Superradiant Effects in Systems of Two-Level Atoms*

C. R. Stroud, Jr.
Institute of Optics, University of Rochester, Rochester, New York 14627

and

J. H. Eberly,† W. L. Lama, and L. Mandel
Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627
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The problem of the evolution in time of a system of two-level atoms that are coupled through their electromagnetic fields, is studied and solved in the framework of semiclassical radiation theory. The atoms may be in any initial states and the radiation reaction is fully taken into account. Both superradiance and time-dependent frequency shift or chirping effects are found. The assumption that all the atoms see the same field is shown to place severe constraints on the evolution of the system, in the form of constants of the motion. These constraints are most easily pictured in terms of the Bloch-vector representation of the atomic system, and they lead to explicit solutions for the time development of each atom. It is shown that the evolution of the complex system is describable by means of a collective super Bloch vector, whose behavior is similar to the behavior of the Bloch vector for a single isolated atom. The constraints on the motion also imply that the system cannot radiate all its energy coherently; some of it remains trapped, to be dissipated ultimately by incoherent processes. Some curves are presented to illustrate the behavior of the system in special cases.

I. INTRODUCTION

Cooperative effects in the radiative decay of assemblies of excited atoms or molecules have been studied extensively, and a wide variety of theoretical treatments is now available. All of these studies have been concerned with one or another of the effects arising from the interatom coherence induced by the electromagnetic fields of the atoms themselves.

Two principal themes have characterized most previous work, and we attempt here to combine them in a unified way. The first of these themes is primarily concerned with the radiation emitted by a very large collection of identical excited atoms. Such radiation can be anomalously intense, in the sense that the radiated intensity can be proportional to the square of the number of atoms participating. The coherent or cooperating quantum states of a large number N of emitting atoms were called "superradiant" by Dicke,[1] and the cooperative emission phenomenon itself is now generally termed superradiance.[2,3]

The second theme which has received repeated attention centers on the reactive (or dispersive) forces acting on an atom due to its own field or the field of another identical atom.[4,5] Studies of such effects have been confined to only one or two atoms at a time.

It seems clear, however, that these two themes are not really distinct. For the superradiant aspects of the emission from N atoms are due precisely to the ability of each atom's field to act on all the other atoms. We will show that, under certain plausible assumptions, it is possible to take careful account of these individual reaction fields and their action on the assembly of emitters. Furthermore, by doing so we are freed from two important restrictive assumptions made in previous work on superradiance.

Our principal assumptions are two: First of all, we assume that all of the atoms in the assembly react to the same electromagnetic field, and second, that semiclassical radiation theory is adequate to describe the motion of the coupled system of atoms. The first assumption requires that the atoms be at least within a wavelength of each other, and implies a restriction to samples smaller than a wavelength. We shall give some justification for the assumption when the number of atoms is very large. The validity of the second assumption is not easy to assess, and we rely upon certain aspects of our results to justify it indirectly. In particular, we will show that, again for sufficiently large numbers of atoms, it leads to the same radiated intensity as comparable calculations[2,3] by quantum electrodynamics. As usual we take the atoms to have only two levels of interest, and we work in the dipole approximation.

The assumptions from which our work is free, which have usually been made heretofore in investigations of supperradiant damping, are these: (i) that there is no atomic dipole frequency modulation during the emission process, and (ii) that the basic dynamics of the total system of N atoms plus emitted radiation is adequately derived from energy-balance arguments.

As a result of the absence of the first restric-

tion, our theory allows the possibility of frequency modulation (or chirping) of the emitted electric field. We find such chirping to be present, and we are able to describe it in detail.

The absence of the second restriction means that we account for the evolution of each atom in the assembly of emitters and, in particular, for the way the electric field emitted by any one of them affects each of the others. In a sense, we have solved a new N-body problem. We have done so in the traditional way, by reducing it to a known one-body problem, already solved by one of us.[5]

The remainder of the paper is organized into six sections and an Appendix. In Sec. II we review the equations of motion appropriate to our N atoms and radiation field, and cast them into the familiar Bloch-vector form. We point out the existence of certain constants of motion. We find that the motion of a "super Bloch vector," defined for all N atoms at once, provides the clue to the evolution of the individual atomic state. In Sec. III we obtain the solutions for the motion of the super Bloch vector, with full account taken of radiation reaction. From the super Bloch-vector solution we find immediately the intensity of the superradiant emission and the time dependence of the frequency modulation of the electric field. Graphs are given to illustrate the principal features of the super Bloch-vector evolution.

In Sec. IV we return to the problem of an individual atom in the assembly and derive explicit but rather complicated analytic expressions for the motion of the individual Bloch vector. We show that, in the coordinate frame in which the rapidly moving super Bloch vector is constant, a clear understanding of the time development of an individual atom is possible. We then exploit these individual atom solutions in Sec. V. We discuss the superradiance to be expected from groups of atoms, when each group is prepared differently. We find energy trapping in most cases, and sometimes nearly complete trapping. Section VI is devoted to a brief discussion of the consequences of our analysis when there is pumping of an assembly of emitters by an external field. Finally, in Sec. VII, we summarize our findings. The paper concludes with an Appendix in which certain aspects of the calculation are clarified.

II. EQUATIONS OF MOTION

We take the interaction energy of the set of two-level atoms to be given by

$$\hat{H} = -\sum_l \hat{\vec{\mu}}_l \cdot \vec{\mathrm{E}}_l \,, \tag{1}$$

where $\hat{\vec{\mu}}_l$ is the dipole-moment operator of the lth atom and $\vec{\mathrm{E}}_l$ is the total electric field seen by this atom.[6] We shall treat this field as a c number, and make the assumption that the atoms are all identical, have energy level separations $\hbar\omega$, and are close together within a region of linear dimensions much smaller than a wavelength. Since the dipole moments associated with different atoms commute, it follows from the form (1) of the interaction that the time evolution operator for the entire system factorizes into a product of time evolution operators for the separate atoms. If the state of the collective atomic system starts as a product state over separate atoms at some time $t = 0$, it will therefore remain a product state indefinitely.

We may therefore write an equation of motion in the interaction picture governing the evolution of the state $|\psi_l(t)\rangle$ of the lth atom in the form

$$-\hat{\vec{\mu}}_l \cdot \vec{\mathrm{E}}_l |\psi_l(t)\rangle = i\hbar \frac{\partial}{\partial t} |\psi_l(t)\rangle \,. \tag{2}$$

For atoms having only one transition of interest, it is customary to write the state of any atom as a time-dependent superposition of upper and lower states

$$|\psi_l(t)\rangle = a_l(t)|+_l\rangle + b_l(t)|-_l\rangle \,, \tag{3}$$

where $|a_l|^2 + |b_l|^2 = 1$ expresses the constant normalization of $|\psi_l(t)\rangle$ for the atom. It is well known that for electric as well as magnetic dipole transitions one can incorporate both Schrödinger's equation and the normalization condition into one vector relation[7]:

$$\frac{d}{dt}\vec{\mathrm{r}}_l = \vec{\Omega}_l \times \vec{\mathrm{r}}_l \,, \tag{4}$$

in which $\vec{\mathrm{r}}_l$ is the so-called Bloch vector characterizing the state of the atom on the unit sphere. The Cartesian components $x_l(t)$, $y_l(t)$, and $z_l(t)$ of the vector $\vec{\mathrm{r}}_l(t)$ are related to the expectation values in state $|\psi_l(t)\rangle$ of the dipole-moment operator, of its time derivative, and of the unperturbed atom energy. In the interaction picture these relations take the form

$$\mu x_l(t) = \langle \hat{\mu}_l \rangle \cos\omega t - \frac{1}{\omega}\left(\frac{d}{dt}\langle \hat{\mu}_l \rangle\right)\sin\omega t \,, \tag{5}$$

$$\mu y_l(t) = -\langle \hat{\mu}_l \rangle \sin\omega t - \frac{1}{\omega}\left(\frac{d}{dt}\langle \hat{\mu}_l \rangle\right)\cos\omega t \,, \tag{6}$$

$$z_l(t) = (2/\hbar\omega)\langle \hat{H}_l^{(A)} \rangle \,. \tag{7}$$

$\hat{H}_l^{(A)}$ is the unperturbed energy of the lth atom. The constants ω and μ are the atomic transition frequency and dipole matrix element, respectively, and the vector frequency $\vec{\Omega}_l(t)$ is given by

$$\vec{\Omega}_l = -2(\mu/\hbar)\vec{\mathrm{n}} \cdot \vec{\mathrm{E}}_l (\vec{\epsilon}_x \cos\omega t - \vec{\epsilon}_y \sin\omega t) \,, \tag{8}$$

where $\vec{\epsilon}_x$ and $\vec{\epsilon}_y$ are unit vectors in the x and y directions on the Bloch sphere, and $\vec{\mathrm{n}}$ is a unit vector in the direction of the dipole moment.[8]

Equations (4)–(8) describe the manner in which the lth atom responds to the total electric field $\vec{E}_l(t)$ seen by it. Our self-consistent approach to the problem of mutually interacting and radiating atoms, or dipoles, now requires that we determine the field $\vec{E}_l(t)$ in terms of the dipole moments present in the assembly of atoms. This field $\vec{E}_l(t)$ seen by the lth atom will be composed of two parts: the field due to all $N-1$ other atoms at atom l, which we denote by $\vec{E}_l^D(t)$, and the radiation reaction field $\vec{E}_l^R(t)$ of the lth atom itself. In a series of papers with several collaborators, Jaynes has considered the effect of this radiation reaction field on a single atomic dipole.[4,5] We shall make use of some of the results in these papers, particularly in the form given by Stroud and Jaynes.[5,9]

The radiation reaction field is given by[5]

$$\vec{E}_l^R(t)=\frac{4\omega^2K}{3\pi c^3}\,\vec{n}\,\langle\hat{\mu}_l\rangle-\frac{2\omega^2}{3c^3}\,\vec{n}\frac{d}{dt}\langle\hat{\mu}_l\rangle\,, \tag{9}$$

where $\vec{n}$ is a unit vector in the direction of the atomic dipole. The constant frequency K depends on the detailed nature of the atomic charge distribution. For any reasonable charge distribution K^{-1} is expected to be roughly equal to a_0/c, the time for a light signal to traverse an atomic radius a_0. Crisp and Jaynes[4] have evaluated K in a particular case.

The field due to another atom, say atom k, acting on atom l, depends on the distance d_{lk} and on the position of atom k. Like $\vec{E}_l^R(t)$, this field $\vec{E}_{lk}(t)$ contains contributions proportional to the expectation value of the dipole moment $\hat{\mu}_k$ and its derivative, and we have, from the general expression for the near field of a dipole,[10]

$$\vec{E}_{lk}(t)=\left(\frac{3(\vec{n}\cdot\vec{u})\vec{u}-\vec{n}}{d_{lk}^{\,3}}+\frac{\frac{1}{2}(\vec{n}\cdot\vec{u})\vec{u}\omega^2+\frac{1}{2}\vec{n}\omega^2}{c^2d_{lk}}\right)\langle\hat{\mu}_k\rangle$$
$$-\frac{2\vec{n}\omega^2}{3c^3}\frac{d}{dt}\langle\hat{\mu}_k\rangle\,, \tag{10}$$

where $\vec{u}$ is a unit vector in the direction of the vector from atom k to atom l. On summing over all atoms $k\neq l$, we arrive at the total field $\vec{E}_l^D(t)$ due to all other atoms seen by atom l. It is convenient to write this sum in the form

$$\vec{E}_l^D(t)=\vec{A}\sum_{k\neq l}\langle\hat{\mu}_k\rangle-B\vec{n}\sum_{k\neq l}\frac{d}{dt}\langle\hat{\mu}_k\rangle\,, \tag{11}$$

where

$$\vec{A}\equiv\sum_{k\neq l}\left(\frac{3(\vec{n}\cdot\vec{u})\vec{u}-\vec{n}}{d_{lk}^{\,3}}+\frac{\frac{1}{2}(\vec{n}\cdot\vec{u})\vec{u}\,\omega^2+\frac{1}{2}\vec{n}\omega^2}{c^2d_{lk}}\right)\langle\hat{\mu}_k\rangle$$
$$\Big/\sum_{k\neq l}\langle\hat{\mu}_k\rangle \tag{11a}$$

and

$$B\equiv 2\omega^2/3c^3\,. \tag{11b}$$

Coefficient B is a constant and identical with the coefficient of the corresponding term in Eq. (9). Coefficient $\vec{A}$ may be regarded as the average over atoms, in a sense, of the corresponding coefficient in Eq. (10). In the following we shall make the simplifying assumption that $\vec{A}$ is substantially similar for all atoms l, and may also be regarded as a constant. By way of justification we first note that, since the sample is much smaller than a wavelength, the term proportional to $1/d_{lk}^{\,3}$ in Eq. (11a) substantially exceeds the one proportional to $1/c^2d_{lk}$. Now, because of the factor $1/d_{lk}^{\,3}$, the principal contribution to the sum comes from atoms in the neighborhood of atom l. When we are dealing with a dense sample of randomly oriented atoms, it therefore seems reasonable to suppose that the sums in Eq. (11a) have similar values for all atoms, except perhaps for those atoms lying close to the boundary of the sample.

The total field $\vec{E}_l(t)$ acting on atom l is of course the sum of $\vec{E}_l^R(t)$ and $\vec{E}_l^D(t)$ given by Eqs. (9) and (11), so that

$$\vec{E}_l(t)=\frac{4\omega^2K}{3\pi c^3}\,\vec{n}\,\langle\hat{\mu}_l\rangle+\vec{A}\sum_{k\neq l}\langle\hat{\mu}_k\rangle-B\vec{n}\sum_k\frac{d}{dt}\langle\hat{\mu}_k\rangle\,.$$

The calculation would obviously be simplified if all atoms, including atom l, contributed symmetrically to $\vec{E}_l(t)$, for we could then combine the first two terms and write

$$\vec{E}_l(t)=\vec{A}\sum_k\langle\hat{\mu}_k\rangle-B\vec{n}\sum_k\frac{d}{dt}\langle\hat{\mu}_k\rangle\,. \tag{12}$$

There may exist geometries and sample densities for which $\vec{A}$ equals or is close to $(4\omega^2K/3\pi c^3)\vec{n}$, so that Eq. (12) is justified directly. But it is not difficult to see that Eq. (12) must be approximately valid irrespective of whether $\vec{A}$ equals $(4\omega^2K/3\pi c^3)\vec{n}$ or not, when the field on atom l due to the other atoms is much larger than its self-field. This will be the case whenever the number of atoms is large and the initial state of the system is prepared by subjecting most of the atoms to the same external perturbation, e.g., a laser pulse. Then the atomic dipoles are not randomly oriented on the Bloch sphere, and the system has a net macroscopic dipole moment. This is just the condition for cooperative superradiant decay to become more important than independent incoherent decay.

In terms of the components of the $\vec{r}_l$ vector we then have from Eqs. (5), (6), and (12),

$$\vec{E}_l(t)=\vec{A}\mu\textstyle\sum_k[x_k(t)\cos\omega t-y_k(t)\sin\omega t]$$
$$+B\omega\mu\vec{n}\textstyle\sum_k[x_k(t)\sin\omega t+y_k(t)\cos\omega t]$$
$$=\mu(\vec{A}X+\omega BY\vec{n})\cos\omega t+\mu(\omega BX\vec{n}-\vec{A}Y)\sin\omega t\,, \tag{13}$$

which is independent of l. X and Y stand for the sums $\sum_k x_k(t)$ and $\sum_k y_k(t)$, respectively.

Having now determined the field at the lth atom, we may insert it into the Schrödinger equation via $\vec{\Omega}_l$ given by Eq. (8). We see immediately that $\vec{\Omega}_l$ is actually independent of the index l and may be denoted by $\vec{\Omega}$, so that there is no longer any difference in the dynamical laws obeyed by the different atoms, and we may write

$$\frac{d}{dt}\vec{r}_l = \vec{\Omega}\times\vec{r}_l \ . \tag{14}$$

Several constants of the motion follow directly from the nondependence of $\vec{\Omega}$ on l. By definition, each of the Bloch vectors has unit length.[8] Moreover, the angle between any two of them is a constant fixed by initial conditions. A proof is trivially constructed by taking the inner product of Eq. (14) with $\vec{r}_k$ and adding the corresponding equation with l and k interchanged. Then we find

$$\begin{aligned}\frac{d}{dt}(\vec{r}_k\cdot\vec{r}_l) &= (\vec{\Omega}\times\vec{r}_k)\cdot\vec{r}_l + (\vec{\Omega}\times\vec{r}_l)\cdot\vec{r}_k \\ &= \vec{\Omega}\cdot(\vec{r}_k\times\vec{r}_l) + \vec{\Omega}\cdot(\vec{r}_l\times\vec{r}_k) \\ &= 0\end{aligned}$$

or

$$\vec{r}_k\cdot\vec{r}_l = \cos\psi_{kl} = \text{const} \ , \tag{15}$$

where ψ_{kl} is the (constant) angle between the Bloch vectors $\vec{r}_k$ and $\vec{r}_l$. The constancy of the angle between the vectors, together with their fixed unit length, implies that the evolution of the N-atom system can be viewed as a rigid rotation of the corresponding set of Bloch vectors.[11] This conclusion is already obvious by inspection of Eq. (14), which describes the motion of a point $\vec{r}_l$ of a rigid body that is rotating with angular velocity $\vec{\Omega}$.

We should emphasize that this conclusion follows from the assumption that all the atoms see the same field and is independent of the assumed form of the field. In Sec. VI we shall consider the effect of including an external field also.

III. SUPER BLOCH VECTOR

As a consequence of the fact that each atomic Bloch vector obeys the same equation of motion we may define a super Bloch vector

$$\vec{R}(t) \equiv \sum_{l=1}^{N} \vec{r}_l(t) \ ,$$

with components $X(t)$, $Y(t)$, and $Z(t)$, which obeys a dynamical law of the same form as the law for an individual Bloch vector;

$$\frac{d}{dt}\vec{R} = \vec{\Omega}\times\vec{R} \ . \tag{16}$$

We now insert the expression (13) for the field into Eq. (8) for $\vec{\Omega}$, and introduce the rotating wave approximation. We then obtain

$$\vec{\Omega} = -\vec{\epsilon}_x(\gamma X + \beta Y) + \vec{\epsilon}_y(\beta X - \gamma Y) \ , \tag{17}$$

where the frequencies β and γ are defined[9] by

$$2\beta = 4\mu^2\omega^3/3\hbar c^3 = (\text{natural atomic lifetime})^{-1} \ , \tag{18a}$$

$$\gamma = \frac{\mu^2}{\hbar}\left(\frac{3(\vec{n}\cdot\vec{u})^2 - 1}{d_{lk}^{\,3}} + \omega^2\frac{(\vec{n}\cdot\vec{u})^2 + 1}{2c^2 d_{lk}}\right)_{\text{average over atoms}} \tag{18b}$$

It is important to note that the super Bloch vector $\vec{R}$ precesses about a time-dependent vector $\vec{\Omega}$ [cf. Eq. (16)], which is a linear homogeneous function of the components of $\vec{R}$ itself [cf. Eq. (17)]. In fact, $\vec{\Omega}$ is the same function of $\vec{R}$ as $\vec{\Omega}_1$ would be of $\vec{r}_1$, if we were dealing with a single isolated atom. It follows that the solution of the radiative decay problem for a single isolated atom in the presence of its own reaction field is also the solution of the problem of the motion of the N-atom super Bloch vector. But the problem of the single-atom decay has already been investigated and solved in detail.[5]

A second consequence of the fact that $\vec{\Omega}$, given by Eq. (17), is a linear, homogeneous function of the super Bloch vector $\vec{R}$ is that the rate of change of the super Bloch vector is R times as rapid as the rate of change of the vector $\vec{r}_1$ describing an isolated single atom. This implies that the N-atom system described by the super Bloch vector will be superradiant, if the length of the super Bloch vector is of order N. The length of the super Bloch vector is easily determined from the initial conditions, because we have shown already that

$$\vec{r}_k(t)\cdot\vec{r}_l(t) = \vec{r}_k(0)\cdot\vec{r}_l(0) \ , \tag{19}$$

so that

$$R^2(t) = \sum_k\sum_l \vec{r}_k(t)\cdot\vec{r}_l(t) = R^2(0) \ . \tag{20}$$

The length $R(0)$, which we denote simply by R, can have any value from 0 to N, depending on the initial alignments of the N individual Bloch vectors. If the vectors are initially aligned, or nearly so, then $R \sim N$ and the system will be superradiant.

We may point out an analogy that goes deeper than notation, between the length of the super Bloch vector $\vec{R}$ and the eigenvalue of the "cooperation number" operator $\hat{R}$ introduced by Dicke.[1] The cooperation number operator satisfies

$$\hat{R}^2 = \hat{R}_1^2 + \hat{R}_2^2 + \hat{R}_3^2 \ , \tag{21}$$

and the component operators are closely related to the components of our super Bloch vector $\vec{R}$ by the equations

$$X = \langle\hat{R}_1\rangle \ , \quad Y = \langle\hat{R}_2\rangle \ , \quad Z = \langle\hat{R}_3\rangle \ . \tag{22}$$

The solution of the equation of motion for the super Bloch vector can be found exactly. On substituting Eq. (17) in Eq. (16) and equating components, we obtain

$$\dot{X} = (\beta X - \gamma Y)Z \,, \tag{23}$$

$$\dot{Y} = (\gamma X + \beta Y)Z \,, \tag{24}$$

$$\dot{Z} = -\beta(X^2 + Y^2) = -\beta(R^2 - Z^2) \,. \tag{25}$$

The solutions are most easily expressed in terms of the polar and azimuthal angles Θ and Φ of the vector $\vec{R}$ in spherical coordinates. If we define

$$X = R\sin\Theta\cos\Phi \,, \quad Y = R\sin\Theta\sin\Phi \,, \quad Z = R\cos\Theta \,, \tag{26}$$

then Eq. (25) reduces to

$$\dot{\Theta} = R\beta\sin\Theta \,, \tag{27}$$

and the combination of Eq. (23) with Eq. (27) yields

$$\dot{\Phi} = R\gamma\cos\Theta \,. \tag{28}$$

Equations (23)–(25) are the same as those given and solved by Bloom[12] in an earlier discussion of radiation damping, except for the appearance here of the frequency-shift parameter γ. The relation (27) has of course long been known.[13] In Eq. (28) we see clearly the importance of γ for radiation reaction damping. Its presence implies a frequency modulation of the atomic dipole oscillation. The same phenomenon is also well known in the decay of coupled classical oscillators.[14]

Equations (27) and (28) can be integrated immediately,[5] to give

$$\Theta(t) = 2\tan^{-1} e^{R\beta(t - t_0)} \tag{29}$$

or

$$\cos\Theta(t) = -\tanh[R\beta(t - t_0)] \,, \tag{30}$$

$$\Phi(t) - \Phi(0) = -\frac{\gamma}{\beta}\ln\left(\frac{\cosh[R\beta(t - t_0)]}{\cosh(R\beta t_0)}\right) \tag{31}$$

or

$$\dot{\Phi}(t) = -R\gamma\tanh[R\beta(t - t_0)] \,. \tag{32}$$

The parameter t_0 is defined by

$$\tan\tfrac{1}{2}\Theta(0) = e^{-R\beta t_0} \,, \tag{33}$$

and depends only on the initial energy of the system.

If we eliminate the time t between Eqs. (29) and (31), we obtain the equation of the orbit of the super Bloch vector;

$$\sin\Theta = \sin\Theta(0)\, e^{-(\beta/\gamma)[\Phi - \Phi(0)]} \,. \tag{34}$$

This trajectory on the Bloch sphere is illustrated in Fig. 1 for the case $\gamma/\beta = 10$.

The total energy $W(t)$ of the atomic system is given by $\frac{1}{2}\hbar\omega Z(t)$ and we see from Eq. (30) that

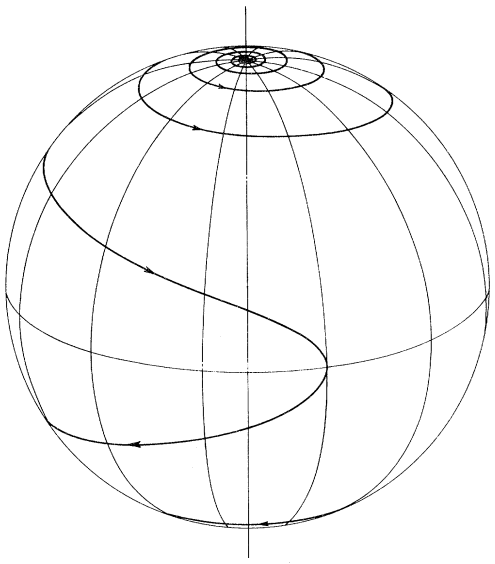

FIG. 1. Trajectory of the super Bloch vector $\vec{R}$ on the sphere, with $\gamma/\beta = 10$.

$$W(t) = \tfrac{1}{2}\hbar\omega R\cos\Theta(t) = -\tfrac{1}{2}\hbar\omega R\tanh[R\beta(t - t_0)] \,, \tag{35}$$

which decays in time as a hyperbolic tangent. This behavior is illustrated in Fig. 2.

The coherent radiation rate is readily calculated. The radiant power $I(t)$ is given by the rate at which the atomic system loses energy, i.e.,

$$I(t) = (-\tfrac{1}{2}\hbar\omega)\,\dot{Z}(t) = \tfrac{1}{2}\hbar\omega R^2\beta\,\mathrm{sech}^2[R\beta(t - t_0)] \,, \tag{36}$$

which can range from 0 to N^2 times the single-atom rate. This result is familiar from other treatments of superradiance. Rehler and Eberly[2] have shown, for example, that for the special case in which all the Bloch vectors are initially aligned, $N \gg 1$, and the linear dimensions of the sample are much smaller than a wavelength,

$$I(t) = \tfrac{1}{2}\hbar\omega N^2\beta\,\mathrm{sech}^2[N\beta(t - t_0)] \,. \tag{37}$$

In the sense that we may have $R < N$, the present solution (36) is more general. The time dependence of $I(t)$ is also illustrated in Fig. 2.

Several features of the solution given by Eqs. (29)–(32) are worth noting. There is a metastable point at $\Theta = 0$, so that, in this theory, a system of atoms prepared exactly in its upper state never radiates. The reason is that, in a semiclassical theory, radiation is possible only when the system has a nonzero dipole moment. However, once perturbed away from the excited state, however slightly, the system does radiate and does so with the same characteristic lifetime $(R\beta)^{-1}$ predicted by quantum electrodynamics.[2,3]

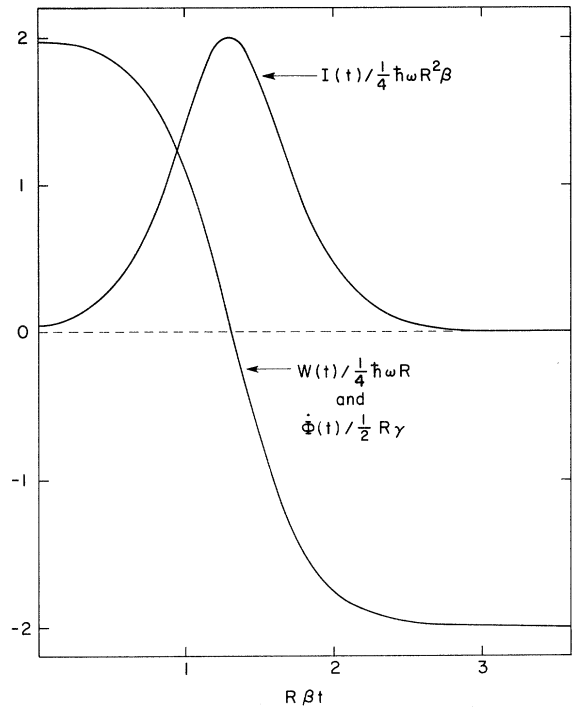

FIG. 2. Time dependence of the total energy $W(t)$ of the atomic system, the power of electromagnetic radiation $I(t)$, and the chirp frequency $\dot{\Phi}(t)$ of the super Bloch vector. The initial conditions are such that $\Theta(0)=0.14$, which makes $R\beta t_0=1.32$.

Another remarkable feature of the solution is that, after the system decays to its steady state in which $\Theta=\pi$, i.e., $Z=-R$, there is in general still energy left trapped in the system. The final state is therefore not the state in which all N of the atoms are in their respective ground states ($Z=-N$). Because of our conservation laws, all of the atoms cannot decay to the ground state. They can only decay to a state such that the total energy is a minimum subject to Eq. (15), i.e., to a state such that their super Bloch vector points straight down, when the total dipole moment vanishes. The system of atoms can then no longer radiate coherently, and the remaining energy will be dissipated by whatever incoherent processes are available to the atoms.

Another interesting feature of the solution concerns the rate of change of the phase angle Φ of the dipole moment, which follows a hyperbolic tangent curve according to Eq. (32). This means that the emitted radiation is chirped. The frequency of the emitted radiation starts at $\omega+R\gamma$ near $\Theta=0$ and decreases monotonically to $\omega-R\gamma$ near $\Theta=\pi$. This behavior is also illustrated in Fig. 2.

It should be pointed out that the system is not normally prepared in a metastable state. In fact it would be very difficult to do so in practice. In general, only a portion of the various curves is traced out as t runs from 0 to ∞. For example, if $\Theta(0)=\frac{1}{2}\pi$, then only half of each curve is traced out, and if $\Theta(0)<\frac{1}{2}\pi$ even less of the curve is traced out, perhaps only the exponential tail.

IV. MOTION OF THE BLOCH VECTOR OF AN INDIVIDUAL ATOM

It is possible to determine the motion, not only of the super Bloch vector, but also of the Bloch vectors of the individual atoms. The simplest way to understand this motion is to return to the equations of motion (14) and (16) for $\vec{r}_l$ and $\vec{R}$, respectively. Both $\vec{R}$ and the individual $\vec{r}_l$ precess about the vector $\vec{\Omega}$, which is itself moving in the x-y plane. The motion can be broken up into a sum of simpler motions if we note that the vector $\vec{\Omega}$, given by Eq. (17), can be written in the form

$$\vec{\Omega}=-\gamma\vec{R}+\gamma Z\vec{\epsilon}_z+\beta(\vec{\epsilon}_z\times\vec{R})\ . \tag{38}$$

From Eqs. (16) and (38), the equation of motion for $\vec{R}$ can then be expressed as

$$\dot{\vec{R}}=\gamma Z(\vec{\epsilon}_z\times\vec{R})+\beta(\vec{\epsilon}_z\times\vec{R})\times\vec{R}\ . \tag{39}$$

The motion of $\vec{R}$ can therefore be pictured as a combination of two motions, a precession about the z axis at a rate γZ, and a precession at a rate $R\beta\sin\Theta$ about the vector $\vec{\epsilon}_z\times\vec{R}$. These same conclusions can of course be drawn more directly from Eqs. (27) and (28). However, this method of decomposing the motion of the vector $\vec{R}$ is also applicable to the motion of the individual $\vec{r}_l$. Thus, the equation of motion for $\vec{r}_l$ may be written in the form

$$\dot{\vec{r}}_l=\gamma Z(\vec{\epsilon}_z\times\vec{r}_l)+\beta(\vec{\epsilon}_z\times\vec{R})\times\vec{r}_l-\gamma(\vec{R}\times\vec{r}_l)\ . \tag{40}$$

The first two terms describe exactly the same motion as Eq. (39) for $\vec{R}$, but the last term describes an additional precession about the super Bloch vector at a constant rate $-\gamma R$. The individual Bloch vectors simply precess about the super Bloch vector at a constant rate $-\gamma R$, while maintaining their original angular separations. This result allows us to obtain closed form solutions for the individual angular coordinates $\theta_l(t)$ and $\phi_l(t)$ (see Appendix). In the rotating frame these solutions may be written

$$\cos\theta_l(t)=-\cos\psi_l\tanh R\beta(t-t_0)+\sin\psi_l\,\mathrm{sech}R\beta(t-t_0)\cos R\gamma(t-t_{0l}) \tag{41}$$

and

$$\phi_l(t)-\phi_l(0)=(\gamma/\beta)\ln[\mathrm{sech}R\beta(t-t_0)/\mathrm{sech}R\beta t_0]+\sin^{-1}[\mathrm{cosec}\theta_l(t)\sin\psi_l\sin R\gamma(t-t_{0l})]\ , \tag{42}$$

where

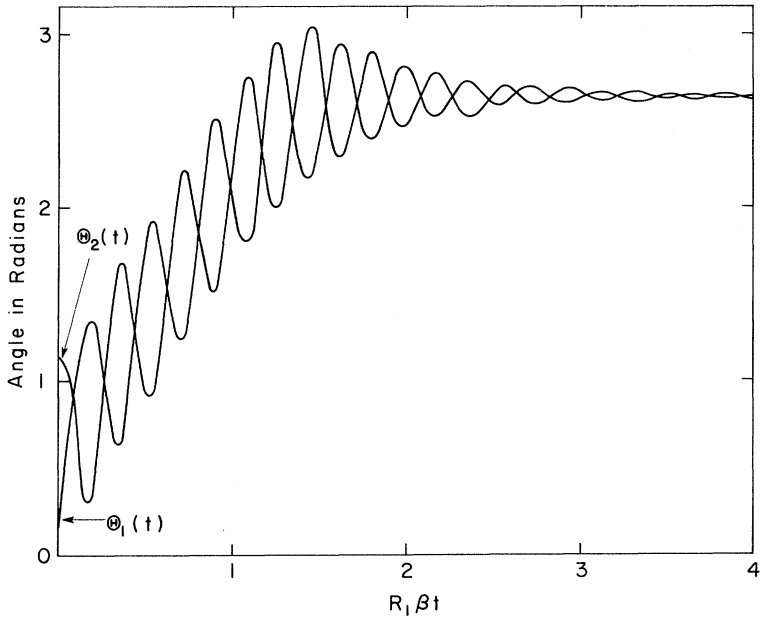

FIG. 3. Time dependence of the two polar angles $\Theta_1(t)$ and $\Theta_2(t)$ for two interacting systems of atoms with equal length super Bloch vectors ($R_1 = R_2$). Both systems are fairly well excited initially, with $\Theta_1(0) = 0.14$, $\Theta_2(0) = 1.14$, and $\Phi_1(0) = 0 = \Phi_2(0)$. Note that the values of $\Theta_1(t)$ and $\Theta_2(t)$ do not tend to π, so that some energy remains trapped.

$$\cos\psi_i = (\vec{r}_i \cdot \vec{R})/R\,, \tag{43a}$$

$$\operatorname{cosec}\theta_i(t) = \{1 - [\cos\psi_i \tanh R\beta(t - t_0) - \sin\psi_i \operatorname{sech} R\beta(t - t_0) \cos R\gamma(t - t_{0i})]^2\}^{-1/2}\,, \tag{43b}$$

and t_{0i} is defined by

$$\cos\theta_i(t_0) = \sin\psi_i \cos R\gamma(t_0 - t_{0i})\,. \tag{43c}$$

Some consequences of these equations will be examined in Sec. V.

V. GROUPS OF ATOMS AND RADIATION TRAPPING

Let us now consider a system composed of two groups of atoms, the atoms in group I being described by the super Bloch vector $\vec{R}_1$, and those in group II by $\vec{R}_2$. This is not really a simplification since any collection of atoms can be similarly de-

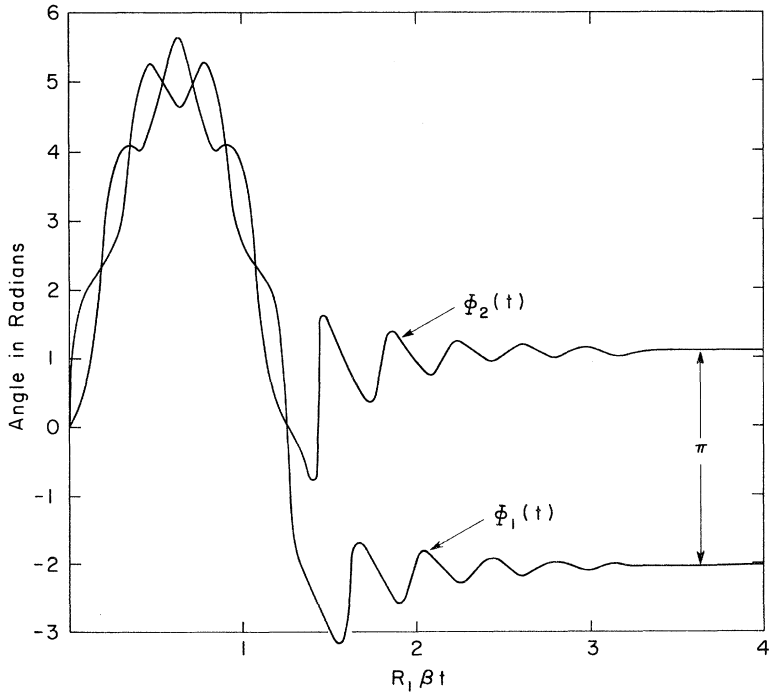

FIG. 4. Time dependence of the two azimuthal angles $\Phi_1(t)$ and $\Phi_2(t)$ for two interacting systems of atoms with equal length super Bloch vectors ($R_1 = R_2$). Both systems are fairly well excited initially, with $\Theta_1(0) = 0.14$, $\Theta_2(0) = 1.14$, and $\Phi_1(0) = 0 = \Phi_2(0)$. Note that the atomic dipoles are aligned in antiphase in the steady state.

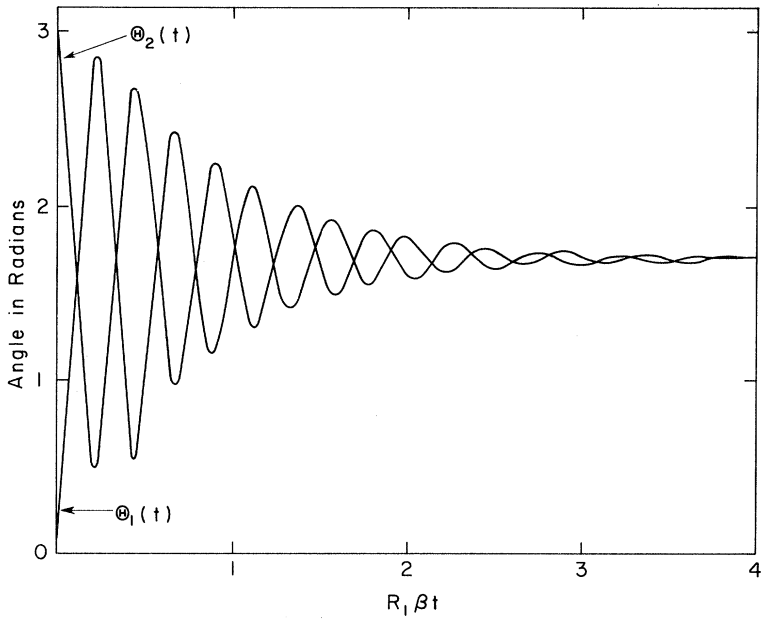

FIG. 5. Time dependence of the two polar angles $\Theta_1(t)$ and $\Theta_2(t)$ for two interacting systems of atoms with equal length super Bloch vectors ($R_1 = R_2$). One system is almost fully excited, while the other one is almost fully deexcited initially, with $\Theta_1(0) = 0.14$, $\Theta_2(0) = 3.0$, and $\Phi_1(0) = 0 = \Phi_2(0)$. Note that energy is continually interchanged between the two systems, and that most of the initial energy remains trapped.

composed. If we are only interested in the evolution of the two super Bloch vectors, this can be obtained immediately from Eqs. (41) and (42), where

$$\cos\psi_l = (\vec{R}_l \cdot \vec{R})/R\,, \quad l = 1, 2 \tag{44}$$

and

$$\vec{R} = \vec{R}_1 + \vec{R}_2\,.$$

Some examples of these solutions for different initial states are illustrated in Figs. 3–6 for the case $R_1 = R_2$. The figures show that the polar angles Θ_l, and therefore also the energies W_l, oscillate while decaying. We also note that the atoms do not settle down in the ground state $\Theta_1 = \pi$ or $\Theta_2 = \pi$, but that some energy is trapped in the system. This energy will ultimately be dissipated by incoherent processes.

Figures 4 and 6 show the phase angles Φ_l. We note that, in each case, the relative phase evolves to π. This is a necessary condition for the system to stop radiating. Moreover, we note that after a long time, $\dot{\Phi}_l = 0$, which means that the frequency modulation of the individual atoms (or groups of atoms) has ended. On the other hand, Eq. (32) shows that, for the super Bloch vector of the complete system, $\dot{\Phi}(\infty) = -R\gamma$. However, there is no contradiction, for the super Bloch vector $\vec{R}$ has zero x and y components at $t \to \infty$, so that Φ and $\dot{\Phi}$ are no longer meaningful quantities.

An interesting application of these ideas is to the problem of incoherent excitation. If a collection of atoms is pumped incoherently, one would expect that most of the atoms would remain in the ground state while some might be pumped to a state represented by a point quite high on the Bloch sphere. Immediately after pumping, the Bloch vectors might be as shown in Fig. 7(a). The small group of atoms represented by the short vector $\vec{e}$ are highly excited, while most of the atoms represented by the long vector $\vec{g}$ are in the ground state. The final state will be the one in which the sum vector $\vec{e}+\vec{g}$ points straight down, Fig. 7(b). But the angle between $\vec{e}$ and $\vec{g}$ is fixed, so that in the final state essentially all of the atoms of group $\vec{e}$ are still excited. They have simply induced a slight amount of dipole moment in the atoms of group $\vec{g}$ to cancel the total dipole moment of the system. One would not expect to see much energy in a superradiant pulse from such a system; most of the energy will be trapped, and ultimately dissipated by incoherent processes.

VI. COHERENT PUMPING

As has been shown, it is only when most of the atoms in the medium are excited that a significant amount of energy is expected to be released in a superradiant pulse. We are therefore led to try to describe coherent pumping of the medium by the same formalism. The equations are easily generalized to include an external applied field. If we simply add to the field seen by the dipoles, given by Eq. (13), an additional external field

$$\vec{E}^{\text{ext}}(t) = \vec{n}\,\mathcal{E}(t)\cos\omega t\,, \tag{45}$$

where $\mathcal{E}(t)$ is a slowly varying envelope, then the equations of motion (27) and (28) become

$$\dot{\Theta} = R\beta\sin\Theta + (\mu/\hbar)\,\mathcal{E}(t)\sin\Phi\,, \tag{46}$$

$$\dot{\Phi} = R\gamma\cos\Theta + (\mu/\hbar)\,\mathcal{E}(t)\cot\Theta\cos\Phi\,. \tag{47}$$

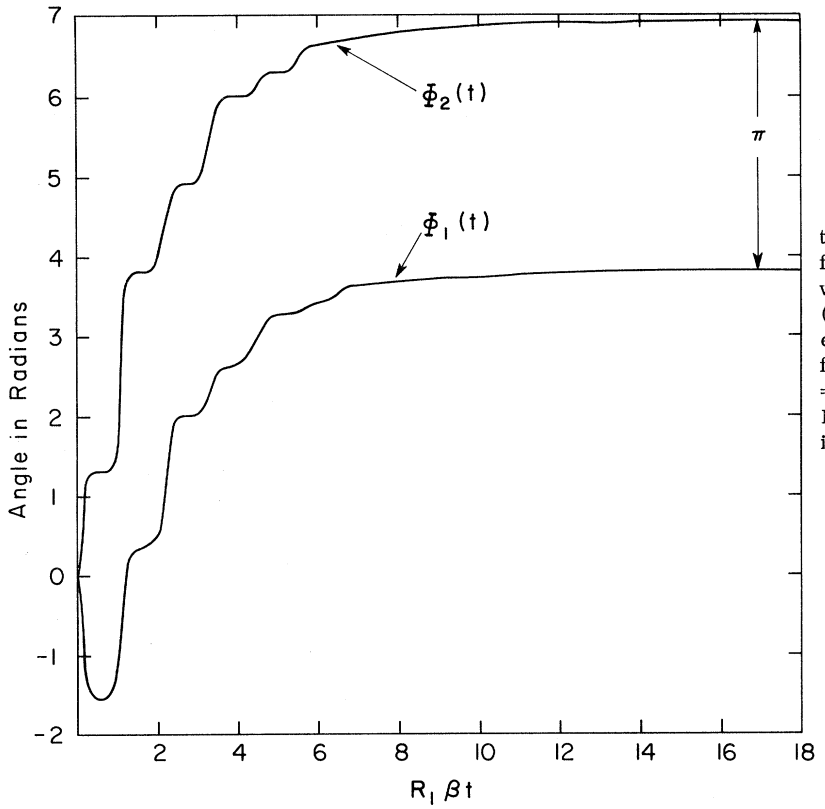

FIG. 6. Time dependence of the two azimuthal angles Φ_1 and $\Phi_2(t)$ for two interacting systems of atoms with equal length super Bloch vectors ($R_1 = R_2$). One system is almost fully excited, while the other one is almost fully deexcited initially, with $\Theta_1(0) = 0.14$, $\Theta_2(0) = 3.0$, and $\Phi_1(0) = 0 = \Phi_2(0)$. Note that the atomic dipoles are aligned in antiphase in the steady state.

We have not obtained the general analytic solutions of these equations, but certain implications can be seen immediately. Whereas the spontaneous damping rate β and the frequency shift γ in Eqs. (46) and (47) are multiplied by the superradiant enhancement factor R, which may be as large as N, the external field $\mathcal{E}(t)$ is not similarly enhanced. An extremely strong applied field will therefore be needed to overcome the enhanced spontaneous emission and to excite the system appreciably.

VII. SUMMARY AND CONCLUSIONS

We have used a semiclassical formalism, in which the expectation values of the atomic dipole-moment operators are taken to be the sources of the classical Maxwell fields, to describe the evolution of a collection of N identical two-level atoms located in a volume with dimensions small compared with a wavelength of the emitted radiation. The effects of dipole-dipole interactions as well as the effects of radiation reaction were included. The resulting equations yielded an analytic solution for the time development of the macroscopic dipole moment of the entire system, for the energy stored in the system, and for the intensity of the radiated field. In the limit of a large number of atoms, the results for the radiated intensity are identical to those obtained previously by other methods, wherever comparison is possible. The radiation produced was found to be chirped; the frequency of the field decreased during the emission. Solutions were also found for the dipole moments and energies of the individual atoms, for arbitrary initial states.

The detailed analytic solutions to the rather complex nonlinear equations were obtained with the help of a number of constants of the motion. The

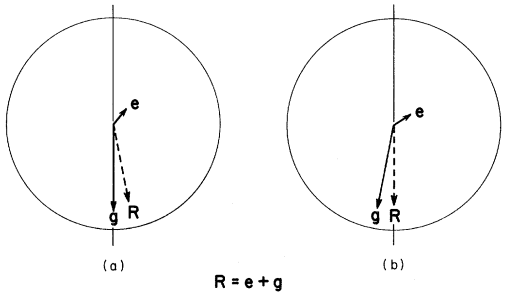

FIG. 7. Illustration of the evolution of a small system of partially excited atoms (super Bloch vector $\vec{e}$) which is interacting with a large system of atoms in the ground state (super Bloch vector $\vec{g}$). The initial state is shown in (a) and the final state in (b). Note that the super Bloch vector $\vec{R}$ turns only slightly, so that very little of the initial energy is radiated coherently.

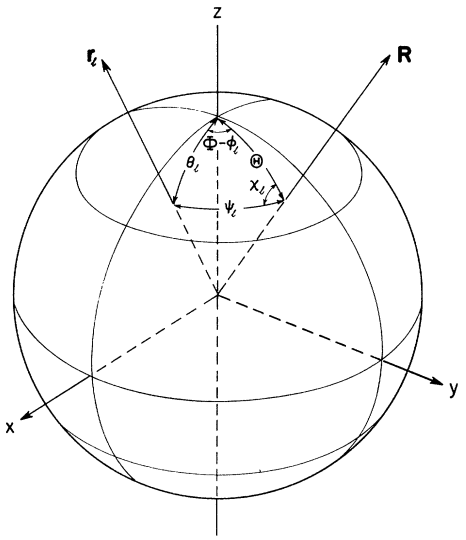

FIG. 8. Relation between the directions of the Bloch vector $\vec{r}_l$ and the super Bloch vector $\vec{R}$.

existence of these constants of the motion, i.e., conservation laws, is perhaps more significant than the solutions themselves, as they determine certain properties of the solutions which are independent of the initial states of the various atoms. The most important conservation law implies that the evolution of the collective atomic system is such that the Bloch vectors on the unit sphere move as a rigidly connected set. This means that the evolution of the entire system can be represented by one super Bloch vector, the vector sum of all the individual Bloch vectors. The conservation law illustrates how strong the coupling is between the atoms. It is so strong that they all evolve together, generating a coherent spontaneous emission pulse which may be N^2 times as intense as the emission from a single isolated atom. The conservation law also requires that if the N atoms are initially not all in exactly the same state, then they cannot all return to their individual ground states by cooperative emission. Some energy will remain trapped in the system, with the dipoles rigidly locked into a configuration with total dipole moment zero. The conservation law results from the assumption that all the atoms are exposed to the same field, which is likely to be only approximately true in practice.

Finally, it should be pointed out again that the results obtained by this semiclassical formalism, sometimes called neoclassical radiation theory, are expected to coincide with those obtained by quantum electrodynamics when the number of atoms is large, as it surely is in practical cases.

APPENDIX: EVOLUTION OF INDIVIDUAL BLOCH VECTORS

To obtain the analytic solutions [Eqs. (41)–(43)] for the motion of the Bloch vector $\vec{r}_l$, it is convenient to transform to a new coordinate system in which the super Bloch vector is at rest (see Fig. 8). From Eq. (40) and the arguments following it one can see that the motion of $\vec{r}_l$ in this coordinate system is a rotation about the axis $\vec{R}$, with a constant polar angle ψ_l and an azimuthal angle χ_l, describing a constant precession at rate $-\gamma R$, i.e., the angle $\chi_l(t)$ defined in Fig. 8 satisfies the equation

$$\chi_l(t) = -R\gamma(t - t_{0l}) \,. \tag{A1}$$

The solutions for the evolution of $\theta_l(t)$ and $\phi_l(t)$ can then be immediately expressed in terms of the known quantities $\Theta(t)$, $\Phi(t)$, and $\chi_l(t)$ by use of some identities of spherical trigonometry. The law of cosines gives (see Fig. 8)

$$\cos\theta_l = \cos\Theta\cos\psi_l + \sin\Theta\sin\psi_l\cos\chi_l \,, \tag{A2}$$

while the law of sines gives

$$\sin(\Phi - \phi_l) = \sin\psi_l \sin\chi_l / \sin\theta_l \,. \tag{A3}$$

On substituting Eqs. (30), (31), and (A1) into Eqs. (A2) and (A3), we obtain the desired results, Eqs. (41)–(43).

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†Address during the 1971–72 academic year: SLAC, Stanford University, Stanford, Calif. 94305.

[1]R. H. Dicke, Phys. Rev. 93, 99 (1954).

[2]One recent treatment of superradiant emission, and a large number of references to other works, is given in N. E. Rehler and J. H. Eberly, Phys. Rev. A 2, 1735 (1971).

[3]G. S. Agarwal, Phys. Rev. A 2, 2038 (1970); Opt. Commun. 2, 357 (1971).

[4]E. T. Jaynes and F. W. Cummings, Proc. IEEE 51, 89 (1963); M. D. Crisp and E. T. Jaynes, Phys. Rev. 179, 1253 (1969); C. S. Chang and P. Stehle, Phys. Rev. A 4, 630 (1971). Other early work is referred to by Chang and Stehle. The previous work most directly important for ours is given in Ref. 5.

[5]C. R. Stroud, Jr. and E. T. Jaynes, Phys. Rev. A 1, 106 (1970); 2, 1613(E) (1970).

[6]We distinguish Hilbert-space operators by the caret.

[7]R. P. Feynman, F. L. Vernon, Jr., and R. W. Hellwarth, J. Appl. Phys. 28, 49 (1956).

[8]It is important to bear in mind that our equations involve vectors in two different spaces; vectors such as $\vec{E}_l$ and $\hat{n}$ are in real space, whereas $\vec{\Omega}_l, \vec{r}_l, \vec{\epsilon}_x$, etc., are vectors in the symbolic space of the Bloch sphere.

[9]It is known (cf. Jaynes *et al.*, Refs. 4 and 5) that a self-consistent semiclassical treatment of single-atom radiation problems can lead to the prediction of radiative level widths and frequency shifts. Whether or not the frequency shifts are precisely the same as those calculated

in quantum electrodynamics has been discussed by Crisp and Jaynes (Ref. 4). Because of our concern with a many-atom system with strong classical analogies we expect the semiclassical results to be completely adequate for our needs.

[10]See, for example, M. Born and E. Wolf, *Principles of Optics*, 4th ed. (Pergamon, New York, 1970), Sec. 2.2.3.

[11]For a discussion of similar results in a slightly different context see: J. H. Eberly, M. L. Report 940, Hansen Laboratories of Physics, 1962 (unpublished); and Ph. D. thesis (Stanford University, 1962) (unpublished); Second Rochester Conference on Coherence and Quantum Optics, 1966 (unpublished).

[12]S. Bloom, J. Appl. Phys. 27, 785 (1956); 28, 800 (1957).

[13]See, for example, N. Bloembergen and R. V. Pound, Phys. Rev. 95, 8 (1954).

[14]See, for example, W. L. Lama, R. Jodoin, and L. Mandel, Am. J. Phys. (to be published).

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Photoprotonic Effect in Hydrides*

I. L. Thomas
Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830
(Received 5 October 1971)

The cross section for the photoejection of a proton from a hydride molecule is derived using the Born approximation for $h\nu \gg I$, and using the long-wavelength approximation for $h\nu \simeq I$, where ν is the frequency of the incident photon and I is the ionization potential of the proton. The cross section is negligible in the high-energy case. Near threshold, the probability for one photon being adsorbed in a path length of 1 cm is

$$\tau = 1.4\times 10^{2}\,\frac{\rho N^{1/2}}{Z^{2}M}\left(\frac{\nu}{b\nu_0}\right)\left(\frac{b\nu_0}{\nu-\nu_c+(b-1)\nu_0}\right)^{(6N+5)/3} f_N^2 ,$$

where ρ is the density, M is the molecular weight, Z is the orbital exponent of the Slater function describing the proton, b is a number between 1 and 2, N is the principal quantum number of the proton, $h\nu_0 = I$, and $h\nu_c$ is the kinetic energy of the center of mass plus the kinetic energy of the relative coordinate in the direction of the motion of the center of mass. The coefficient f_N oscillates as N increases. For the range in N considered (88–125), f_N varies from a minimum of 7.793 at $N=103$ to a maximum of 175.4 at $N=115$ beyond which it decreases again. Since N is large, τ is large only near threshold.

I. INTRODUCTION

The ionization potential of a proton is the energy required to remove a proton with zero kinetic energy from a molecule. This ionization potential can be calculated by subtracting the energy of the molecule from that of the negative ion which is formed when a proton is removed. For example, McLean and Yoshimine[1] got −100.0704 hartree for the ground state of HF, and Clementi[2] got −99.4594 hartree for the ground state of F^-. Therefore, the protonic ionization potential in HF is 0.6210 hartree or 16.8 eV, which is in fair agreement with the experimental appearance potential of 15.9 eV.[3]

Another way to estimate the protonic ionization potential is to use a two-particle model for HF. The energy levels in this case are

$$E_N = -\frac{\hbar^2 z^2}{2\mu N^2 A_0^2} = -\frac{\hbar^2 Z^2}{2\mu a_0^2}\ (\text{cgs})$$

$$= -\frac{Z^2}{2\mu}\ (\text{hartree}) , \qquad (1)$$

where z is the effective charge attracting the proton, μ is the reduced protonic mass, $A_0 = \hbar^2/(\mu\epsilon^2)$, where $-\epsilon$ is the electronic charge, and a_0 is the Bohr radius. The ionization potential would equal the ground-state energy. This model was used to discuss the vibrational energy levels in HF where 1.067 hartree was found for the ground-state energy.[4] However, the value of z was derived from the orbital exponent of the Slater function which described the proton, and Slater has shown that the effective z of the wave function is larger than the effective z which should be used in the energy expression if the central-field model is used.[5] Therefore, the true ionization potential is given by

$$I = -E_N/b , \qquad (2)$$

where b is a number between 1 and 2.

Consider a monochromatic plane wave traveling in the z direction and polarized in the x direction incident on a hydride molecule. If $h\nu \geq I$, a proton may be excited to the continuum leaving a proton and a negative ion. The energy relation is