^B(\vec{X}' \vec{K}', b)] \rho_{nn}(t). \tag{10}$$

The time derivative of the singlet and doublet densities has been used to denote the more rigorous finite difference expressions of Eqs. (5) and (6). The indices n and n' denote the initial and final states, respectively, that are available in a given system. The terms $N_{n'n'}^A(\vec{X}, \vec{K}, a)$ and $N_{n'n'}^B(\vec{X}' \vec{K}', b)$ are simply the matrix representatives of $\hat{N}^A(\vec{X}, \vec{K}, a)$ and $\hat{N}^B(\vec{X}', \vec{K}', b)$ for the available final states in a diagonalizing representation of particle and photon numbers. Likewise, $N_{nn}^A(\vec{X}, \vec{K}, a)$, $N_{nn}^B(\vec{X}', \vec{K}', b)$, and $\rho_{nn}(t)$ are the matrix representatives of $\hat{N}^A(\vec{X}, \vec{K}, a)$, $\hat{N}^B(\vec{X}', \vec{K}', b)$, and $\rho(t)$ for the available initial states of a system. Off-diagonal matrix elements of $\rho(t)$ have been neglected. The quantity $W_{n'n}(\tau)$ is defined by

$$W_{n'n}(\tau) \equiv (1/\tau) \left| [e^{-(i/\hbar)(H_0 + I)\tau}]_{n'n} \right|^2, \tag{10}$$

where H_0 is comprised of those terms in the system Hamiltonian representative of the unperturbed (free) radiation field, plus the free-particle contributions. The term I is comprised of the "interaction terms" of the system Hamiltonian (see Ref. 5 for calculational details of this development). The equations, as they are written up to this point, are in a sufficiently general form to accommodate a description of a broad class of systems. To focus attention on the simple class of radiating systems of present concern, it is convenient here to adopt a particular system model that corresponds to a sufficiently large, uniform, homogeneous system of two-level hydrogen atoms and an isotropic radiation distribution. Bound-bound radiative transitions are taken to be the dominant interaction process. Since it is intended to eventually discuss matters in quantitative terms, Lyman-α transitions will be taken to be a representative bound-bound radiative transition in a two-level hydrogen-atom model. There is, of course, no great difficulty in extending the treatment to a larger number of arbitrary levels. So, for present purposes, the kinetic equations will be reduced to a form that applies to atoms in the $2P$ and $1S$ levels and the corresponding line radiation. Inasmuch as excited atomic states have finite lifetimes, damping theory[6,7] is invoked to evaluate the terms $W_{n'n}(\tau)$ – thereby deductively accounting for finite radiation linewidths in the present formulation. It will be convenient to sum atomic singlet and doublet densities over momenta (recall that we are working in a discrete, coarse-grained coordinate system). We will also assume photon polarizations to be random, and to pass to the continuum in frequency for the radiation. Assuming all singlet and doublet densities to be spatially constant, the equation for the singlet density of photons can be written as

$$\frac{df_1^R(\nu, t)}{dt} = M(\nu) g(\nu) \left[\frac{F_1^{Au}(2P, t)}{g_u} \rho_\nu + \left(\frac{F_1^{Au}(2P, t)}{g_u} - \frac{F_1^{Al}(1S, t)}{g_l} \right) f_1^R(\nu, t) + \frac{v^{AuR}(2P, \nu, t)}{g_u} - \frac{v^{AlR}(1S, \nu, t)}{g_l} \right], \tag{11}$$

where[8] $f_1^R(\nu,t)d\nu$ is defined in the continuum as the "system-averaged" expected number of photons per unit volume in $d\nu$ about ν, at time t. The cross covariance of photons (in the continuum) with atoms in the upper state, denoted by $v^{AuR}(2P,\nu,t)$, is defined as

$$v^{AuR}(2P,\nu,t) \equiv f_2^{AuR}(2P,\nu,t) - F_1^{Au}(2P,t) f_1^R(\nu,t), \tag{12}$$

where $f_2^{AuR}(2P,\nu,t)d\nu$ is defined as the "system-averaged" expectation value of the product per unit (volume)^2 of the number of atoms in the upper state and the number of photons in $d\nu$ about ν, at time t. The quantity $v^{AlR}(1S,\nu,t)$ is defined in like manner for photons and atoms in the lower state ($1S$ in our examples). The quantities g_u and g_l are the statistical weight factors associated with the upper and lower atomic states and take values of $(2l+1)$, where l is the azimuthal quantum number for the atomic state of interest. The origin of the factors g_u and g_l, as they appear herein, is derived from the assumption of natural excitation of the ml quantum states, in the absence of external fields. That is, by designating the internal states of the atom by the quantum labels n, l, and m_l, we have written

$$\hat{N}(\vec{X},\vec{K},n,l,m_l) = (1/2l+1)\hat{N}(\vec{X},\vec{K},n,l).$$

The spin orientation, often denoted by the label m_s, of the bound electron has been assumed to be random. The quantity $M(\nu)$ is given by

$$M(\nu) = (4\pi^2 e^2 \nu/3\hbar)(R_{nl}^{n'l'})^2, \tag{13}$$

$$\text{where} \quad [R_{nl}^{n'l'}]^2 \equiv \left[\int_0^\infty R_{n'l'}(r) R_{nl}(r) r^3 dr\right]^2, \tag{14}$$

and $R_{nl}(r)$ is the radial eigenfunction for the initial state designated by the quantum numbers nl (primes denote the final state). The density of available photon states, ρ_ν, is defined by

$$\rho_\nu \equiv 8\pi\nu^2/c^3. \tag{15}$$

The quantity $s(\nu)$ is a line-shape factor arising from the damping theoretic computation of $W_{n'n}(\tau)$. If γ_i and γ_f represent the widths associated with the lifetimes of the initial and final atomic states, respectively, and $h\nu_{ul}$ the mean energy difference between the upper and lower atomic states, it is readily shown that $s(\nu)$ approaches $\delta(\nu-\nu_{ul})$ when $\gamma_i\tau \ll 1$, $\gamma_f\tau \ll 1$, and $\tau > 1/2\pi\nu_{ul}$. This, of course, corresponds to the result obtained from first-order perturbation theory. But these conditions are too restrictive for our purposes, inasmuch as our kinetic equations are to be used over time scales that may very often be long compared to the lifetimes of excited states. On the other hand, the more general considerations of damping theory lead to a Lorentzian line-shape factor (normalized to unity) with only the conditions $\gamma_i\tau \ll 1$, $\gamma_f\tau \gg 1$ (or vice versa). This more suitably describes the physics of the systems of present concern, and the interpretability of the kinetic equations is likewise improved. The equations for $F_1^{Au}(2P,t)$ and $F_1^{Al}(1S,t)$ are written as

$$dF_1^{Au}(2P,t)/dt = \int d\nu\, M(\nu) s(\nu) \Big\{ -[\rho_\nu + f_1^R(\nu,t)] F_1^{Au}(2P,t)/g_u + (F_1^{Al}(1S,t)/g_l) f_1^R(\nu,t) + v^{AlR}(1S,\nu,t)/g_l - v^{AuR}(2P,\nu,t)/g_u \Big\} \tag{16}$$

$$\text{and} \quad dF_1^{Al}(1S,t)/dt = -dF_1^{Au}(2P,t)/dt. \tag{17}$$

Since the equations for the particle distributions are dependent upon integrals over radiation distributions (singlets and doublets as well as cross doublets), it is convenient to also integrate equations for radiation distributions over the line of interest. To simplify the calculations, the Lorentzian shape factor is replaced by a unit impulse function centered about ν_{ul}. That is, for a line of width $\Delta\nu$, $s(\nu)$ is now taken to be

$$s(\nu) = [H(\nu - \nu_{ul} + \Delta\nu/2) - H(\nu - \nu_{ul} - \Delta\nu/2)]/\Delta\nu,$$

where $H(\nu - x)$ is the unit step function, having value unity for $\nu \geq x$ and zero for $\nu < x$. Of course, as $\Delta\nu$ approaches zero, $s(\nu)$ approaches $\delta(\nu - \nu_{ul})$. If we define

$$F_1^R(t) \equiv \int d\nu\, f_1^R(\nu,t), \tag{18}$$

$$V^{AuR}(2P,t) \equiv \int d\nu\, v^{AuR}(2P,\nu,t), \tag{19}$$

$$V^{AlR}(1S,t) \equiv \int d\nu\, v^{AlR}(1S,\nu,t), \tag{20}$$

and $$V^{RR}(t) \equiv \int d\nu \int d\nu' v^{RR}(\nu,\nu',t), \tag{21}$$

the set of kinetic equations reduces finally to the following solvable form:

$$\frac{dF_1^R(t)}{dt} = \frac{M(\nu_{ul})}{\Delta\nu}\left[\frac{F_1^{Au}(2P,t)}{g_u}\rho_{\nu_{ul}}\Delta\nu + \left(\frac{F_1^{Au}(2P,t)}{g_u} - \frac{F_1^{Al}(1S,t)}{g_l}\right)F_1^R(t) + \frac{V^{AuR}(2P,t)}{g_u} - \frac{V^{AlR}(1S,t)}{g_l}\right], \tag{22}$$

$$dF_1^{Au}(2P,t)/dt = -dF_1^R(t)/dt, \tag{23}$$

$$dF_1^{Al}(1S,t)/dt = -dF_1^{Au}(2P,t)/dt, \tag{24}$$

$$\begin{aligned}\frac{dV^{AuR}(2P,t)}{dt} = \frac{M(\nu_{ul})}{\Delta\nu}\Bigg[&\left(\frac{F_1^{Au}(2P,t) - g_u F_1^{Al}(1S,t)}{g_l} - \rho_{\nu_{ul}}\Delta\nu - F_1^R(t)\right)\frac{V^{AuR}(2P,t)}{g_u} \\ &+ \frac{[\rho_{\nu_{ul}}\Delta\nu + F_1^R(t)]\ V^{AuAu}(2P,2P,t)}{g_u} - \frac{F_1^R(t)V^{AuAl}(2P,1S,t)}{g_l} + \frac{F_1^R(t)V^{AlR}(1S,t)}{g_l} \\ &+ \left(\frac{F_1^{Al}(1S,t)}{g_l} - \frac{F_1^{Au}(2P,t)}{g_u}\right)V^{RR}(t) - \frac{1}{v_s}C(t)\Bigg],\end{aligned} \tag{25}$$

where $$C(t) \equiv \frac{F_1^{Au}(2P,t)}{g_u}\rho_{\nu_{ul}}\Delta\nu + \left(\frac{F_1^{Au}(2P,t)}{g_u} + \frac{F_1^{Al}(1S,t)}{g_l}\right)F_1^R(t) + \frac{V^{AuR}(2P,t)}{g_u} + \frac{V^{AlR}(1S,t)}{g_l}, \tag{26}$$

$$\begin{aligned}\frac{dV^{AlR}(1S,t)}{dt} = \frac{M(\nu_{ul})}{\Delta\nu}\Bigg[&\left(\frac{g_l F_1^{Au}(2P,t)}{g_u} - F_1^{Al}(1S,t) - F_1^R(t)\right)\frac{V^{AlR}(1S,t)}{g_l} + [\rho_{\nu_{ul}}\Delta\nu + F_1^R(t)]\frac{V^{AuR}(2P,t)}{g_u} \\ &+ [\rho_{\nu_{ul}}\Delta\nu + F_1^R(t)]\frac{V^{AuAl}(2P,1S,t)}{g_u} + \left(\frac{F_1^{Au}(2P,t)}{g_u} - \frac{F_1^{Al}(1S,t)}{g_l}\right)V^{RR}(t) - \frac{V^{AlAl}(1S,1S,t)}{g_l}F_1^R(t) + \frac{1}{v_s}C(t)\Bigg],\end{aligned} \tag{27}$$

$$\begin{aligned}\frac{dV^{RR}(t)}{dt} = \frac{M(\nu_{ul})}{\Delta\nu}\Bigg[&2\left(\frac{F_1^{Au}(2P,t)}{g_u} - \frac{F_1^{Al}(1S,t)}{g_l}\right)V^{RR}(t) + 2[\rho_{\nu_{ul}}\Delta\nu + F_1^R(t)]\frac{V^{AuR}(2P,t)}{g_u} \\ &- \frac{2F_1^R(t)V^{AlR}(1S,t)}{g_l} + \frac{1}{v_s}C(t)\Bigg],\end{aligned} \tag{28}$$

$$\begin{aligned}\frac{dV^{AuAl}(2P,1S,t)}{dt} = \frac{M(\nu_{ul})}{\Delta\nu}\Bigg[&-\left(\rho_{\nu_{ul}}\Delta\nu + \frac{F_1^R(t)(g_u+g_l)}{g_l}\right)\frac{V^{AuAl}(2P,1S,t)}{g_u} + \frac{F_1^R(t)V^{AlAl}(1S,1S,t)}{g_l} \\ &+ \left(\frac{F_1^{Au}(2P,t)}{g_u} - \frac{F_1^{Al}(1S,t)}{g_l}\right)V^{AuR}(2P,t) + \left(\frac{F_1^{Al}(1S,t)}{g_l} - \frac{F_1^{Au}(2P,t)}{g_u}\right)V^{AlR}(1S,t)\end{aligned}$$

$$+[\rho_{\nu_{ul}}\Delta\nu + F_1{}^R(t)]\frac{V^{AuAu}(2P,2P,t)}{g_u} - \frac{1}{v_s}C(t)\Bigg], \tag{29}$$

$$\frac{dV^{AuAu}(2P,2P,t)}{dt} = \frac{M(\nu ul)}{\Delta\nu}\Bigg[-2[\rho_{\nu_{ul}}\Delta\nu + F_1{}^R(t)]\frac{V^{AuAu}(2P,2P,t)}{g_u}$$

$$+2\left(\frac{F_1{}^{Al}(1S,t)}{g_l} - \frac{F_1{}^{Au}(2P,t)}{g_u}\right)V^{AuR}(2P,t) + \frac{2F_1{}^R(t)V^{AuAl}(2P,1S,t)}{g_l} + \frac{1}{v_s}C(t)\Bigg], \tag{30}$$

$$\frac{dV^{AlAl}(1S,1S,t)}{dt} = \frac{M(\nu_{ul})}{\Delta\nu}\Bigg[-\frac{2F_1{}^R(t)V^{AlAl}(1S,1S,t)}{g_l} + 2\left(\frac{F_1{}^{Au}(2P,t)}{g_u} - \frac{F_1{}^{Al}(1S,t)}{g_l}\right)V^{AlR}(1S,t)$$

$$+2[\rho_{\nu_{ul}}\Delta\nu + F_1{}^R(t)]\frac{V^{AuAl}(2P,1S,t)}{g_u} + \frac{1}{v_s}C(t)\Bigg]. \tag{31}$$

The terms $(1/v_s)C(t)$ in the above equations may be termed "self-correlation" terms. These terms are originally (i.e., before "system-averaging") nonzero only within a given phase cell, and space averaging the equations [see Eqs. (7) and (8)] accounts for the factor $(1/v_s)$. This merely indicates that, as the volume of the total system gets sufficiently large, contributions from a single phase cell ordinarily assume very little importance. Therefore in large systems, for which our present consideration of an infinite homogeneous model best applies, it is a good, and consistent, approximation to neglect the "self-correlation" terms. On the other extreme, the system volume may be on the order of the phase cell volume in configuration space. An important class of measurements in this regime are numerous optical experiments in which quantum fluctuations within a single phase cell are observable. For systems of this type, solutions can be deduced for Eqs. (22)–(31) with the "self-correlation" terms retained, but in the absence of spatial dependence such solutions should only be viewed inferentially, at most.

III. EXAMPLES THAT LEAD SIMPLY TO PLANCKIAN RESULTS

In order to lay out areas in which clear-cut trends characterize system behavior, it is well to first of all consider two sets of conditions that lead simply to a Planckian characterization at steady state – particularly since Planckian behavior is, in one way or another, associated with well-recognized notions that underlie a sizable segment of existing work on radiating systems. Accordingly the results of this section are neither surprising nor novel. They do, however, provide a logical framework for discussion of later results, and perhaps most important are the dynamic features that, with some extension, will also carry over to more general cases in the next section.

A. Collision-Dominated Systems

The main feature that characterizes a collision-dominated system is that the atomic populations in the upper and lower levels are held in a ratio given by Boltzmann's distribution law. In our examples this ratio is assumed to be constant, and is given by

$$F_1{}^{Au}(2P)/F_1{}^{Al}(1S) = g_u e^{-h\nu_{ul}/\theta}/g_l, \tag{32}$$

where θ is Boltzmann's constant multiplied by the temperature of the *atoms*. In sufficiently dense systems, atomic collisions can provide this condition to a good approximation. That is also to say that radiative effects are sufficiently weak in such instances so as not to appreciably perturb the atomic distributions from a Boltzmann ratio. If one neglects the cross-covariances $V^{AuR}(2P,t)$ and $V^{AlR}(1S,t)$, it is a straightforward matter to substitute Eq. (32) into Eq. (22) and obtain the following expression for the expected number of photons per unit volume in the line of width $\Delta\nu$ about ν_{ul} at time t:

$$F_1{}^R(t) = \frac{8\pi\nu_{ul}^2\Delta\nu(1-e^{-\Lambda t})}{c^3(e^{h\nu_{ul}/\theta}-1)} + F_1{}^R(0)\,e^{-\Lambda t}, \tag{33}$$

where $\Lambda \equiv M(\nu_{ul})F_1{}^{Au}(2P)(e^{h\nu_{ul}/\theta}-1)/g_u\Delta\nu$. (34)

If the result of Eq. (33) is divided by $\Delta\nu$ and multiplied by $hc\nu_{ul}$, it is evident that, asymptotically the conventional Planck distribution is approached at the *atomic* temperature. That is,

$$\lim_{t\to\infty} I(\nu_{ul},t) \to 8\pi h\nu_{ul}{}^3/c^2(e^{h\nu_{ul}/\theta}-1). \tag{35}$$

While the temporal evolution given by Eq. (33) is in a sense trivial, there is one feature of the results of Eq. (33) and (34) that is not often explicitly demonstrated. This refers to the dependence of the equilibration time upon some measure of the radiative level width (in this case $\Delta\nu$ is that measure). From the definition of Λ and the time de-

pendence given by Eq. (33), it is clear that this class of systems approaches equilibrium relatively faster for narrow radiative widths than for broader widths. This can be briefly explained mechanistically by noting from Eq. (22) that the ratio of photon emission to absorption is proportional to $[1+\rho_{\nu_{ul}}\Delta\nu/F_1{}^R(t)]$. Therefore, for $\Delta\nu$ relatively large (broad radiative line widths), emission processes become relatively more important with respect to absorption. For a given radiation density, this has the resultant effect of assigning relatively greater importance to the spontaneous emission process (because of the second term in the square brackets immediately above). But except for very rare systems (which will be taken up in Sec. IV), the spontaneous emission rate is slow in comparison to the rate of net absorption. It is therefore evident that, within the limits of present considerations, a collision-dominated system is expected to approach steady state relatively more slowly for broad radiative line widths than for narrow ones. Extensions of this result will be seen in later sections.

B. Collisionless Systems

In collisionless systems the radiation and atomic distributions are completely coupled by the dominance of radiative interactions. Therefore, given a set of initial conditions, the temporal evolution can be deduced by solving a sufficiently complete set of coupled equations. In this section, terms of second statistical order will be neglected, and Eqs. (22)–(24) can be solved directly for $F_1{}^R(t)$, $F_1{}^{Au}(2P,t)$, $F_1{}^{Al}(1S,t)$. It can first be noted from Eqs. (22)–(24) that the following conservation conditions obtain for the present models:

$$F_1{}^{Au}(2P,t)+F_1{}^{Al}(1S,t)=N_A\,, \tag{36}$$

$$F_1{}^{Au}(2P,t)+F_1{}^{R}(t)=c_1\,, \tag{37}$$

$$F_1{}^{Al}(1S,t)-F_1{}^{R}(t)=c_2\,, \tag{38}$$

where N_A is the total number density of atoms in the system. It also follows that

$$c_1+c_2=N_A\,. \tag{39}$$

By applying these conservation relationships to eliminate $F_1{}^{Au}(2P,t)$ and $F_1{}^{Al}(1S,t)$, Eq. (22) takes the form

$$dF_1{}^R(t)/dt=a[F_1{}^R(t)]^2+bF_1{}^R(t)+c\,, \tag{40}$$

where

$$a\equiv-[M(\nu_{ul})/g_u\Delta\nu](g_u+g_l)/g_l\,, \tag{41}$$

$$b\equiv\left[N_A-\rho_{\nu_{ul}}\Delta\nu-\frac{(g_u+g_l)}{g_l}c_2\right]\frac{M(\nu_{ul})}{g_u\Delta\nu}\,, \tag{42}$$

and

$$c\equiv[M(\nu_{ul})\rho_{\nu_{ul}}\Delta\nu/g_u\Delta\nu](N_A-c_2)\,. \tag{43}$$

of course, Eq. (40) can be written in the alternative form

$$dF_1{}^R(t)/dt=a[F_1{}^R(t)-p][F_1{}^R(t)-q]\,, \tag{44}$$

where

$$p=[-b+(b^2-4ac)^{1/2}]/2a \tag{45}$$

$$q=-[b+(b^2-4ac)^{1/2}]/2a\,. \tag{46}$$

The solution for $F_1{}^R(t)$ is then given by

$$F_1{}^R(t)=\frac{q[F_1{}^R(0)-p]-p[F_1{}^R(0)-q]e^{a(q-p)t}}{[F_1{}^R(0)-p]-[F_1{}^R(0)-q]e^{a(q-p)t}}\,. \tag{47}$$

It is evident that

$$\lim_{t\to\infty}F_1{}^R(t)\to q\,. \tag{48}$$

From Eqs. (41), (42), (46), and the conservation condition expressed by Eq. (38), it can be shown that q, and therefore the steady-state radiation density, approaches $F_1R(0)$ when $F_1R(0)$ is sufficiently large in comparison to N_A, the total number density of atoms in the system. That is, the number of photons created and destroyed by the available atoms is so small that the initial radiation distribution is, to good approximation, not affected by subsequent interaction with the system. By simultaneously solving for $F_1{}^{Au}(2P,t)$ and $F_1{}^{Al}(1S,t)$, it can be shown that

$$\lim_{t\to\infty}\frac{F_1{}^{Al}(1S,t)}{F_1{}^{Au}(2P,t)}=\frac{g_l}{g_u}e^{h\nu_{ul}/\theta_R}\,, \tag{49}$$

where

$$e^{h\nu_{ul}/\theta_R}\equiv[8\pi\nu_{ul}{}^2\Delta\nu/c^3F_1{}^R(0)]+1\,. \tag{50}$$

Thus it is noted that a sufficiently strong radiation source will force atomic level populations into a Boltzmann ratio at the temperature of the initial *radiation* distribution. A cursory view of the exponential factors in Eq. (47) shows that the terms $a(q-p)$ are manifestly negative and on the order of $F_1R(0)$ in the present case. It follows that steady state is approached more rapidly for increasing radiation densities, which is to imply that absorption and stimulated emission are the dominant processes. This is expected to be true for large radiation and/or particle densities, but the possibility remains that, even though $F_1R(0)\gg N_A$, there may exist a regime in which the absolute values of $F_1R(0)$ and N_A are not sufficiently large (for a given line width $\Delta\nu$) to strictly allow the dominance of absorption and stimulated emission over the spontaneous emission process. Indeed it will be found (in Sec. IV) that sufficiently rare systems will not approach the Planckian behavior at θ_R as defined above. These extensions are of both practical and theoretical interest, as indicated in earlier remarks, and a more detailed view of some aspects of these departures consti-

tutes the remainder of this paper.

IV. A MORE GENERAL CONSIDERATION OF COLLISIONLESS SYSTEMS

A. Higher-Order Terms Neglected

It is profitable to continue the neglect of all terms of higher statistical order for the present, inasmuch as it can be shown (see Part B of this section) that mean densities are not appreciably affected by the inclusion, or the neglect of, higher-order terms in the simple two-level models of present interest. This is not to suggest that the higher-order terms are, in fact, small. For in many instances they are not small. Having already viewed a collisionless system that approaches Planckian behavior at the initial radiation temperature, let us consider a conceptually useful counterexample. This example is one in which all atoms are initially in the upper level, and there is no radiation at time zero. Equation (47) reduces to

$$F_1^R(t) = pq[1 - e^{a(q-p)t}]/[p - qe^{a(q-p)t}], \quad (51)$$

and, as before, we have[10]

$$\lim_{t\to\infty} F_1^R(t) \to q \text{ for } p \neq 0. \quad (52)$$

The initial conditions lead to the term c_2 taking the value zero, and Eq. (52) is readily evaluated for two extreme cases. Case I is dominated by spontaneous emission and may be designated as having rare system behavior. Case II is dominated by absorption and stimulated emission and may be designated as having non-rare system behavior.

Case I

Rare system behavior is characterized by the condition that $N_A \ll \rho_{\nu_{ul}}\Delta\nu$. It follows that $b \simeq -M(\nu_{ul})\rho_{\nu_{ul}}/g_u$ (from Eq. (42), and that

$$\lim_{t\to\infty} F_1^R(t) \to N_A, \quad (53a)$$

$$\lim_{t\to\infty} F_1^{Au}(2P,t) \to 0, \quad (53b)$$

$$\lim_{t\to\infty} F_1^{Al}(1S,t) \to N_A. \quad (53c)$$

The essence of this result is evident both intuitively and analytically. Indeed it is to be expected that, in a very rare system, the probability for a photon to be "seen" by an atom in the ground level and to undergo an absorption event is very small. Thus atoms in the upper state simply decay away (by predominantly spontaneous emission) with only a minute fraction ever getting restored to the upper state by photon absorption. For this reason, rare system behavior has been loosely characterized as a case in which spontaneous emission is the dominant process.

Case II

Non-rare system behavior is characterized by the condition that $N_A \gg \rho_{\nu_{ul}}\Delta\nu$. In this case[11]

$$b \simeq M(\nu_{ul})N_A/g_u\Delta\nu,$$

and it follows that

$$\lim_{t\to\infty} F_1^R(t) \to N_A g_l/(g_u + g_l). \quad (54)$$

For Lyman-α transitions in hydrogen $g_u = 3$ and $g_l = 1$, thereby giving the following steady-state solutions:

$$\lim_{t\to\infty} F_1^R(t) \to N_A/4, \quad (55a)$$

$$\lim_{t\to\infty} F_1^{Au}(2P,t) \to 3N_A/4, \quad (55b)$$

$$\lim_{t\to\infty} F_1^{Al}(1S,t) \to N_A/4. \quad (55c)$$

In the familiar example of nondegenerate two-level oscillators (i.e., the upper level is nondegenerate as well as the lower level), we find that all densities approach the value of $N_A/2$.[12] As before, the origin of this result is clearly seen both intuitively and analytically – this type of system simply approaches a situation in which the rates of photon absorption and stimulated emission are sufficiently great that these two processes become dominant over spontaneous emission. Thus in non-rare system behavior the asymptotic level populations simply balance out in direct proportion to the number of available states associated with a given level.

While these latter two collisionless cases are exceedingly simple, they nevertheless point up the interesting fact that, in the absence of strong radiation fields, the radiation distribution obeys distinctly and basically different laws for opposing density extremes. Therefore in nonequilibrium systems, it may be profitable to *mechanistically* view departures from the simple Planckian behavior of Sec. III. Recall that the role of linewidths with respect to equilibration times may also be reduced to the same set of mechanistic considerations and will be further expanded upon forthwith.

To examine the temporal evolution in more detail for arbitrary system models, we will proceed by considering specific numerical examples. By performing a brief parameter study, major trends in dynamic characteristics can be noted. In the interest of brevity, only two basic sets of initial conditions will be considered in detail. In each instance we are considering Lyman-α transitions in hydrogen (such that $g_u = 3$ and $g_l = 1$). The examples may be broken down further as

Case A:

$$F_1^{Au}(2P, 0) = N_i$$

$$F_1^{Al}(1S, 0) = F_1^{R}(0) = 0$$

Case B:

$$F_1^{R}(0) = F_1^{Au}(2P, 0) = N_i$$

$$F_1^{Al}(1S, 0) = 1.65 N_i \ .^{13}$$

The quantity N_i is simply the number of atoms per cm³ in the upper atomic level at $t = 0$, and N_i takes values of 10^3 cm^{-3}, 10^{12} cm^{-3}, and 10^{16} cm^{-3} in each case. These densities correspond in a crude way to certain regions of interstellar space, to some laboratory plasmas, and to some regions of stellar interiors, respectively. The parametrization with respect to $\Delta\nu$ will allow $\Delta\nu$ to take values corresponding to the natural radiative linewidth (narrow), for an arbitrary intermediate linewidth, and for an arbitrary broad line. The parametrization with respect to linewidth merits some discussion, and it will be helpful to first tabulate (in Table I) the equilibration time, τ_{eq} (this is the time at which steady state is first reached). The radiation and atomic densities at steady state, denoted by $F_1^R(\infty)$, $F_1^{Au}(2P, \infty)$, and $F_1^{Al}(1S, \infty)$ are also tabulated in Table I for each case. When τ_{eq} happens to be less than $\tau_{ul} \equiv 1/\Delta\nu$, the entry in Table I will be labeled accordingly, and the value for the densities will be deleted – for these instances are recognized to violate constraints that are ordinarily associated with kinetic equations in conventional form. They will be discussed further below.

The most prominent feature of the data in Table I is that rare system behavior persists for all instances in which $N_i = 1.0 \times 10^3$ cm^{-3}. That is, in the absence of exceptionally strong radiation fields, it is expected that a major fraction of atoms will be in the ground level for a rare system at steady state. It can also be noted that τ_{eq} increases with $\Delta\nu$ in all of the rare systems that were observed. This is an indication that, for relatively large $\Delta\nu$ (and therefore relatively short lifetimes of upper states), spontaneous emission becomes more and more dominant, with restitutive net photon absorption events occurring less and less frequently. But to achieve steady state the rate of restoring atoms to the upper level must, of course, become equal to the rate at which they leave it. Therefore, as restituting events become relatively less likely (with increasing $\Delta\nu$), the upper level must become more and more depopulated (by spontaneous emission in this case) in order for all the rates to equalize. This additional depopulation simply requires more and more time, thereby accounting for increasing τ_{eq} with increasing $\Delta\nu$. In regard to the entries in Table I for $F_1^{Au}(2P, \infty) \sim 0$, it is realized that some vanishingly small value of $F_1^{Au}(2P, \infty)$ is, in fact, required to maintain steady state. Like-

TABLE I. Equilibrium times and asymptotic densities for Cases A and B with $N_i = 10^3$, 10^{12}, and 10^{16} cm^{-3}.

		Case A				Case B			
		τ_{eq}(sec)	$F_1^R(\infty)$	$F_1^{Au}(2P, \infty)$	$F_1^{Al}(1S, \infty)$	τ_{eq}(sec)	$F_1^R(\infty)$	$F_1^{Au}(2P, \infty)$	$F_1^{Al}(1S, \infty)$
$N_i = 1.0 \times 10^3$	cm^{-3}								
$\Delta\nu = 6.25 \times 10^8$	sec^{-1}	4.0×10^{-8}	1.0×10^3	~ 0	1.0×10^3	3.5×10^{-8}	2.0×10^3	~ 0	2.65×10^3
$\Delta\nu = 2.40 \times 10^{12}$		5.0×10^{-8}	1.0×10^3	~ 0	1.0×10^3	5.0×10^{-8}	2.0×10^3	~ 0	2.65×10^3
$\Delta\nu = 2.40 \times 10^{15}$		6.0×10^{-8}	1.0×10^3	~ 0	1.0×10^3	6.0×10^{-8}	2.0×10^3	~ 0	2.65×10^3
$N_i = 1.0 \times 10^{12}$	cm^{-3}								
$\Delta\nu = 6.25 \times 10^8$	sec^{-1}	$\tau_{eq} < \tau_{ul}$	. . .	. . .	. . .	$\tau_{eq} < \tau_{ul}$	. . .	. . .	. . .
$\Delta\nu = 2.40 \times 10^{12}$		1.1×10^{-8}	8.502×10^{11}	1.498×10^{11}	8.502×10^{11}	7.4×10^{-9}	1.416×10^{12}	5.836×10^{11}	2.066×10^{12}
$\Delta\nu = 2.40 \times 10^{15}$		3.0×10^{-8}	9.998×10^{11}	2.200×10^8	9.998×10^{11}	2.4×10^{-8}	1.999×10^{12}	1.165×10^9	2.649×10^{12}
$N_i = 1.0 \times 10^{16}$	cm^{-3}								
$\Delta\nu = 6.25 \times 10^8$	sec^{-1}	$\tau_{eq} < \tau_{ul}$	. . .	. . .	. . .	$\tau_{eq} < \tau_{ul}$	. . .	. . .	. . .
$\Delta\nu = 2.40 \times 10^{12}$		3.0×10^{-11}	2.510×10^{15}	7.490×10^{15}	2.510×10^{15}	1.3×10^{-10}	3.289×10^{14}	1.967×10^{16}	6.829×10^{15}
$\Delta\nu = 2.40 \times 10^{15}$		4.4×10^{-9}	5.401×10^{15}	4.599×10^{15}	5.401×10^{15}	2.7×10^{-9}	6.776×10^{15}	1.322×10^{16}	1.328×10^{16}

wise $F_1^{Al}(1S,\infty)$ and $F_1^R(\infty)$ are within this very small value of 1.0×10^3, even though in these cases the value of 1.0×10^3 has been entered in Table I.

As one goes to higher densities ($N_i = 1.0\times10^{12}$ cm^{-3} and 1.0^{16} cm^{-3}), it is first noticed that $\tau_{eq} < \tau_{ul}$ for the natural radiative width $\Delta\nu = 6.25\times10^8$ sec^{-1}. But it can be noted that this value of the width corresponds only to the spontaneous emission process.[9] In more dense systems it is clear that the stimulated emission process becomes important as well. Thus $\Delta\nu$ appearing in the kinetic equations must correspond to the *total* radiative width. The total radiative width is, of course, proportional to $[\rho_\nu + f_1^R(\nu,t)]$ (see Ref. 6);[14] and for this reason, even in collisionless systems, $\Delta\nu$ may take values of greater than 6.25×10^8 sec^{-1}. We have allowed ourselves the approximation that the system models herein can be simply parametrized by constant linewidths of greater or lesser values [as opposed to widths that are functionals of $F_1^R(t)$]. It is now interesting to note (from Table I) in Case A with $\Delta\nu = 2.4\times10^{12}$ sec^{-1} that, for $N_i = 1.0\times10^{12}$ cm^{-3}, rare system behavior obtains. However, for Case A with $\Delta\nu = 2.4\times10^{12}$ sec^{-1} and $N_i = 1.0\times10^{16}$ cm^{-3} non-rare system behavior obtains. When $\Delta\nu$ takes the value of 2.4×10^{15} sec^{-1}, it is seen that Case A for $N_i = 1.0\times10^{16}$ cm^{-3} tends to be somewhere between rare and non-rare system behavior. These examples simply point up the fact that, in density ranges that are of "laboratory interest," radiative coupling effects may, in general, become manifest in the form of one, or the other, or as a competition of the two characteristic trends that have been indicated herein. The same appears to be true for Case B with $N_i = 1.0\times10^{12}$ cm^{-3} and 1.0×10^{16} cm^{-3}.[15] Thus, even though one has gained a notion of the underlying factors that are at play, it still appears necessary in most nontrivial examples to simply solve each case of interest on its own merits. The time scales for equilibration of non-rare system behavior are found to be shorter than those for rare system behavior, as expected. A final point that can be made, but does not seem to merit extensive tabulations, relates to the effect of statistical weights. It can readily be seen [Eqs. (22)–(24) that, as the statistical weight for the upper level assumes smaller values, the transition rates for the associated emission processes become greater. In this case the dynamic and steady-state features may be expected to tend more toward the characteristics of rare system behavior. Explicit calculations on a hypothetical system of nondegenerate two-level oscillators ($g_u = g_l = 1$) for the cases considered in Table I bears this out upon comparison to the data in Table I and the subsequent graphical representations of the dynamic behavior for the above cases. In Figs. 1–6 the full temporal evolution for the cases of Table I are presented. Only the photon singlet density $F_1^R(t)$ is plotted versus time, since the atomic singlet densities $F_1^{Au}(2P,t)$ and $F_1^{Al}(1S,t)$ follow trivially from the conservation conditions [Eqs. (36)–(38)], the initial conditions, and the value of $F_1^R(t)$.[16]

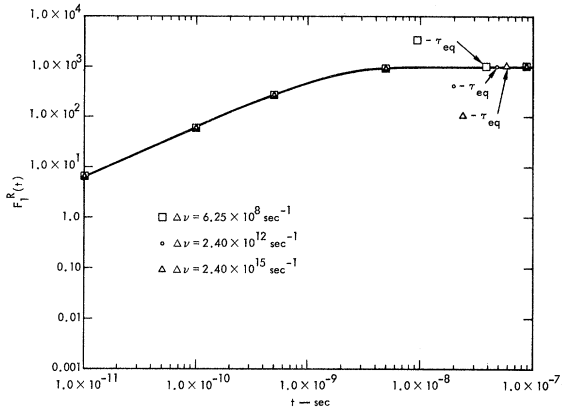

FIG. 1. The photon singlet density $F_1^R(t)$ versus time for $F_1^{Au}(2p,0) = 1.0\times10^3$ cm^{-3}, $F_1^{Al}(1S,0) = F_1^R(0) = 0$ (Case A with $N_i = 1.0\times10^3$ cm^{-3}).

B. Higher-Order Terms Included

By retaining all higher-order terms, one must now solve the set of nine coupled differential equations [Eqs. (22)–(31)]. In addition to the conservation conditions that are given by Eqs. (36)–(38), we have the conditions:

$$V^{AuAu}(2P,2P,t) + V^{AlAl}(1S,1S,t) + 2V^{AuAl}(2P,1S,t) = d_1, \quad (56)$$

$$V^{RR}(t) + V^{AlAl}(1S,1S,t) - 2V^{AlR}(1S,t) = d_2, \quad (57)$$

$$V^{RR}(t) + V^{AuAu}(2P,2P,t) + 2V^{AuR}(2P,t) = d_3, \quad (58)$$

$$V^{RR}(t) + V^{AuR}(2P,t) - V^{AlR}(1S,t) - V^{AuAl}(2P,1S,t) = d_4, \quad (59)$$

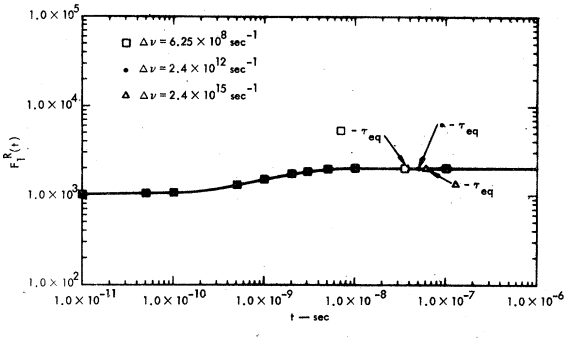

FIG. 2. The photon singlet density $F_1^R(t)$ versus time for $F_1^R(0) = F_1^{Au}(2P,0) = 1.0\times10^3$ cm^{-3}, $F_1^{Al}(1S,0) = 1.65\times10^3$ cm^{-3} (Case B with $N_i = 1.0\times10^3$ cm^{-3}).

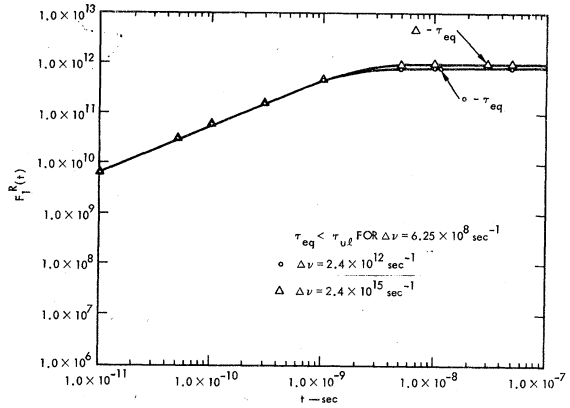

FIG. 3. The photon singlet density $F_1{}^R(t)$ versus time for $F_1{}^{Au}(2P, 0) = 1.0 \times 10^{12}$ cm^{-3}, $F_1{}^{Al}(1S, 0) = F_1{}^R(0) = 0$ (Case A with $N_i = 1.0 \times 10^{12}$ cm^{-3}).

$$V^{AuR}(2P, t) + V^{AlR}(1S, t) + V^{AuAu}(2P, 2P, t) + V^{AuAl}(2P, 1S, t) = d_5, \tag{60}$$

and

$$V^{AuR}(2P, t) + V^{AlR}(1S, t) - V^{AuAl}(2P, 1S, t) - V^{AlAl}(1S, 1S, t) = d_6. \tag{61}$$

In principle, the set of nine differential equations can be solved analytically, given a set of initial conditions. In practice, it is quite tedious to do so. On the other hand, a numerical solution of these nine equations (or even a much larger set of first-order differential equations) is exceedingly simple when standard numerical methods are applied. The results in this section have therefore been obtained numerically, using a fourth-order Runge-Kutta method.[17,18] As previously noted, it is found that the inclusion of variances and cross-covariances for photons and atoms does not alter the solutions obtained (to three significant figures) above for photon and atomic singlet densities in the absence

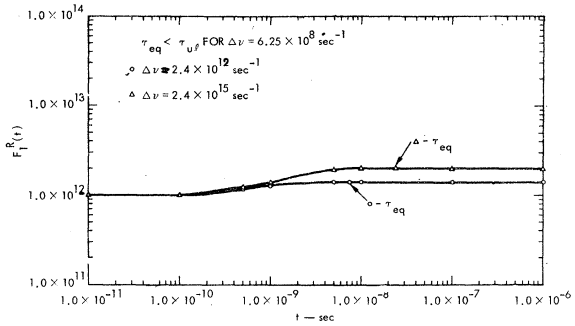

FIG. 4. The photon singlet density $F_1{}^R(t)$ versus time for $F_1{}^R(0) = F_1{}^{Au}(2P, 0) = 1.0 \times 10^{12}$ cm^{-3}, $F_1{}^{Al}(1S, 0) = 1.65 \times 10^{12}$ cm^{-3} (Case B with $N_i = 1.0 \times 10^{12}$ cm^{-3}).

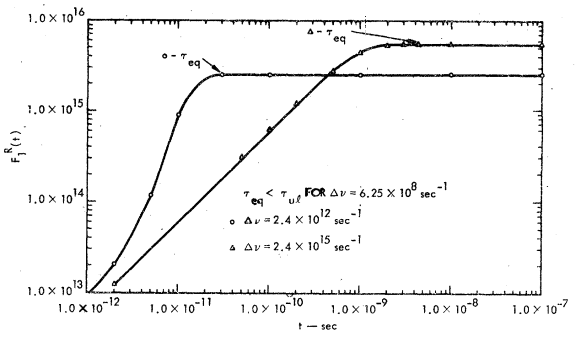

FIG. 5. The photon singlet density $F_1{}^R(t)$ versus time for $F_1{}^{Au}(2P, 0) = 1.0 \times 10^{16}$ cm^{-3}, $F_1{}^{Al}(1S, 0) = F_1{}^R(0) = 0$ (Case A with $N_i = 1.0 \times 10^{16}$ cm^{-3}).

of higher-order terms. It will also be seen that, for the system models that we are considering, the higher-order terms may be small, or large, depending upon initial conditions and other factors. This is only to say that, in the systems that we have investigated, the singlet (mean) densities are for practical purposes unaffected by the magnitude of the fluctuations.[19] In view of the constancy of singlet densities in the present models, the solutions for the higher-order terms under various sets of conditions are, nevertheless, of some interest. Perhaps primary interest arises because recent advances in quantum-optical measurement techniques have made the observation of fluctuations more or less commonplace. Secondly, there is theoretical interest stemming from a continuing suspicion that, particularly in less simple non-equilibrium systems (than those we have considered herein), the higher-order terms may yet account nontrivially for statistical coupling between the various distributions. If nothing more, the present work represents a first step toward systematically writing and explicitly solving a set of transport equations descriptive of a radiating system that are consistently coupled to second statistical order. In view of the examples already considered, in which variances and cross-covariances were neglected, let us consider here just two of the more

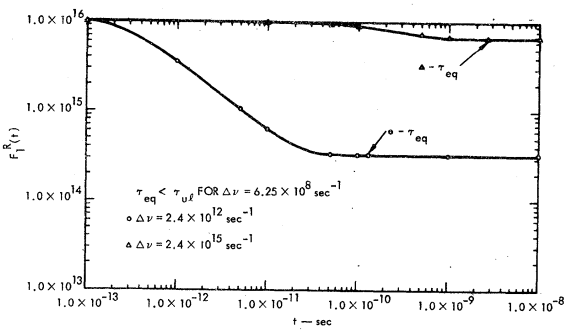

FIG. 6. The photon singlet density $F_1{}^R(t)$ versus time for $F_1{}^R(0) = F_1{}^{Au}(2P, 0) = 1.0 \times 10^{16}$ cm^{-3}, $F_1{}^{Al}(1S, 0) = 1.65 \times 10^{16}$ cm^{-3} (Case B with $N_i = 1.0 \times 10^{16}$ cm^{-3}).

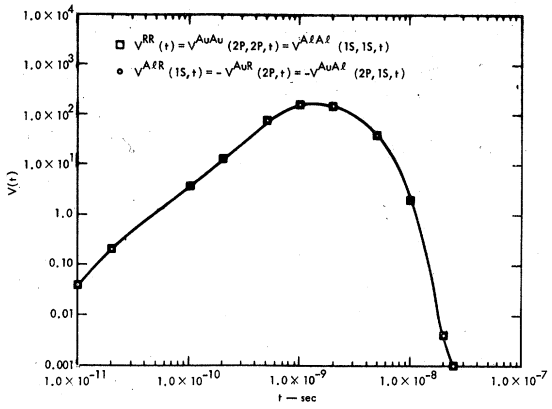

FIG. 7. Variances and cross-covariances versus time for $F_1^{Au}(2P,0) = 1.0 \times 10^3$ cm^{-3}, $F_1^{Al}(1S,0) = F_1^R(0) = 0$, $\Delta\nu = 6.25 \times 10^8$ sec^{-1}, all higher-order terms initially zero, and $1/\mathcal{V}_S = 1$.

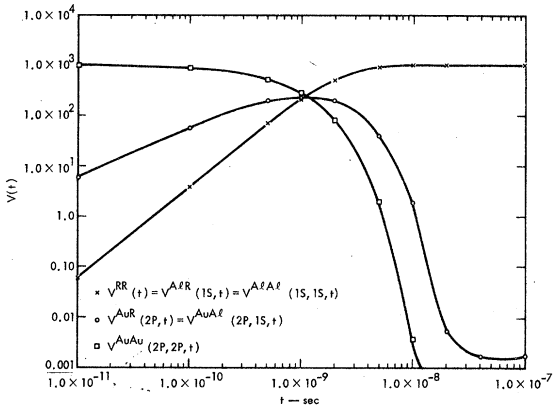

FIG. 9. Variances and cross-covariances versus time for $F_1^{Au}(2P,0) = 1.0 \times 10^3$ cm^{-3}, $F_1^{Al}(1S,0) = F_1^R(0) = 0$, $\Delta\nu = 6.25 \times 10^8$ sec^{-1}, all higher-order terms initially zero except $V^{AuAu}(2P,2P,0) = 1.0 \times 10^3$ cm^{-6}, and $1/\mathcal{V}_S = 0$.

basic system types that come to mind – that is, one which typifies rare behavior and one which typifies non-rare behavior. Each case corresponds to the Cases A in Table I [$F_1^{Au}(2P,0) = N_i$, $g_u = 3$, and $F_1^{Al}(1S,0) = F_1^R(0) = 0$]. For rare system behavior, we take $N_i = 1.0 \times 10^3$ cm^{-3} and $\Delta\nu = 6.25 \times 10^8$ sec^{-1}. For non-rare system behavior we take $N_i = 1.0 \times 10^{16}$ cm^{-3} and $\Delta\nu = 2.4 \times 10^{12}$ sec^{-1}. Recall that, for sufficiently large systems, terms proportional to $1/\mathcal{V}_S$ may be neglected. For the opposite extreme, we arbitrarily take $1/\mathcal{V}_S$ to be unity. All that remains to be specified in order to solve the full set of equations are the initial conditions on the variances and the cross-covariances.

One obvious case can be readily dispatched. That is the case in which all variances and cross-covariances are initially zero and $1/\mathcal{V}_S = 0$. It is found that the variances and cross-covariances remain zero for all time (recall that singlet densities in all cases are the same as in Figs. 1–6). However, when this example is solved with $1/\mathcal{V}_S = 1$, the variances and cross covariances do not remain zero, which indicates that the "self-correlation" terms are introducing fluctuations and coupling between distributions as the system evolves. This behavior is shown graphically in Figs. 7 and 8 for rare and non-rare behavior, respectively.

By comparing the above data, it is clear that the variances and cross-covariances grow to relatively large values in the early history of rare system behavior (for this set of initial conditions) and then diminish back to very small values asymptotically. The non-rare system behavior appears to evolve in more or less the inverse manner, thereby ending up with appreciable fluctuations and coupling within, and between, the distributions at steady state. It was also found in these examples that, for rare system behavior, the equilibration times for the variances and cross-covariances are equal to τ_{eq} for the singlet densities (and therefore, to τ_{eq} for the corresponding examples in Table I). In the case of non-rare behavior it took somewhat longer (as much as twice τ_{eq} for the singlet densities) before all of the variances and cross-covariances finally reached steady state. These higher-order terms were, however, quite

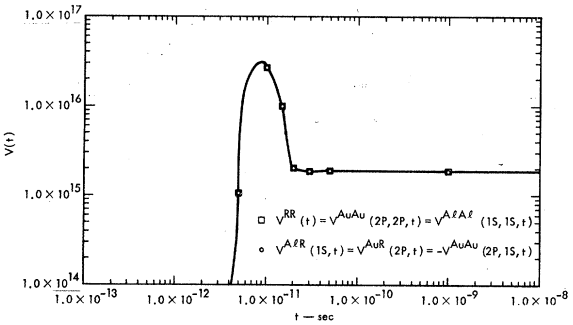

FIG. 8. Variances and cross-covariances versus time for $F_1^{Au}(2P,0) = 1.0 \times 10^{16}$ cm^{-3}, $F_1^{Al}(1S,0) = F_1^R(0) = 0$, $\Delta\nu = 2.4 \times 10^{12}$ sec^{-1}, all higher-order terms initially zero, and $1/\mathcal{V}_S = 1$.

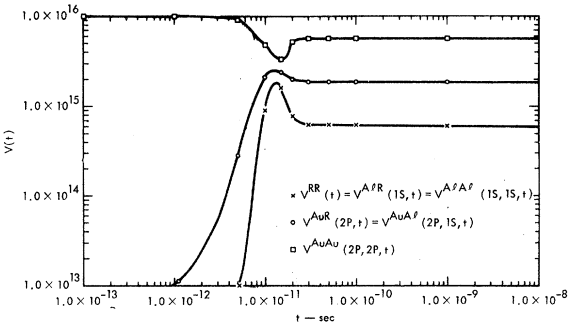

FIG. 10. Variances and cross-covariances versus time for $F_1^{Au}(2P,0) = 1.0 \times 10^{16}$ cm^{-3}, $F_1^{Al}(1S,0) = F_1^R(0) = 0$, $\Delta\nu = 2.4 \times 10^{12}$ sec^{-1}, all higher-order terms initially zero except $V^{AuAu}(2P,2P,0) = 1.0 \times 10^{16}$ cm^{-6}, and $1/\mathcal{V}_S = 0$.

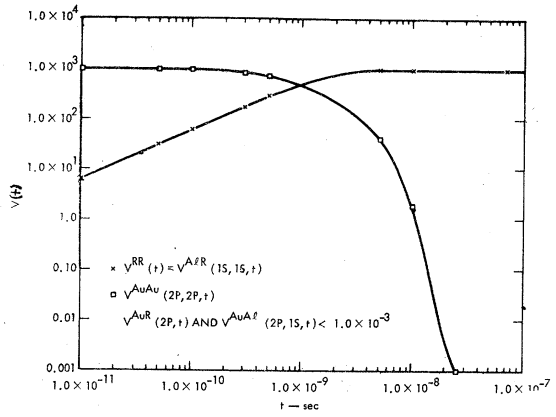

FIG. 11. Variances and cross-covariances versus time for $F_1^{Au}(2P, 0) = 1.0 \times 10^3$ cm^{-3}, $F_1^{Al}(1S, 0) = F_1^R(0) = 0, \Delta\nu = 6.25 \times 10^8$ sec^{-1}, all higher-order terms initially zero except $V^{AuAu}(2P, 2P, 0) = 1.0 \times 10^3$ cm^{-6}, and $1/\mathcal{V}_S = 1$.

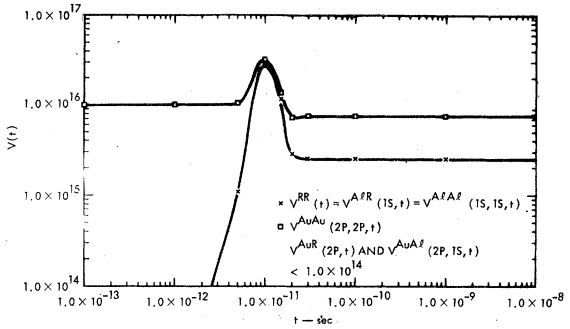

FIG. 12. Variances and cross-covariances versus time for $F_1^{Au}(2P, 0) = 1.0 \times 10^{16}$ cm^{-3}, $F_1^{Al}(1S, 0) = F_1^R(0) = 0, \Delta\nu = 2.4 \times 10^{12}$ sec^{-1}, all higher-order terms initially zero except $V^{AuAu}(2P, 2P, 0) = 1.0 \times 10^{16}$ cm^{-6}, and $1/\mathcal{V}_S = 1$.

near (within a few percent) their steady-state values at τ_{eq} for the singlet densities. It can also be pointed out that, in these particular examples (see Figs. 7 and 8), the signs of the cross-covariances clearly point up the nature of the coupling between various distributions. For example, the negative sign of $V^{AuR}(2P, t)$ indicates that the creation of a photon correspondingly annihilates an atom in the upper state (and vice versa, the annihilation of a photon accounts for the creation of an atom in the upper state). An analogous statement obtains for the negative sign of $V^{AuAl}(2P, 1S, t)$. On the other hand, positive $V^{AlR}(1S, t)$ indicates positive coupling between photons and atoms in the lower state, inasmuch as the creation of a photon correspondingly accounts for the creation of an atom in the lower state, and *vice versa*.

A final set of examples is identical to those just considered (Figs. 7 and 8), with the single exception that $V^{AuAu}(2P, 2P, 0)$ is now set equal to $F_1^{Au}(2P, 0) = N_i$. The results in graphical form are displayed in Figs. 9–12. Comparison of the data in Figs. 9–12 to that in Figs. 7 and 8 is more or less self-evident. In the rare systems (Figs. 9 and 11) the initial fluctuation in the upper-level (atomic) distribution diminishes monotonically with decay from that state; and in effect, this initial fluctuation is transmitted to the photon and lower-level atomic distributions. In the non-rare case the fluctuations at steady state are once again appreciable and are significantly larger for $1/\mathcal{V}_S = 1$ than for $1/\mathcal{V}_S = 0$. The times necessary to reach a steady state follow the same pattern in this set of examples as when the initial fluctuations were taken to be zero. It is apparent that no attempt is to be made at this point to suggest general rules and/or mechanistic trends of the higher-order terms. The main intent in this latter segment has been simply to give a preliminary indication of some of the ways that the higher-order quantities *may* behave, dynamically and asymptotically, in the absence of the usual constraints associated with thermodynamic equilibrium.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge Ronald Ott for considerable assistance at numerous stages of this investigation.

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[1]This includes cross-doublet densities between different atomic states as well as between atomic states and photons – as opposed to, and in addition to, self-doublet densities for atomic states and for photons.

[2]R. H. Fowler, Statistical Mechanics (Cambridge University Press, London, 1936), 2nd ed.

[3]R. J. Gelinas and R. K. Osborn, Phys. Rev. 163, 162 (1967).

[4]R. J. Gelinas and R. K. Osborn, in Proceedings of the A. E. C. Symposium Series No. 9 on Neutron Noise, Wave, and Pulse Propagation, University of Florida, 1966 (U. S. A. E. C. Division of Technical Information Extension, Oak Ridge, Tennessee 1967).

[5]R. K. Osborn, Phys. Rev. 130, 2142 (1963).

[6]W. Heitler, The Quantum Theory of Radiation (Oxford University Press, London, 1954), 3rd ed.

[7]A. Z. Akcasu, The University of Michigan, Office of Research Administration Technical Report No. 04836 – 1 – T, 1963 (unpublished), under ORA Project 04836.

[8]In many applications the specific light intensity $I(\nu, t)$ is employed. $I(\nu, t)$ is defined as $I(\nu, t) \equiv h\nu f_1^R(\nu, t)$, where h is Planck's constant and c is the speed of light.

[9]H. Bethe and E. E. Salpeter, Quantum Mechanics of One- and Two-Electron Atoms (Springer-Verlag, Berlin, 1957).

[10]A case in which $p = 0$ is the trivial one in which, ini-

tially, all atoms are in the ground level and no radiation is present. It may then be easily shown that $F_1^R(t)$ remains zero for all time. This is expected, because in this model there is no way to get atoms out of the ground state and to create photons. Therefore nothing happens.

[11]In both Case I and Case II, it can be noted that $4ac \ll b^2$, thereby allowing the expansion $(b^2-4ac)^{1/2} \simeq |b|(1-2ac/b^2)$.

[12]J. Premanand and D. L. Falkoff, Phys. Rev. 163, 178 (1967).

[13]The factor 1.65 has arbitrary origin. It is the value taken by the Boltzmann factor, $(g_l/g_u)\exp(h\nu_{ul}/\theta)$ for $\theta = 2h\nu_{ul}$ in a hypothetical nondegenerate two-level system ($g_u = g_l = 1$). It is perhaps best to view 1.65 as nothing more than an arbitrary factor, inasmuch as the notion of a temperature is as often an encumbrance as an aid when the system is not at or near equilibrium.

[14]Recall that $f_1^R(\nu, t)$ is defined in the continuum. See Eq. (11) *et seq.*

[15]Cases B were included primarily because they represent a set of initial conditions that are more likely to exist, physically, than the conceptually useful Cases A (Cases A correspond to the relaxation of an initial distribution of atoms with an extreme measure of population inversion). Cases B may be viewed to correspond, at least crudely, to a system of atoms (initially distributed in the ratio of 1.65) that is instantaneously irradiated by an arbitrary light source at $t = 0$.

[16]It may finally be noted that, for any of the examples in Table I and/or Figs. 1–6, the asymptotic radiation density can be used, after the fact, to define a Planck temperature that also turns out to satisfy Boltzmann's distribution law for the corresponding asymptotic atomic level populations. This calculated asymptotic temperature is generally found to be different from any of the initial temperatures (the initial temperatures being determined from the initial atomic level populations and the initial radiation densities). At any rate, temperature is simply a defined quantity here and is in no way essential to the basic formulation and solution of our kinetic equations. However, if one were to persist with a wish to keep track of time-dependent temperatures for the various components of a given system, it would still be necessary to first solve for the distribution functions, *per se*. Then, with this knowledge and a set of appropriate (and reasonably well-agreed-upon) definitions, the corresponding evolution of temperatures could be computed. It has, of course, been demonstrated in this section that such an evolution is very much dependent upon system parameters such as linewidths, statistical weights, and particle and radiation densities. With the availability of large, high-speed computers, the approach that we are presenting is feasible for much more complicated physical systems as well.

[17]T. Suyehiro, Lawrence Radiation Laboratory, Livermore, California, C.I.C. Report No. D2.3-001, 1965 (unpublished).

[18]The results of the above cases, in which higher-order terms were neglected (thereby reducing to a set of three simultaneous differential equations), have also been verified by this numerical method.

[19]This point may also have been noticed by Premanand and Falkoff.[12] For they noted that the fluctuations may be non-negligible, and that the higher-order terms should, in general, be included in conventional detailed-balance equations for systems not at equilibrium. They did not, however, indicate that the mean values (singlet densities) *must*, in general, change noticeably by the inclusion of these additional terms (although it is suspect). Indeed, the present work explicitly shows that, not only asymptotically but at all times, the singlet densities of photons and atoms are essentially unaffected by the inclusion of, or the neglect of, the higher-order terms in the simple system models under present consideration.

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Coupling of Density and Spin Fluctuations to Quasiparticles in a Fermi Liquid*

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The general interaction between quasiparticles and particle-hole excitations in a neutral Fermi system at $T = 0$ is investigated. Density and spin-density fluctuations and collective modes are treated on an equal basis. Use is made of Ward identities to relate the vertex function Γ to the quasiparticle self-energy Σ, and logarithmic corrections to the self-energy are obtained. The coefficients of the logarithmic terms are calculated analytically in terms of known Landau parameters.

I. INTRODUCTION

The microscopic theory of a normal Fermi liquid,[1-4] formulated in terms of Green's functions, rests on certain assumptions about the regularity of the mass operator, Σ (single-particle self-energy). Specifically, it is assumed that the real part of the mass operator, $\mathrm{Re}\Sigma \equiv M$, can be expanded in a power series near the Fermi surface, i.e., when $|\vec{q}| = q \to k_F$, $\epsilon \to 0$: