(in first order): $\alpha_0^T = 0.76035$; $\alpha_1^T = 0.02480$.

Total values of the constant through first order, $\tilde{\alpha}^T = \alpha_0^T + Z^{-1} \alpha_1^T$, for various Z values, are listed in Table III.

IV. DISCUSSION

The constant α^T as calculated by applying criterion B exhibits Z dependence for these ions. The Z^{-1} perturbation expansion of α appears to be rapidly convergent. In applying criterion A, one finds that the total energy through first order is independent of α , and that the second-order energy involves α in zero order. The values α_0^T and $\tilde{\alpha}^T$, found by forcing the total energy $E_{\rm SA}$ to be exactly the HF total energy, are different from, but quite close to, the value α_0^m found by minimizing the energy in

*Research supported by grants from the National Research Council of Canada and the Ontario Department of University Affairs.

¹J. C. Slater, Phys. Rev. <u>81</u>, 385 (1951).

²R. Gaspar, Acta Phys. Hung. <u>3</u>, 263 (1954).

 $^3W.$ Kohn and L. Sham, Phys. Rev. <u>140</u>, A1133 (1966). 4We employ atomic units in this section.

⁵F. Herman and S. Skillman, Atomic Structure Calcu-

lations (Prentice-Hall, Englewood Cliffs, N. J., 1963).

⁶B. Y. Tong and D. J. Sham, Phys. Rev. <u>144</u>, 1 (1966). ⁷D. J. McNaughton and V. H. Smith, Jr., Intern. J.

Quantum Chem. <u>3S</u>, 775 (1970).

 8 M. Berrondo and O. Goscinski, Phys. Rev. <u>184</u>, 10 (1969).

⁹Excellent reviews and extensive bibliographies of the local exchange problem may be found in (a) J. C. Slater, MIT Solid-State and Molecular Theory Group Report No. 71, 1969 (unpublished); (b) J. C. Slater and J. H. Wood, Intern. J. Quantum Chem. <u>4S</u>, 3 (1971).

¹⁰I. Lindgren, Arkiv Fysik <u>31</u>, 59 (1966).

¹¹J. C. Slater, T. M. Wilson, and J. H. Wood, Phys. Rev. <u>179</u>, 28 (1969).

¹²D. J. Stukel, R. N. Euwema, T. C. Collins, and V. H. Smith, Jr., Phys. Rev. B 1, 779 (1970).

criterion A. Minimum energy thus found is higher than, but quite close to, the HF energy in second order. The zero-order values α_0^T and α_0^m are in the neighborhood of the corresponding values for the three-electron ions, ²⁵ but show a slight decrease with increasing number of electrons. The firstorder corrections α_1^T increase rather rapidly with increasing number of electrons. However, the total value $\tilde{\alpha}^T$ shows a decrease with increasing number of electrons.

ACKNOWLEDGMENT

One of us (V. H. S.) wishes to thank Professor J. C. Slater for several discussions on local exchange and for reports of Refs. 9 and 19 prior to publication.

¹³T. M. Wilson, J. H. Wood, and J. C. Slater, Phys. Rev. A 2, 620 (1970).

¹⁴E. A. Kmetko, Phys. Rev. A <u>1</u>, 37 (1970).

¹⁵See, e.g., P. O. Löwdin, J. Mol. Spectry. <u>3</u>, 46 (1959).

¹⁶K. Schwarz (unpublished).

¹⁷W. F. Rudge, Phys. Rev. <u>181</u>, 1033 (1969).

¹⁸M. Ross, Phys. Rev. <u>179</u>, 612 (1969).

¹⁹J. C. Slater and J. H. Wood (unpublished).

²⁰L. J. Sham, Phys. Rev. A <u>1</u>, 969 (1970).

²¹J. C. Slater (unpublished).

²²It is a common misconception that the local exchange approximations of Slater and Gaspar-Kohn-Sham neglect the self-exchange term because of the large number of electrons considered in the statistical approximation. See Refs. 1 and 2 and J. C. Slater, *Quantum Theory of Atomic Structure* (McGraw-Hill, New York, 1960), Vol. II, Chap. 17; *Quantum Theory of Matter* (McGraw-Hill, New York, 1968), Chap. 17.

²³J. Linderberg, Phys. Rev. <u>121</u>, 816 (1961).

 $^{24} \rm Numerical$ integration was required in some parts of the evaluation of these coefficients, due to the presence of $\tau_{0}.$

²⁵S. R. Singh and V. H. Smith, Jr. (unpublished).

PHYSICAL REVIEW A

VOLUME 4, NUMBER 5

NOVEMBER 1971

Rotating-Wave Approximation and Spontaneous Emission*

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Spontaneous emission from two systems, namely, N identical harmonic oscillators and N identical two-level atoms, is studied without the use of the rotating-wave approximation. Certain new features of spontaneous emission, for instance, the dependence of the radiation rate on the initial dipole moment phase, are discussed.

It is well known¹ that the Hamiltonian characterizing the interaction between a quantized radiation field and identical two-level atoms has the form

$$H = \omega_0 \sum_j S_j^x + \sum_{kS} \omega_{kS} a_{kS}^{\mathsf{T}} a_{kS} + \sum_{kSj} \{g_{kS} a_{kS}(S_j^{\mathsf{T}} + S_j^{\mathsf{T}}) + \text{H.c.}\},$$
(1)

where

$$g_{kS} = (-i\omega_0/c) \ (2\pi c/L^3)^{1/2} \ k^{-1/2} \ (\bar{\mathbf{d}})_{12} \cdot \bar{\boldsymbol{\epsilon}}_{kS} \ . \tag{2}$$

Here $(\tilde{d})_{12}$ is the dipole matrix element and S_i^* are the spin angular momentum operators. All other symbols have the usual meanings. In quantum optics^{1,2} one invariably makes the "rotating-wave approximation" (RWA), which amounts to ignoring the rapidly oscillating terms. In this approximation the Hamiltonian for the interaction between the radiation field and two-level atoms has the form

$$H = \omega_0 \sum_j S_j^z + \sum_{kS} \omega_{kS} a_{kS}^{\dagger} a_{kS} + \sum_{kSj} \left\{ g_{kS} a_{kS} S_j^{\dagger} + \text{H.c.} \right\}.$$
(3)

Following the classic work of Bloch and Siegert³ (in which the interaction of two-level systems with external *c*-number fields⁴ was considered), it has *a priori* been assumed that RWA is a good approximation, provided that the interaction between the radiation field and matter is weak. However, to our knowledge no calculation has been carried out for the problem of interaction between radiation and matter which explicitly justifies the above assertion.⁵ The present investigation is concerned with this question.

We consider spontaneous emission⁶⁻⁸ from two systems, namely, N identical two-level atoms and N identical harmonic oscillators. We obtain, without the use of the RWA, the master equation which describes the spontaneous emission. We calculate the initial radiation rate I(0) from these two systems under various conditions of initial excitation and it is found that I(0), in contrast to the radiation rate $I^{(r)}$ (0) obtained by making the RWA, depends strongly on the phase of the dipole moment at t = 0. In particular, if the system is initially excited to a state with zero dipole moment, for instance, the state $|\frac{1}{2}N, M\rangle$ introduced by Dicke, ⁶ then I(0) is identical with $I^{(r)}(0)$. We also show explicitly for the case of harmonic oscillators that the radiation rate I(t) for later times reduces to $I^{(r)}(t)$ in the weak-coupling limit and if averaging operation over a few optical cycles is carried out.

We first examine a system of harmonic oscillators which are emitting spontaneously. We have studied spontaneous emission from this system in the RWA in detail in II. Our treatment was based on the master equation for the reduced phase-space distribution function characterizing the oscillator system alone.⁹ When we do not make the RWA, we find that the reduced phase-space distribution function satisfies the following master equation¹⁰:

$$\frac{\partial F^{(A)}}{\partial t} = i\omega_0 \sum_{\mathbf{z}} \left(z_i \frac{\partial}{\partial z_i} - z_i^* \frac{\partial}{\partial z_i^*} \right) F^{(A)} + \gamma_0 \sum_{ij} \left\{ \frac{\partial}{\partial z_i} \left[(z_j - z_j^*) F^{(A)} \right] + \mathrm{c.c.} \right\}.$$
(4)

The notation is the same as that of II. Here $F^{(A)}$ corresponds to the reduced density operator in the Schrödinger picture. We recall that in our¹¹ terminology, $(1/\pi)F^{(A)}$ is the same as the Sudarshan-Glauber phase-space distribution function.¹² Equation (4) is the desired Fokker-Planck equation which can be used to calculate all the properties of the spontaneously emitted radiation. It is clear that the Langevin equations¹³ equivalent to (4) are

$$\dot{z}_i = -i\omega_0 z_i - \gamma_0 \sum_j (z_j - z_j^*).$$
(5)

It should be noted that Eqs. (5) do not contain any fluctuating forces. We can also write down the operator form of the master equation by using the general theory developed in Ref. 11. It is found by straightforward but long calculations that the reduced density operator ρ satisfies the following equation^{14, 15}:

$$\frac{\partial \rho}{\partial t} = -i\omega_0 \sum_j \left[a_j^{\dagger} a_j, \rho \right] - \gamma_0 \sum_{ij} \left\{ a_i^{\dagger} a_j \rho - 2a_j \rho a_i^{\dagger} \right. \\ \left. + \rho a_i^{\dagger} a_j + a_i a_j \rho - a_i \rho a_j - a_i^{\dagger} \rho a_i^{\dagger} + \rho a_i^{\dagger} a_i^{\dagger} \right\}.$$
(6)

We first calculate the radiation rate I(t), which is given by

$$I(t) = -\omega_0 \frac{d}{dt} \sum_j \langle a_j^{\dagger}(t) a_j(t) \rangle .$$
⁽⁷⁾

On using (6) and (7) we find that

$$I(t) = 2\omega_{0}\gamma_{0} \left[\langle D^{\dagger}(t) D(t) \rangle - \frac{1}{2} \langle D^{\dagger}(t) D^{\dagger}(t) \rangle - \frac{1}{2} \langle D(t) D(t) \rangle \right],$$
(8)

where the operator D(t), which determines the properties of the emitted radiation, is given by

$$D(t) = \sum_{j} a_{j}(t). \tag{9}$$

The mean values appearing in (8) are, of course, to be obtained from the solution of the master equation. We now present the solution of (4). It is easily shown that the Green's function $K^{(A)}(\{z_i\}, \{z_i^*\}, t | \{z_i^0\}, \{z_i^{0*}\}, 0)$ associated with the Fokker-Planck equation (4) is

$$K^{(A)}(\{z_i\}, \{z_i^*\}, t | \{z_i^0\}, \{z_i^{0*}\}, 0) = \prod_i \delta^{(2)}(z_i - \overline{z}_i(t)),$$
(10)

where $\overline{z}_i(t)$ is obtained from the solution of (5) subject to the initial condition $z_i(0) = z_i^0$. The time dependence of the phase-space distribution function is then given by

$$F^{(A)}(\{z_i\}, \{z_i^*\}, t) = \int d^2 \{z_i^0\} K^{(A)}$$
$$\times (\{z_i\}, \{z_i^*\}, t | \{z_i^0\}, \{z_i^{0*}\}, 0\} F^{(A)}(\{z_i^0\}, \{z_i^{0*}\}, 0), \quad (11)$$

where $F^{(A)}(\{z_i\}, \{z_i^*\}, 0)$ is the distribution function at time t = 0.

We first consider the case when each of the oscillators at t=0 was excited to a coherent state $|z_0\rangle$ (which is a state with finite dipole moment), i.e.,

$$F^{(A)}(\{z_i^0\}, \{z_i^0\}, 0) = \prod_i \{\pi \delta^{(2)}(z_i^0 - z_0)\}.$$
(12)

On combining (10)-(12) we find that

$$F^{(A)}(\{z_i\},\{z_i^*\},t) = \prod_i \{\pi \delta^{(2)}(z_i - z(t))\}.$$
(13)

We thus find that the oscillator system remains in a coherent state whose amplitude is given by

$$z(t) = [\cos\omega t - (i\omega_0/\omega)\sin\omega t] z_0 e^{-N\gamma_0 t} + (N\gamma_0/\omega)\sin\omega t z_0^* e^{-N\gamma_0 t}, \quad (14)$$

where

$$\omega = (\omega_0^2 - \gamma_0^2 N^2)^{1/2} . \tag{15}$$

On using (13), we find that the radiation is given by

$$I(t) = 4\gamma_0 \omega_0 N^2 |z(t)|^2 \sin^2 \varphi(t), \quad \varphi(t) = \operatorname{Im} \ln z(t) .$$
(16)

The corresponding result in the rotating-wave approximation is given by

$$I^{(r)}(t) = 2\gamma_0 \omega_0 N^2 |z_0|^2 e^{-2N\gamma_0 t} .$$
⁽¹⁷⁾

On comparing (16) and (17), it is clear that I(0) may be very different from $I^{(r)}(0)$ depending on the phase of the initial coherent amplitude. On the other hand, if the system is excited initially to a state with zero dipole moment [e.g., a Fock state or the state characterized by $F^{(A)} = \prod_i \{\delta(|z_i| - |z_0|)/2|z_0|\}$], then the last two terms on the right-hand side of (8) do not contribute and I(0) becomes identical with $I^{(r)}(0)$.

We now study the limiting form of the radiation rate (16) in the weak-coupling limit $N\gamma_0 \ll \omega_0$. In this limit (16) reduces to

$$I(t) - 2I^{(r)}(t)\sin^2(\varphi_0 + \omega t), \quad \varphi_0 = \text{Im} \ln z_0.$$
 (18)

It is clear that I(t) is the same as $I^{(r)}(t)$, provided that the rapidly oscillating terms are ignored. One may also argue that such a rapid oscillation cannot be observed by means of available photodetectors and in practice an averaging operation over a few optical periods is always performed. On the other hand, the correlation function of the form

$$\langle a_j^{\dagger}(t)a_k(0)\rangle = [\cos\omega t + (i\omega_0/\omega)\sin\omega t] |z_0|^2 e^{-Nr_0 t} + (N\gamma_0/\omega)\sin\omega t z_0^2 e^{-Nr_0 t}$$
(19)

shows the oscillation at the optical frequency. For the problem of the interaction of radiation and matter, our analysis indicates the sense in which the rotating-wave approximation is to be interpreted. Similar results hold if the system is initially excited to a state with zero dipole moment. For example, for the initial Fock-state excitation and for N=1, it is found that

$$\frac{I(t) - I^{(r)}(t)}{I^{(r)}(t)} = \frac{2\gamma_0}{\omega^2} \left\{ \gamma_0 \sin \omega t - \omega \cos \omega t \right\} \sin \omega t \ . \ (20)$$

It is again clear from (20) that in the weak-coupling

limit I(t) is equal to $I^{(r)}(t)$.

We will now briefly examine spontaneous emission from a system of N identical two-level atoms. The master equation in the RWA was obtained in I. On carrying out calculations similar to those given in I, we find that the master equation (IA7) is modified to

$$\frac{\partial \rho}{\partial t} = -i\omega_0 \sum_j [S_j^s, \rho] - \gamma_0 \sum_{ij} \{S_i^* S_j^- \rho - 2S_j^- \rho S_i^* + \rho S_i^* S_j^- + S_i^- S_j^- \rho S_j^- - S_i^- \rho S_j^- - S_i^- \rho S_j^+ + \rho S_i^* S_j^+ \} .$$
(21)

On using (21) one can show that the radiation rate I(t) is given by

$$I(t) = -\omega_0 \frac{d}{dt} \sum_j \langle S_j^{\mathbf{e}} \rangle$$

= $2\omega_0 \gamma_0 \{ \langle S^*(t) S^-(t) \rangle - \frac{1}{2} \langle S^*(t) S^*(t) \rangle$
 $- \frac{1}{2} \langle S^-(t) S^-(t) \rangle \}, \qquad (22)$

where we have introduced the collective operators⁶ S^* defined by

$$S^{\star}(t) = \sum_{j} S_{j}^{\star}(t) \quad . \tag{23}$$

We will compute the radiation rate at t = 0 for two types of initial excitation: (a) If the system is excited to a Dicke state⁶ $|\frac{1}{2}N, M\rangle$ (which is a state with zero dipole moment), then

$$I(0) = 2\omega_0 \gamma_0 (\frac{1}{2}N - M + 1) (\frac{1}{2}N + M) .$$
 (24)

This result is identical to the one obtained by making the RWA. (b) If the system is excited to a state given by

$$\left|\psi\right\rangle = \prod_{i} \left\{ \sin\frac{1}{2}\theta \, e^{-i\varphi/2} \left|+\right\rangle_{i} + \cos\frac{1}{2}\theta \, e^{i\varphi/2} \left|-\right\rangle_{i} \right\},\tag{25}$$

then the initial radiation rate is given by

$$I(0) = 2\omega_0 \gamma_0 N \{ \cos^2(\frac{1}{2}\theta) + \frac{1}{2}(N-1)\sin^2\theta \sin^2\varphi \} .$$
(26)

Equation (26) shows the strong dependence of I(0) on the dipole moment phase φ . The corresponding result in the RWA is

$$I^{(r)}(0) = 2\omega_0 \gamma_0 N \left\{ \cos^2(\frac{1}{2}\theta) + \frac{1}{4}(N-1)\sin^2\theta \right\}.$$
 (27)

For $\theta = \frac{1}{2}\pi$, we find that

$$I(\mathbf{0}) = \omega_0 \gamma_0 N \left\{ \cos^2 \varphi + N \sin^2 \varphi \right\}, \ I^{(\tau)}(\mathbf{0}) = \frac{1}{2} \omega_0 \gamma_0 N (N+1) ,$$
(28)

which shows that for $\theta = \frac{1}{2}\pi$, the emission, in contrast to the result obtained by making the RWA, is *not* necessarily superradiant. The radiation rate I(t) from *one* two-level atom is the same as $I^{(r)}(t)$ because $(S^{\pm})^2 = 0$. However, the time dependence of the dipole moment is different from the one obtained by making the RWA:

$$\langle S^{-}(t) \rangle = [\cos \omega t - (i\omega_{0}/\omega) \sin \omega t] \langle S^{-}(0) \rangle e^{-\gamma_{0}t}$$

+
$$(\gamma_0/\omega) \sin \omega t \langle S^*(0) \rangle e^{-\gamma_0 t}$$
. (29)

The corresponding result in the RWA is

$$\langle S^{-}(t) \rangle = \langle S^{-}(0) \rangle e^{-i\omega_{0}t - \gamma_{0}t}$$

We have thus shown that (i) I(0) is identical to $I^{(r)}(0)$ if the system is initially excited to a state with zero dipole moment; (ii) if the system is excited to a state with finite dipole moment, then I(0) could be very different from $I^{(r)}(0)$, depending upon

*Research supported by the U. S. Army Research Office (Durham).

¹See, for example, H. Haken, in *Handbuch der Physik*, edited by S. Flügge (Springer, Berlin, 1970), Vol.XXV/ 2C, pp. 28-32, 176, 179.

²W. E. Lamb, Jr., in *Quantum Optics and Electronics*, edited by C. De Witt, A. Blandin, and C. Cohen-Tannoudji (Gordon and Breach, New York, 1965), p. 329.

³F. Bloch and A. Siegert, Phys. Rev. <u>57</u>, 522 (1940).
⁴I. F. Rabi, N. F. Ramsey, and J. Schwinger, Rev. Mod. Phys. <u>26</u>, 167 (1954); A. Abragam, *Principle of Nuclear Magnetic Resonance* (Clarendon, London, 1962).

 ${}^{5}An$ exception to this is the recent work of L. Mandel and D. Meltzer [Phys. Rev. <u>188</u>, 198 (1969)], where photoelectric detection of light (absorption process) without the use of the RWA was studied and certain new features of the photoelectric emission process were found.

⁶R. H. Dicke, Phys. Rev. <u>93</u>, 99 (1954).

⁷G. S. Agarwal, Phys. Rev. A $\underline{2}$, 2038 (1970); $\underline{3}$, 1783 (1971); this issue, $\underline{4}$, 1791 (1971). These papers are referred to as I, II, and III, respectively.

⁸J. H. Eberly and N. Rehler, Phys. Rev. A <u>3</u>, 1735 (1971); R. H. Lehmberg, *ibid*. <u>2</u>, 883 (1970); D. Dialetis, *ibid*. A <u>2</u>, 599 (1970); R. Bonifacio, P. Schwendimann, and F. Haake *ibid*. (to be published).

⁹For the master equation techniques, see, for example, G. S. Agarwal, Phys. Rev. <u>178</u>, 2025 (1969), and the references therein.

¹⁰This Fokker-Planck equation is easily obtained from the general theory developed in Ref. 9. A generalized form of the Fokker-Planck equation (4) corresponding to finite temperature has been studied in detail by the author [Phys. Rev. A $\underline{4}$, 739 (1971)] in connection with the Brownian motion of a quantum oscillator.

¹¹G. S. Agarwal and E. Wolf, Phys. Rev. D <u>2</u>, 2161 (1970); 2, 2187 (1970).

¹²E. C. G. Sudarshan, Phys. Rev. Letters <u>10</u>, 277 (1963); R. J. Glauber, Phys. Rev. <u>131</u>, 2766 (1963).

¹³For this equivalence see, for example, R. L. Strat-

the phase of the initial dipole moment; and (iii) for the case when each two-level atom is replaced by a harmonic oscillator, I(t) in the weak-coupling limit reduces to $I^{(r)}(t)$, provided that an averaging over a few optical periods is performed. We expect that such a result will also hold good for the problem of two-level atoms emitting spontaneously.

The author is pleased to acknowledge several interesting conversations with Professor L. Mandel, Professor J. H. Eberly, and Professor E. Wolf on the subject of the rotating-wave approximation.

onovich, Topics in the Theory of Random Noise (Gordon and Breach, New York, 1963), Vol. I.

¹⁴It should be noted that terms like $a_i^{\dagger} a_j^{\dagger} \rho$, $\rho a_i a_j$, and $a_i^{\dagger} \rho a_j$ (or their *c*-number analogs) do not appear in the master equation (6) [or in Eq. (4)], for these terms are multiplied by a coefficient of the form $\sum_{ks} |g_{ks}|^2 \delta(\omega_{kS} + \omega_0)$. This coefficient is identically equal to zero since all the frequencies are by definition positive quantities. The same is true for the case of two-level atoms.

 15 In deriving (6) [or, equivalently, (4)] we have ignored the "frequency shift" terms. When these terms are taken into account, Eq. (4) is modified to

$$\frac{\partial F^{(A)}}{\partial t} = i\omega_0 \sum_i \left(z_i \frac{\partial}{\partial z_i} - z_i^* \frac{\partial}{\partial z_i^*} \right) F^{(A)} + \gamma_0 \sum_{ij} \left\{ \frac{\partial}{\partial z_i} (z_j - z_j^*) F^{(A)} + \text{c.c.} \right\} + i\Delta \sum_{ij} \left\{ \frac{\partial}{\partial z_i} (z_j + z_j^*) F^{(A)} - \text{c.c.} \right\}, \quad (R1)$$

where $\Delta = p \sum_{kS} |g_{kS}|^2 (\omega_{kS} - \omega_0)^{-1}$ and is related to the frequency shift. The corresponding Langevin equations are

$$\dot{z}_{i} = -i\omega_{0}z_{i} - \gamma_{0}\sum_{j}(z_{j} - z_{j}^{*}) - i\Delta\sum_{j}(z_{j} + z_{j}^{*}).$$
(R2)

On introducing the real coordinates q_j , p_j and the collective coordinates Q, P by the relations

$$z_{j} = \left(\frac{m\omega_{0}}{2}\right)^{1/2} q_{j} + i \left(\frac{1}{2m\omega_{0}}\right)^{1/2} p_{j}, \quad Q = \sum_{j} q_{j}, \quad P = \sum_{j} p_{j}, \quad (R3)$$

we find that (R2) reduces to

$$Q = P/m, P = -m\omega_{\rm RN}^2 Q - 2\gamma_0 NP.$$
 (R4)

Here $\omega_{\rm RN}$ is the renormalized frequency and is given by $\omega_{\rm RN}^2 = \omega_0^2 + 2\omega_0 \Delta N$. Thus the effect of the last term on the right-hand side of (R1) can be taken into account by "frequency renormalization" insofar as one is concerned with the properties of the spontaneously emitted radiation which are determined by the properties of the "collective coordinates" [cf. Eq. (9)] Q and P.