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Atomic coherence decay due to interactions with strong coherent radiation

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The time development of N two-level atoms linearly coupled to a monochromatic, long-wavelength radiation field is studied. Both systems are assumed to be initially in coherent states, the radiation mode being strongly populated with an average number n of photons larger than N. The decay of atomic coherence can be followed up to a time t^*, after which the perturbation expansion in powers of $n^{-1/2}$ used to solve the problem breaks down. We show that it increases proportional to t^2 at large times, as measured from the variances of the components of the total angular momentum of the atoms. The results are briefly discussed in connection with the problem of conversion of incoherent into coherent radiation.

I. INTRODUCTION

The present paper is the third of a series[1,2] in which we report the results of investigations on the coherence properties of the interaction between monochromatic electromagnetic fields and atomic two-level systems. The aim of these investigations is to understand the influence of the quantum-mechanical nature of both field and atoms on the dynamical development of the coupled system in typically macroscopic situations, that is, when the number of photons in the field and the number of atoms are large although finite. In order to evidence these quantum-mechanical effects, situations have been considered where the field is initially coherent in the sense defined by Glauber,[3] and the N two-level systems are coherent in the sense defined by Radcliffe[4] so that the coupled system can be considered to be as classical as possible at $t=0$, and deviations from coherence at relatively large times have been found to develop gradually. In particular we have previously shown the following:

(i) When the average number of photons n at $t=0$ is much smaller than N, the two-level atoms being initially in their ground states,[1] the initial coherence of the coupled system tends to disappear after the energy has been exchanged between atoms and field a number of times approximately given by N/n.

(ii) At the other end of the scale, i.e., for $n \gg N$, a similar loss of coherence takes place in the field at large times[2] the variance of the field amplitude increasing from 0 to ever larger values. This increase can be followed by our techniques up to times such that the total spin system (which we use to describe the two-level atoms) has exchanged energy with the field about $(n/N)^{1/2}$ times.

This behavior sharply contrasts with that of linearly coupled harmonic oscillators, which have been shown by Glauber[5] to conserve rigorously at all times the initial coherence properties. The behavior also seems to indicate that for times large enough the classical limit is not attainable for finite systems of atoms interacting with a radiation field, however large one takes the numbers of photons and atoms in the system. Moreover, this might also put some fundamental limits of a quantum-mechanical nature on the efficiency of ideal machines which transform incoherent into coherent radiation by exploiting a set of few-level objects in a maserlike or laserlike fashion.[6] Previous work by other authors on coherence decay from initially coherent states in the radiation-matter interaction generally falls into two categories. Either few-photon processes are considered; this amounts to being able to follow the system only for short times,[7] or few isolated atoms are treated as interacting with the radiation field,[8] which tends to make dubious the extrapolation of the conclusions to the more realistic many-atom case (in fact, we shall see that for large times the behavior of many atoms is qualitatively different from that of an isolated atom).

In view of these facts it has seemed worthwhile to extend the investigations to cover the statistical properties of the *atomic* system in the case of $n \gg N$. In this sense the present paper should be considered a completion of the work[2] described in (ii). The resonant Dicke Hamiltonian[9] for long-wavelength monochromatic radiation and in the rotating-wave approximation (RWA) is our usual starting point:

$$\mathcal{H} = \mathcal{H}_0 + V ;$$
$$\mathcal{H}_0 = \omega(S_z + \alpha^\dagger \alpha) ; \quad V = \tfrac{1}{2}\epsilon (S_+\alpha + S_-\alpha^\dagger) , \qquad (1.1)$$

where we have used collective spin operators

$$S_i = \sum_{l=1}^{N} S_i^l \quad (i = +, -, z)$$

in a subspace with maximum cooperation number $S=\frac{1}{2}N$, S_i^l being the single-atom spin operators represented by 2×2 Pauli matrices. The state of the field at $t=0$ is assumed to be a Glauber coherent state

$$|a\rangle = D|0\rangle = \exp(a\alpha^\dagger - a^*\alpha)|0\rangle ,$$

with $a=\sqrt{n}\,e^{-i\varphi}$, $n=|a|^2$ being the average photon number at $t=0$. We take the atomic system at $t=0$ to be $|-S\rangle$, such that

$$S_-|-S\rangle = 0 ; \quad S_z|-S\rangle = -S|-S\rangle .$$

This is the lowest possible in energy among the Radcliffe states, and has the important properties of being both coherent and an eigenstate of $\mathcal{H}_0$. We indicate the state of the total system at $t=0$ by $|a, -S\rangle$, and the same state at time t becomes

$$|\psi\rangle = e^{-i\mathcal{H}t}D|0,-S\rangle = e^{-i\mathcal{H}_0 t}e^{-iVt}D|0,-S\rangle , \tag{1.2}$$

since $[\mathcal{H}_0, V]=0$. Introducing the rotation operator

$$R_z = e^{-i\omega t S_z} ,$$

enables us to put the free-field evolution operator in the form

$$e^{-i\mathcal{H}_0 t} = e^{-i\omega t \alpha^\dagger\alpha}R_z , \tag{1.3}$$

while using the unitary properties of D we can write

$$e^{-iVt}D = DD^{-1}\sum_{m=0}^{\infty}\frac{1}{m!}(-iVt)^m D = D\exp(-iD^{-1}VDt) . \tag{1.4}$$

Moreover,

$$D^{-1}VD = \tfrac{1}{2}\epsilon[(S_+a + S_-a^*) + (S_+\alpha + S_-\alpha^\dagger)]$$

can be used in (1.4), so that (1.2) can be cast in the form

$$|\psi\rangle = e^{-i\omega t\alpha^\dagger\alpha}DR_z\, e^{-(i/2)\tau(X+Y/\sqrt{n})}|0,-S\rangle , \tag{1.5}$$

where

$$X = e^{-i\varphi}S_+ + e^{i\varphi}S_- ; \quad Y = \alpha S_+ + \alpha^\dagger S_- ; \quad \tau = \epsilon\sqrt{n}\,t . \tag{1.6}$$

The aim of the present work is to investigate the statistical properties of the atomic system and their time development, deducing them by suitable approximations to state (1.5). In Sec. II we shall discuss the techniques necessary to obtain these approximations, which shall be used to calculate the average values of the total angular momentum and their variances in Secs. III and IV in two different frames of reference. We shall finally discuss the results obtained together with their limits of validity in Sec. V.

II. GENERAL THEORY

The advantage of putting the state of the system in the form (1.5) is that in this form the "classical" features of the rotating field are represented by X, while the "quantum" fluctuations are included in Y. The effects of these two operators on the dynamics of the system, however, are not immediately separable in (1.5), since $[X,Y]\neq 0$ and

$$e^{-(i/2)\tau(X+Y/\sqrt{n})} \neq e^{-(i/2)\tau X}e^{-(i/2)\tau Y/\sqrt{n}} .$$

On the other hand, it is easily seen from (1.5) that for large n the effects of the quantum fluctuations should be small, at least for finite τ. Hence it is plausible that for finite τ, and n large enough, one can write in zero order

$$|\psi\rangle \simeq e^{-i\mathcal{H}_0 t}De^{-(i/2)\tau X}|0,-S\rangle ,$$

which is equivalent to neglecting terms $O(n^{-1/2})$ in the exponent of (1.5). Furthermore, in this approximation,

$$e^{-i\mathcal{H}_0 t}D \simeq e^{-i\omega t n}R_z D .$$

Furthermore, we find the well-known classical result of the spin system rotating rigidly and without loss of coherence with frequency $\epsilon\sqrt{n}$ about an axis on the (x,y) plane making an angle φ with the x axis at $t=0$, which in turn rotates about the z axis at frequency ω. Moreover, in this approximation the average number of photons does not change with time, the field remaining in a coherent state with its phase changing with angular velocity ω, due to its rotation about the z axis. In what follows we shall put $\varphi=0$ which amounts to fixing the initial phase of the rotating field as coincident with the x axis at $t=0$.

The above discussion leads us to consider expansions in powers of $n^{-1/2}$ of the exponential in (1.5) to take into account the effects of the quantum fluctuations as successive approximations to the exact expression. In order to obtain such an expansion, we put

$$\exp[-(i/2)\tau(X+Y/\sqrt{n})] \equiv \phi(\tau) ,$$

and observe that ϕ is the formal solution of operator equation

$$i\frac{d}{d\tau}\phi = \frac{1}{2}\left(X+\frac{Y}{\sqrt{n}}\right)\phi ; \quad \phi(0)=1 . \tag{2.1}$$

Equation (2.1) on the other hand is formally identical to the equation for the Schrödinger time-development operator of a system described by a Hamiltonian

$$H = \tfrac{1}{2}X + Y/\sqrt{n}$$

and with a scaled time τ. For n large enough, $\frac{1}{2}X$ can be considered as the unperturbed Hamiltonian, $Y/2\sqrt{n}$ being the perturbation. The well-known expansions familiar from time-dependent perturbation theory[10] should thus be applicable to our case. We proceed first by introducing the "interaction representation"

$$\phi' = e^{(i/2)\tau X}\phi\,; \quad Y' = e^{(i/2)\tau X} Y e^{-(i/2)\tau X}\,, \tag{2.2}$$

which transforms (2.1) into

$$i\frac{d}{d\tau}\phi' = \frac{1}{2\sqrt{n}}Y'\phi'\,; \quad \phi'(0)=1\,. \tag{2.3}$$

The last equation is then transformed into the integral equation

$$\phi'(\tau) = 1 + \frac{1}{i2\sqrt{n}}\int_0^\tau Y'(\tau_1)\phi'(\tau_1)\,d\tau_1\,,$$

which can be iterated to yield

$$\phi'(\tau) = 1 + \frac{1}{i\,2\sqrt{n}}\int_0^\tau Y'(\tau_1)\,d\tau_1 + \left(\frac{1}{i2\sqrt{n}}\right)^2\int_0^\tau\int_0^{\tau_1} Y'(\tau_1)Y'(\tau_2)\,d\tau_1\,d\tau_2 + \cdots = 1 + \phi_1' + \phi_2' + \cdots\,,$$

where

$$\phi_s'(\tau) = \left(\frac{1}{i2\sqrt{n}}\right)^s \int_0^\tau\int_0^{\tau_1}\cdots\int_0^{\tau_{s-1}} Y'(\tau_1)Y'(\tau_2)\cdots Y'(\tau_s)\,d\tau_1\,d\tau_2\cdots d\tau_s \tag{2.4}$$

is the term $O(n^{-s/2})$. We thus obtain

$$\phi = e^{-(i/2)\tau X}\left(1 + \sum_{s=1}^{\infty}\phi_s'\right).$$

In order to calculate ϕ_s' from (2.4) we need Y' from (2.2). Since

$$e^{(i/2)\tau X}S_- e^{-(i/2)\tau X} = S_-\cos^2\tfrac{1}{2}\tau + S_+\sin^2\tfrac{1}{2}\tau + iS_z\sin\tau\,, \quad e^{(i/2)\tau X}S_z e^{-(i/2)\tau X} = S_z\cos\tau + \tfrac{1}{2}i(S_- - S_+)\sin\tau\,, \tag{2.5}$$

we find after some straightforward algebra

$$Y'(\tau) = (\alpha S_+ + \alpha^\dagger S_-) + \tfrac{1}{2}[(\alpha^\dagger - \alpha)(S_- - S_+)](\cos\tau - 1) + i(\alpha^\dagger - \alpha)S_z\sin\tau\,. \tag{2.6}$$

Substituting (2.6) in (2.4) with $s=1$ we find immediately, after an elementary integration

$$\phi_1' = (1/2\sqrt{n})\{-(\alpha^\dagger - \alpha)S_z(\cos\tau - 1) - i[(\alpha S_+ + \alpha^\dagger S_-)\tau + \tfrac{1}{2}(S_- - S_+)(\alpha^\dagger - \alpha)(\sin\tau - \tau)]\}\,. \tag{2.7}$$

It is convenient to calculate the second-order approximation as

$$\phi_2' = \frac{1}{i2\sqrt{n}}\int_0^\tau Y'(\tau_1)\phi_1'(\tau_1)\,d\tau_1\,. \tag{2.8}$$

The procedure is conceptually simple but rather tedious, and here we only report the final result

$$\begin{aligned}\phi_2' = (1/4n)[&i(\alpha S_+ + \alpha^\dagger S_-)(\alpha^\dagger - \alpha)S_z I_1 + (\alpha S_+ + \alpha^\dagger S_-)^2 I_2 + \tfrac{1}{2}(\alpha S_+ + \alpha^\dagger S_-)(\alpha^\dagger - \alpha)(S_- - S_+)I_3\\ &+ \tfrac{1}{2}i(\alpha^\dagger - \alpha)^2(S_- - S_+)S_z I_4 + \tfrac{1}{2}(\alpha^\dagger - \alpha)(S_- - S_+)(\alpha S_+ + \alpha^\dagger S_-)I_5 + \tfrac{1}{4}(\alpha^\dagger - \alpha)^2(S_- - S_+)^2 I_6 + (\alpha^\dagger - \alpha)^2 S_z^2 I_7\\ &+ i(\alpha^\dagger - \alpha)S_z(\alpha S_+ + \alpha^\dagger S_-)I_8 + \tfrac{1}{2}i(\alpha^\dagger - \alpha)^2 S_z(S_- - S_+)I_9]\,,\end{aligned} \tag{2.9}$$

where

$$\begin{aligned}&I_1 = \sin\tau - \tau\,;\quad I_2 = -\tfrac{1}{2}\tau^2\,;\quad I_3 = \cos\tau - 1 + \tfrac{1}{2}\tau^2\,;\quad I_4 = \tfrac{1}{2}\sin\tau\cos\tau - 2\sin\tau + \tfrac{3}{2}\tau\,;\quad I_5 = -\tau\sin\tau - (\cos\tau - 1) + \tfrac{1}{2}\tau^2\,;\\ &I_6 = -\tfrac{1}{2}(\sin\tau - \tau)^2\,;\quad I_7 = \tfrac{1}{2}(\cos\tau - 1)^2\,;\quad I_8 = \tau\cos\tau - \sin\tau\,;\quad I_9 = \tfrac{1}{2}\sin\tau\cos\tau - \tau\cos\tau + \sin\tau - \tfrac{1}{2}\tau\,.\end{aligned} \tag{2.10}$$

We have thus explicitly obtained the expression for ϕ up to second-order terms in $n^{-1/2}$, which is as far as we wish to push our calculations. Hence the first-order approximation to the average value of an operator A is given by

$$\langle A\rangle = \langle 0,-S|(1+\phi_1'^\dagger)e^{(i/2)\tau X}D^{-1}e^{i\mathcal{H}_0 t}Ae^{-i\mathcal{H}_0 t}De^{-(i/2)\tau X}(1+\phi_1')|0,-S\rangle = \langle 0,-S|(\tilde{A}' + \phi_1'^\dagger\tilde{A}' + \tilde{A}'\phi_1')|0,-S\rangle\,, \tag{2.11}$$

where

$$\begin{aligned}\tilde{A} &= D^{-1}e^{i\mathcal{H}_0 t}Ae^{-i\mathcal{H}_0 t}D\\ &= R_z^{-1}D^{-1}e^{i\omega t\alpha^\dagger\alpha}Ae^{-i\omega t\alpha^\dagger\alpha}DR_z\,.\end{aligned} \tag{2.12}$$

The second-order approximation is given by

$$\begin{aligned}\langle A\rangle &= \langle 0,-S|(1+\phi_1'^\dagger + \phi_2'^\dagger)\tilde{A}'(1+\phi_1' + \phi_2')|0,-S\rangle\\ &= \langle 0,-S|(\tilde{A}' + \phi_1'^\dagger\tilde{A}' + \tilde{A}'\phi_1' + \phi_2'^\dagger\tilde{A}'\\ &\qquad + \tilde{A}'\phi_2' + \phi_1'^\dagger\tilde{A}'\phi_1')|0,-S\rangle\,.\end{aligned} \tag{2.13}$$

Expressions (2.12) and (2.13) give the average values of an operator in the laboratory frame of reference; we shall find it more convenient to calculate first the corresponding values in a frame which rotates with angular velocity $\epsilon\sqrt{n}$ about the direction of the rotating electromagnetic (em) field. We call this the doubly rotating frame of reference, in which the operators are averaged over the state

$$|\psi_R\rangle = e^{(i/2)\tau X}R_z^{-1}|\psi\rangle = e^{-i\omega t\alpha^\dagger\alpha}D\phi'|0,-S\rangle,$$

as

$$\langle A\rangle_R = \langle 0,-S|(\tilde{A}'' + \phi_1'^\dagger\tilde{A}'' + \tilde{A}''\phi_1')|0,-S\rangle \quad (2.14)$$

in the first-order approximation, and as

$$\langle A\rangle_R = \langle 0,-S|(\tilde{A}'' + \phi_1'^\dagger\tilde{A}'' + \tilde{A}''\phi_1' + \phi_2'^\dagger\tilde{A}'' + \tilde{A}''\phi_2' + \phi_1'^\dagger\tilde{A}''\phi_1')|0,-S\rangle, \quad (2.15)$$

in the second-order approximation, where

$$\tilde{A}'' = R_z e^{-(i/2)\tau X}\tilde{A}' e^{(i/2)\tau X}R_z^{-1} = D^{-1}e^{i\omega t\alpha^\dagger\alpha}Ae^{-i\omega t\alpha^\dagger\alpha}D. \quad (2.16)$$

We remark that up until now the formalism developed is valid also for any initial-spin state, so that the same formulas would apply to any state $|a,\mu\rangle$ at $t=0$, with the only substitution of $|0,\mu\rangle$ instead of $|0,-S\rangle$. From now on, however, we shall exploit the simplicity of the initial state of our choice, which permits us a noticeable reduction of mathematical labor. We recall that

$$\begin{aligned}&\alpha|0,-S\rangle = 0;\quad S_-|0,-S\rangle = 0;\\ &S_z|0,-S\rangle = -S|0,-S\rangle;\quad \alpha\alpha^\dagger|0,-S\rangle = |0,-S\rangle;\\ &S_-S_+|0,-S\rangle = 2S|0,-S\rangle;\\ &S_zS_+|0,-S\rangle = (1-S)S_+|0,-S\rangle,\end{aligned} \quad (2.17)$$

so that

$$\begin{aligned}&(\alpha S_+ + \alpha^\dagger S_-)|0,-S\rangle = 0;\\ &(\alpha^\dagger-\alpha)S_z|0,-S\rangle = -S\alpha^\dagger|0,-S\rangle;\\ &(\alpha^\dagger-\alpha)(S_- - S_+)|0,-S\rangle = -\alpha^\dagger S_+|0,-S\rangle.\end{aligned} \quad (2.18)$$

From (2.18) and (2.7) we find

$$\phi_1'|0,-S\rangle = (1/2\sqrt{n})\{[S(\cos\tau-1) + \tfrac{1}{2}iS_+(\sin\tau-\tau)]\alpha^\dagger\}|0,-S\rangle. \quad (2.19)$$

Moreover, on the basis of (2.17)

$$\begin{aligned}&(\alpha S_+ + \alpha^\dagger S_-)(\alpha^\dagger-\alpha)S_z|0,-S\rangle = -SS_+|0,-S\rangle,\\ &(\alpha S_+ + \alpha^\dagger S_-)(\alpha^\dagger-\alpha)(S_- - S_+)|0,-S\rangle = -(S_+^2 + 2S\alpha^{\dagger 2})|0,-S\rangle,\\ &(\alpha^\dagger-\alpha)^2(S_- - S_+)S_z|0,-S\rangle = S(\alpha^{\dagger 2}-1)S_+|0,-S\rangle,\\ &(\alpha^\dagger-\alpha)^2(S_- - S_+)^2|0,-S\rangle = (\alpha^{\dagger 2}-1)(S_+^2-2S)|0,-S\rangle,\\ &(\alpha^\dagger-\alpha)^2S_z^2|0,-S\rangle = S^2(\alpha^{\dagger 2}-1)|0,-S\rangle,\\ &(\alpha^\dagger-\alpha)^2S_z(S_- - S_+)|0,-S\rangle = (S-1)(\alpha^{\dagger 2}-1)S_+|0,-S\rangle,\end{aligned} \quad (2.20)$$

which together with (2.9) yields

$$\phi_2'|0,-S\rangle = (1/4n)\{(\tfrac{1}{2}I_6S - I_7S^2) - i[I_1S + \tfrac{1}{2}I_4S + \tfrac{1}{2}I_9(S-1)]S_+ + \tfrac{1}{2}i[I_4S + I_9(S-1)]S_+\alpha^{\dagger 2} - (I_3S + \tfrac{1}{2}I_6S - I_7S^2)\alpha^{\dagger 2} - \tfrac{1}{2}(I_3 + \tfrac{1}{2}I_6)S_+^2 + \tfrac{1}{4}I_6S_+^2\alpha^{\dagger 2}\}|0,-S\rangle. \quad (2.21)$$

We shall now proceed to calculate the average values of the dynamical quantities of the system relevant to its statistical properties.

III. DOUBLY ROTATING FRAME

We first calculate $\langle S_z\rangle_R$. From (2.16) we immediately have $\tilde{S}_z'' = S_z$ so that from (2.15),

$$\langle S_z+S\rangle_R = \langle 0,-S|[(S_z+S) + \phi_1'^\dagger(S_z+S) + (S_z+S)\phi_1' + \phi_1'^\dagger(S_z+S)\phi_1' + \phi_2'^\dagger(S_z+S) + (S_z+S)\phi_2']|0,-S\rangle. \quad (3.1)$$

Since

$$(S_z+S)|0,-S\rangle = 0,$$

we immediately find from (2.19),

$$\langle S_z+S\rangle_{R0} = 0;\quad \langle S_z+S\rangle_{R1} = 0;$$

$$\begin{aligned}\langle S_z+S\rangle_{R2} &= \langle 0,-S|\phi_1'^\dagger(S_z+S)\phi_1'|0,-S\rangle\\ &= (1/4n)\langle 0,-S|[S(\cos\tau-1) - \tfrac{1}{2}iS_-(\sin\tau-\tau)]\alpha(S_z+S)\\ &\quad\times[S(\cos\tau-1) + \tfrac{1}{2}iS_+(\sin\tau-\tau)]\alpha^\dagger|0,-S\rangle,\end{aligned}$$

where by $\langle A\rangle_{Ri}$ we have indicated the terms in $\langle A\rangle_R$ of the ith order in $n^{-1/2}$. After some fairly simple algebra the above expression can be put into the form

$$\langle S_z+S\rangle_{R2} = (S/8n)(\sin\tau-\tau)^2$$

and

$$\langle S_z\rangle_R = -S[1-(1/8n)(\sin\tau-\tau)^2]. \tag{3.2}$$

We also calculate $\langle S_+\rangle_R$, for which we also have $\tilde{S}_+'' = S_+$ and

$$\begin{aligned}\langle S_+\rangle_{R0} &= \langle 0,-S|S_+|0,-S\rangle = 0;\\ \langle S_+\rangle_{R1} &= \langle 0,-S|(\phi_1'^\dagger S_+ + S_+\phi_1')|0,-S\rangle;\\ \langle S_+\rangle_{R2} &= \langle 0,-S|(\phi_1'^\dagger S_+\phi_1' + \phi_2'^\dagger S_+ + S_+\phi_2')|0,-S\rangle\\ &= \langle 0,-S|(\phi_1'^\dagger S_+\phi_1' + \phi_2'^\dagger S_+)|0,-S\rangle.\end{aligned} \tag{3.3}$$

From (2.19) and (2.21) we easily find

$$\langle 0,-S|\phi_1'^\dagger S_+\phi_1'|0,-S\rangle = -(i/4n)S^2(\sin\tau-\tau)(\cos\tau-1);$$

$$\langle 0,-S|\phi_2'^\dagger S_+|0,-S\rangle$$
$$= (i/4n)[2S^2(\sin\tau-\tau)+S^2(\sin\tau-\tau)(\cos\tau-1)-SI_9],$$

which after substitution in (3.3) yield

$$\begin{aligned}\langle S_+\rangle_{R2} &= \langle S_+\rangle_R\\ &= (i/4n)[2S^2(\sin\tau-\tau)\\ &\quad -S(\tfrac{1}{2}\sin\tau\cos\tau-\tau\cos\tau+\sin\tau-\tfrac{1}{2}\tau)].\end{aligned} \tag{3.4}$$

From (3.4) we deduce immediately

$$\begin{aligned}\langle S_x\rangle_R &= \mathrm{Re}(\langle S_+\rangle_R) = 0;\\ \langle S_Y\rangle_R &= \mathrm{Im}(\langle S_+\rangle_R)\\ &= (1/4n)[2S^2(\sin\tau-\tau)-S(\tfrac{1}{2}\sin\tau\cos\tau-\tau\cos\tau\\ &\quad +\sin\tau-\tfrac{1}{2}\tau)].\end{aligned} \tag{3.5}$$

Expressions (3.2) and (3.5) allow us to make immediately some interesting considerations on the dynamics of the system. We remark that in the classical approximation the total S should remain fixed along z in the doubly rotating frame, while from (3.5) and (3.2) we see that the total angular momentum develops a y component, the x one always remaining zero and the z one decreasing gradually. This means that S tends to lag in phase and rotates about the direction of the electromagnetic field at an angular velocity smaller than $\epsilon\sqrt{n}$. We shall call this effect also "residual rotation." We can try to explain this qualitatively by taking into account the absorption and emission processes by the atomic system, which on the average decrease the mean number of photons from n to $n-S$, thereby reducing the angular velocity of S from $\epsilon\sqrt{n}$ to

$$\epsilon(n-S)^{1/2} \sim \epsilon\sqrt{n}(1-S/2n)$$

on the average. This means that the spins should lag by an average angle

$$\delta = -S\tau/2n. \tag{3.6}$$

This simple argument would yield for the z and y components

$$\begin{aligned}\langle S_z\rangle_R &\simeq -S\cos\delta \sim -S(1-\tfrac{1}{2}\delta^2)\\ &= -S(1-S^2\tau^2/8n^2),\end{aligned} \tag{3.7}$$

$$\langle S_Y\rangle_R \simeq S\sin\delta \sim S\delta = -S^2\tau/2n. \tag{3.8}$$

While (3.8) agrees with part of (3.5) for large enough τ, (3.7) is in sharp disagreement with (3.2) which indicates a different decrease than that predicted by (3.7); the latter is in fact $O(n^{-2})$. Therefore the phase-lag of S cannot be the whole story. In fact (3.2) and part of (3.5) must be explained in terms of loss of coherence within the atomic system. This can be shown to be the case by calculating $\langle \vec{S}\rangle^2$ which should give S^2 for a coherent state. In our case we have from (3.5) and (3.2),

$$\langle S_x\rangle^2 + \langle S_y\rangle^2 + \langle S_z\rangle^2 \sim S^2(1-\tau^2/4n),$$

where we have neglected terms $O(n^{-2})$, and where we have assumed $\tau \gg 1$. This shows that the coherence of the atomic system is in fact decreasing.

In order to see more clearly the growing up of disorder within the atomic system, we now turn to calculate the variances of the angular momentum components. First we calculate

$$\begin{aligned}\langle S_z^2-S^2\rangle_{R0} &= \langle 0,-S|(S_z^2-S^2)|0,-S\rangle;\\ \langle S_z^2-S^2\rangle_{R1} &= \langle 0,-S|[\phi_1'^\dagger(S_z^2-S^2)+(S_z^2-S^2)\phi_1']|0,-S\rangle;\\ \langle S_z^2-S^2\rangle_{R2} &= \langle 0,-S|[\phi_1'^\dagger(S_z^2-S^2)\phi_1'+\phi_2'^\dagger(S_z^2-S^2)\\ &\quad +(S_z^2-S^2)\phi_2']|0,-S\rangle\\ &= \langle 0,-S|\phi_1'^\dagger(S_z^2-S^2)\phi_1'|0,-S\rangle,\end{aligned} \tag{3.9}$$

since

$$(S_z^2-S^2)|0,-S\rangle = 0.$$

Moreover using (2.19) in (3.9) we get after some algebra

$$\langle S_z^2-S^2\rangle_{R2} = (1/4n)\tfrac{1}{2}S(1-2S)(\sin\tau-\tau)^2,$$

which gives

$$\langle S_z^2\rangle_R = S^2 + (1/4n)\tfrac{1}{2}S(1-2S)(\sin\tau-\tau)^2. \tag{3.10}$$

On the other hand from (3.2) we have

$$\langle S_z\rangle_R^2 = S^2[1-(1/4n)(\sin\tau-\tau)^2],$$

from which we can find the variance

$$(\Delta S_z)_R^2 = \langle S_z^2\rangle_R - \langle S_z\rangle_R^2 = (S/8n)(\sin\tau-\tau)^2. \tag{3.11}$$

As for the variance of the transverse components. we remark that the variance of S_- is given by the length of the vector

$$(S_- - \langle S_-\rangle_R)|\psi_R\rangle,$$

so that

$$(\Delta S_-)_R^2 = \langle\psi_R|S_+S_-|\psi_R\rangle - |\langle\psi_R|S_-|\psi_R\rangle|^2. \tag{3.12}$$

Following the usual procedure we calculate

$$\langle S_+S_-\rangle_{R0}=\langle 0,-S\,|\,S_+S_-\,|\,0,-S\rangle=0;$$
$$\langle S_+S_-\rangle_{R1}=\langle 0,-S\,|\,(\phi_1'^\dagger S_+S_-+S_+S_-\phi_1')\,|\,0,-S\rangle=0;$$
$$\begin{aligned}\langle S_+S_-\rangle_{R2}&=\langle 0,-S\,|\,(\phi_1'^\dagger S_+S_-\phi_1'+\phi_2'^\dagger S_+S_-+S_+S_-\phi_2')\,|\,0,-S\rangle\\&=\langle 0,-S\,|\,\phi_1'^\dagger S_+S_-\phi_1'\,|\,0,-S\rangle\\&=(S^2/4n)(\sin\tau-\tau)^2.\end{aligned}\tag{3.13}$$

Further, from (3.4) we find that $|\langle S_-\rangle_R|^2$ is of $O(n^{-2})$ and we must neglect it in (3.12). Consequently we find

$$(\Delta S_-)_R^2=(S^2/4n)(\sin\tau-\tau)^2.\tag{3.14}$$

Comparing (3.11) and (3.14) with (A5) and (A7) in the Appendix, we see that the variances in the doubly rotating frame cannot be attributed to the residual rotation of S. In fact for large τ we have from (3.11) and (3.14), respectively,

$$(\Delta S_z)_R^2\sim S\tau^2/8n,\quad (\Delta S_-)_R^2\sim S^2\tau^2/4n,\tag{3.15}$$

while the variances due to the residual rotation can be approximately calculated from (A5) and (A7) with $\theta=\delta$ given by (3.6) as

$$(\Delta S_z)_R^2\sim S^3\tau^2/8n^2,\quad (\Delta S_-)_R^2\sim S^5\tau^4/2^7n^4,$$

which are much smaller than (3.15). Thus the increase in the variances is intrinsically quantum mechanical and should imply an increasing loss of coherence within the atomic system.

IV. LABORATORY FRAME

In this section we report the main results of our theory in the laboratory frame of reference. For each quantity we have first to calculate

$$\tilde A'=e^{(i/2)\tau X}\tilde A e^{-(i/2)\tau X},$$

$\tilde A$ being given by (2.12). Thus we obtain

$$\begin{aligned}\tilde S_z'&=e^{(i/2)\tau X}R_z^{-1}D^{-1}e^{i\omega\tau\alpha^\dagger\alpha}S_z e^{-i\omega\tau\alpha^\dagger\alpha}DR_z e^{-(i/2)\tau X}\\&=e^{(i/2)\tau X}R_z^{-1}S_zR_z e^{-(i/2)\tau X}=e^{(i/2)\tau X}S_z e^{-(i/2)\tau X}\\&=S_z\cos\tau+\tfrac{1}{2}i(S_--S_+)\sin\tau,\end{aligned}\tag{4.1}$$

where use has been made of (2.5). Analogously we find

$$\begin{aligned}\tilde S_+'&=e^{i\omega\tau}(S_+\cos^2\tfrac{1}{2}\tau+S_-\sin^2\tfrac{1}{2}\tau-iS_z\sin\tau),\\\tilde S_-'&=e^{-i\omega\tau}(S_-\cos^2\tfrac{1}{2}\tau+S_+\sin^2\tfrac{1}{2}\tau+iS_z\sin\tau).\end{aligned}\tag{4.2}$$

For the longitudinal component of the total angular momentum one has from (4.1), (2.19), and (2.21) after some algebraic labor

$$\begin{aligned}\langle S_z\rangle_0&=\langle 0,-S\,|\,\tilde S_z'\,|\,0,-S\rangle\\&=\langle 0,-S\,|\,[S_z\cos\tau+\tfrac{1}{2}i(S_--S_+)\sin\tau]\,|\,0,-S\rangle\\&=-S\cos\tau;\\\langle S_z\rangle_1&=\langle 0,-S\,|\,(\phi_1'^\dagger\tilde S_z'+\tilde S_z'\phi_1')\,|\,0,-S\rangle=0;\\\langle S_z\rangle_2&=\langle 0,-S\,|\,(\phi_1'^\dagger\tilde S_z'\phi_1'+\phi_2'^\dagger\tilde S_z'+\tilde S_z'\phi_2')\,|\,0,-S\rangle\\&=(1/4n)[\tfrac{1}{2}S\cos\tau(\sin\tau-\tau)^2\\&\quad+2S^2\sin\tau(\sin\tau-\tau)-S\sin\tau I_9].\end{aligned}\tag{4.3}$$

Summing up results (4.3) we finally get

$$\begin{aligned}\langle S_z\rangle&=-S\cos\tau[1-(1/8n)(\sin\tau-\tau)^2]\\&\quad+(1/2n)[S^2\sin\tau(\sin\tau-\tau)\\&\quad-\tfrac{1}{2}S\sin\tau(\tfrac{1}{2}\sin\tau\cos\tau\\&\quad-\tau\cos\tau+\sin\tau-\tfrac{1}{2}\tau)].\end{aligned}\tag{4.4}$$

The transverse components can be calculated from (4.2), (2.19), and (2.21) as

$$\begin{aligned}\langle S_+\rangle_0&=\langle 0,-S\,|\,\tilde S_+'\,|\,0,-S\rangle=iSe^{i\omega\tau}\sin\tau;\\\langle S_+\rangle_1&=\langle 0,-S\,|\,\phi_1'^\dagger\tilde S_+'+\tilde S_+'\phi_1')\,|\,0,-S\rangle=0;\\\langle S_+\rangle_2&=\langle 0,-S\,|\,(\phi_1'^\dagger\tilde S_+'\phi_1'+\phi_2'^\dagger\tilde S_+'+\tilde S_+'\phi_2')\,|\,0,-S\rangle\\&=(ie^{i\omega t}/4n)S[-\tfrac{1}{2}\sin\tau(\sin\tau-\tau)^2\\&\quad+2S\cos\tau(\sin\tau-\tau)\\&\quad-\cos\tau(\tfrac{1}{2}\sin\tau\cos\tau\\&\quad-\tau\cos\tau+\sin\tau-\tfrac{1}{2}\tau)],\end{aligned}\tag{4.5}$$

and summing up results (4.5) we get

$$\begin{aligned}\langle S_+\rangle&=ie^{i\omega t}S\{\sin\tau[1-\tfrac{1}{8}(\sin\tau-\tau)^2]\\&\quad+(1/4n)2S\cos\tau(\sin\tau-\tau)\\&\quad-\cos\tau(\tfrac{1}{2}\sin\tau\cos\tau\\&\quad-\tau\cos\tau+\sin\tau-\tfrac{1}{2}\tau)\}\end{aligned}\tag{4.6}$$

The x and y component of S are easily found from (4.6) as

$$\begin{aligned}\begin{bmatrix}\langle S_x\rangle\\\langle S_y\rangle\end{bmatrix}&=\begin{bmatrix}\mathrm{Re}\\\mathrm{Im}\end{bmatrix}\langle S_+\rangle\\&=\begin{bmatrix}-\sin\omega t\\\cos\omega t\end{bmatrix}\{S\sin\tau[1-(1/8n)(\sin\tau-\tau)^2]\\&\quad+(1/2n)[S^2\cos\tau(\sin\tau-\tau)-\tfrac{1}{2}S\cos\tau(\tfrac{1}{2}\sin\tau\cos\tau-\tau\cos\tau+\sin\tau-\tfrac{1}{2}\tau)]\}.\end{aligned}\tag{4.7}$$

We now wish to analyze in some detail the results hitherto obtained in the laboratory frame of reference. Comparing (3.2) to (4.4) we find again in the latter the term proportional to

$$(S/8n)(\sin\tau - \tau)^2,$$

which we have already ascribed to the shortening of the total angular momentum due to loss of coherence within the atomic system. Other terms of order S^2/n appear in (4.4), however, which are likely to become important for large S, and these are probably due to the phase-lag effect discussed in Sec. III. In fact, in the laboratory frame of reference we should have

$$\begin{aligned}\langle S_z\rangle &= -S(\cos\tau + \delta) \sim -S(\cos\tau - \delta\sin\tau)\\ &= -S\cos\tau - (S^2/2n)\tau\sin\tau,\end{aligned} \tag{4.8}$$

where only the phase-lag effect has been taken into account and where δ is defined in (3.6). Comparing (4.4) and (4.8) we see that the S^2 terms are indeed explainable in terms of the residual rotation. Moreover, it is easily seen that the time development of the transverse components in (4.7) is essentially of the same form as that of $\langle S_z\rangle$ in (4.4), apart from appropriate phase factors.

Finally we report here the average value of S_z^2 without giving the details of calculation

$$\begin{aligned}\langle S_z^2\rangle = {} & S^2\cos^2\tau + \tfrac{1}{2}S\sin^2\tau + (1/4n)S(2S-1)\\ & \times\{-\tfrac{1}{2}\cos^2\tau(\sin\tau - \tau)^2\\ & -\sin\tau\cos\tau[2S(\sin\tau-\tau) - \tfrac{1}{2}\sin\tau\cos\tau\\ & + \tau\cos\tau - \sin\tau + \tfrac{1}{2}\tau]\\ & + \sin^2\tau(\cos\tau - 1 + \tfrac{1}{2}\tau^2)\}.\end{aligned} \tag{4.9}$$

Expression (4.9) is useful to derive the variance of S_z. We shall not do this here, however, and shall be content with the variances already derived in the doubly rotating frame of reference.

V. DISCUSSION

We are now ready to discuss the results obtained in the preceding sections, starting from a discussion on the limits of validity of our perturbation expansion in powers of $n^{-1/2}$. We shall assume that when the perturbation terms in the time-dependent expressions for the quantities we have calculated become comparable with the "classical" terms, our perturbation procedure breaks down. We shall consider the case $\tau \gg 1$ in the laboratory frame and two cases for S:

(i) $S \sim 1$. Here $S^2 \simeq S$, and neglecting terms of order τ with respect to those of order τ^2, we get from (4.4) and (4.7)

$$\begin{aligned}\langle S_x\rangle &= -S\sin\omega t\sin\tau(1-\tau^2/8n),\\ \langle S_y\rangle &= -S\cos\omega t\sin\tau(1-\tau^2/8n),\\ \langle S_z\rangle &= -S\cos\tau(1-\tau^2/8n),\end{aligned} \tag{5.1}$$

which clearly shows that our perturbation breaks down for $\tau = \tau^* \sim (8n)^{1/2}$. This allows us to define a time

$$t^* = (8n)^{1/2}(\epsilon\sqrt{n})^{-1}, \tag{5.2}$$

after which we cannot follow the time development of our system. Since $\epsilon\sqrt{n}$ is the frequency of the precession of S about the rotating field, (5.2) means that by our techniques we can follow the motion of the atoms until the total angular momentum has performed $\sim\sqrt{n}$ precessions, which seems to be quite satisfactory.

(ii) $S \gg 1$. In this case we neglect S with respect to S^2 in (4.4) and (4.7), while keeping terms in $S^2\tau$ and $S\tau^2$. We thus obtain

$$\begin{aligned}\langle S_x\rangle &= -S\sin\omega t[(1-\tau^2/8n)\sin\tau + (\tau S/2n)\cos\tau],\\ \langle S_y\rangle &= S\cos\omega t[(1-\tau^2/8n)\sin\tau + (\tau S/2n)\cos\tau],\\ \langle S_z\rangle &= -S[\cos\tau(1-\tau^2/8n) - (\tau S/2n)\sin\tau].\end{aligned} \tag{5.3}$$

Keeping in mind our interpretation of the phase lag, it is convenient to rewrite (5.3) in the form, correct up to terms $O(n^{-1})$,

$$\begin{aligned}\langle S_x\rangle &= -S(1-\tau^2/8n)\sin\omega t\sin\Omega t,\\ \langle S_y\rangle &= S(1-\tau^2/8n)\cos\omega t\sin\Omega t,\\ \langle S_z\rangle &= -S(1-\tau^2/8n)\cos\Omega t,\end{aligned} \tag{5.4}$$

where

$$\Omega = \epsilon\sqrt{n}(1 - S/2n).$$

Expressions (5.4) describe a total spin precessing at ω about the static field along z, and at Ω about the rotating field. In this case the limits of validity of our approach are given by the more restrictive of the two conditions

$$\tau^2/8n \sim 1 \quad \text{or} \quad \tau S/2n \sim 1.$$

Consequently, depending on the total number of atoms, we can give two breakdown times

$$t^* = \begin{cases}(8n)^{1/2}(\epsilon\sqrt{n})^{-1}, & 1 < S < \sqrt{n},\\ (2n/S)(\epsilon\sqrt{n})^{-1}, & \sqrt{n} < S < n.\end{cases} \tag{5.5}$$

The first of which tells us that when the atoms are relatively few, the prevailing phenomenon is loss of coherence within the atomic system, which spreads off the total angular momentum before the phase-lag effect can perform its work. The opposite is true when t^* is given by the second of (5.5), which implies, if our interpretation in terms of residual rotation is correct, that when the number of atoms is large enough the phase lag of the total angular momentum becomes noticeable before the loss of coherence can play its role in invalidating our perturbation expansion.

We now turn to the variances, which we consider in the doubly rotated frame of reference, as given by (3.11) and (3.14). We observe that between the variances of the field[2] $(\Delta\alpha)^2$ and the variance of S_- the following relationship is valid:

$$(\Delta S_-)^2_R = 2S(\Delta\alpha)^2\,,$$

which is the same as found in previous work[1] with $n \ll S$. The above relationship illustrates the well-known[14] close connection between field and atom statistics. Further, inspection of asymptotic expressions (3.15) shows that the variance of the longitudinal component of S is smaller than those of the transverse components for large numbers of atoms, but the difference tends to vanish for few atoms ($S^2 \sim S$). We may now define on the basis of (3.15) physically meaningful critical times, as those times after which the spread of a component is of the same order as its possible maximum value. For the transverse component we have

$$\tau_s = 2\sqrt{n} \quad \text{or} \quad t_s = 2\sqrt{n}(\epsilon\sqrt{n})^{-1}\,, \tag{5.6}$$

which shows that after $2\sqrt{n}$ precessions around the rotating field the spread in the transverse component of the angular momentum has become very large. From

$$\Delta\alpha = \sqrt{n}\,,$$

we also find the analogous time for the photon field[2]

$$\tau_p = 2\sqrt{2}n/\sqrt{S} \quad \text{or} \quad t_p = (2\sqrt{2}n/\sqrt{S})(\epsilon\sqrt{n})^{-1}\,. \tag{5.7}$$

The ratio

$$t_s/t_p = (S/n)^{1/2}$$

tells us that the time of coherence loss for the spins is smaller than the corresponding time for the photon field, since $S < n$ is a basic assumption of this work. The question now arises: Are these times within reach of our perturbation theory, or, in other words, are t_s and t_p smaller than t^*? From (5.5), (5.6), and (5.7) we can distinguish two cases:

(i) $1 < S < \sqrt{n}$. Here we have

$$t^*/t_s \sim 1\,; \quad t^*/t_p \sim (S/n)^{1/2} < 1\,,$$

and we can follow pretty well the system up to the critical time for atoms, but not for the field.

(ii) $\sqrt{n} < S < n$. Then

$$t^*/t_s \sim \sqrt{n}/S < 1\,, \quad t^*/t_p = (25)^{-1/2} < 1\,,$$

in which case we can only follow the initial part of the decay of the total angular momentum's and of the photon field's coherent properties. It should be noted however that for $t \sim t^*$ the process of coherence loss in the system is already well developed.

It is also interesting to remark that our approximation (5.1) with $S = \frac{1}{2}$ coincides with the expansion for S_z up to $O(n^{-1})$ terms which has been obtained by Meystre *et al.*[8] for the decrease of the z component of an isolated spin in a coherent field. In this sense the first of (5.1) can be considered an extension to a large number of spins of their results, the latter being apparently valid for times larger than ours.

We now turn to the short-time behavior of the spin system, with particular reference to $\frac{1}{2}\pi$ and π pulses; in other words, we investigate the effects of short pulses of strong radiation, which in the so-called classical limit should rotate the angular momentum of the atoms by integer multiples of $\frac{1}{2}\pi$. The effects of π pulses in the laboratory frame of reference is limited essentially to a shortening of the total momentum such that an equivalent fraction $\pi^2/8n \sim 1/n$ of the total number of atoms has suffered quantum-mechanical disorder, as can be easily seen from (4.4); in fact all the phase-lag terms in (4.4) vanish for $\tau = \pi$. This is, in general, quite a negligible fraction, except perhaps if we consider the case of few atoms and few photons. As an example, if we have $n = 10$, about 12% of the atoms (N must be smaller than 10) have been disturbed, which means at most one atom in 8 or 9. The situation is quite different in the case of $\frac{1}{2}\pi$ pulses, where the S^2 terms in (4.4) can play an important role in determining the phase lag. If the latter is in fact ascribed only to the S^2 term, at the end of the pulse the decrease in S_z should amount to a fraction

$$(N/n)(\tfrac{1}{2}\pi - 1)$$

of the N atoms, which gives a phase lag

$$\varphi = \arctan\left[(N/n)(\tfrac{1}{2}\pi - 1)\right].$$

If, e.g., $n = 10N$, the phase lag after a $\frac{1}{2}\pi$ pulse should be over 3 deg.

In conclusion, we wish to stress the fact, clearly emerging from our theory, that every interaction between radiation and two-level objects involves some loss of coherence both in the atomic system and in the radiation field, which over a period of time manifests itself in the form of a slow but steady increase of the variances. This loss of coherence is indeed of a fundamental nature, and it is due to the interplay of the Heisenberg uncertainty principle and the form of the angular-momentum commutation relations.

Another remarkable fact is that the ability of the atomic system to exchange energy with the electromagnetic field gradually decreases with time, due to the "shortening" of the total angular momentum [of course $\langle S_x^2\rangle + \langle S_y^2\rangle + \langle S_z^2\rangle$ is always $S(S+1)$; here we mean that $\langle S_x\rangle^2 + \langle S_y\rangle^2 + \langle S_z\rangle^2$ gets smaller than S^2]; it is as if some energy, initially in the field, were stored in the atomic system, which is not able to give it back to the field in the form of coherent energy. It is this part which has probably to be taken into account in any theory of the efficiency of a machine made up of atoms which converts incoherent into coherent radiation. It should be noted that this energy, which should be considered as effectively lost for the machine when calculating the amount of coherent energy produced per cycle, increases quadratically with time, leading to the concept of short-duration cycles being "cleaner" than long-duration ones.

Similar features to those discussed above have previously been shown to develop in the decay from the fully excited state in the presence of radiation fields initially in a photon number state.[11] The interesting point about our result is that these features should develop also in the presence of fields which have been made to look as classical as possible.

In this paper we do not wish to pursue further the discussion on the conversion of incoherent into coherent energy, which still requires some more detailed investigations on the interaction between few-level objects and radiation before it can be done in a properly thorough way, as is desirable in view of the conceptual and possibly practical importance of the problem.

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APPENDIX

In this appendix we report a short compendium of properties of Radcliffe states. As it is well known[12] a coherent spin state pointing toward a direction which forms an angle θ with the negative z axis and whose projection on the (x,y) plane forms an angle φ with the x axis can be expressed as

$$|\theta,\varphi\rangle = R_{\theta,\varphi}|-S\rangle ,$$

where R is the rotation operator discussed by Arecchi *et al.* The mean value of an operator A on this state is given by

$$\langle A\rangle = \langle\theta,\varphi|A|\theta,\varphi\rangle = \langle -S|R^{-1}_{\theta,\varphi}AR_{\theta,\varphi}|-S\rangle . \quad \text{(A1)}$$

Since $R^{-1}_{\theta,\varphi} = R_{-\theta,\varphi}$, we find

$$\begin{aligned} S'_- &\equiv R^{-1}_{\theta,\varphi}S_-R_{\theta,\varphi} \\ &= S_-\cos^2\tfrac{1}{2}\theta - S_+e^{-i2\varphi}\sin^2\tfrac{1}{2}\theta - S_z e^{-i\varphi}\sin\theta , \\ S'_+ &\equiv R^{-1}_{\theta,\varphi}S_+R_{\theta,\varphi} \\ &= S_+\cos^2\tfrac{1}{2}\theta - S_-e^{i2\varphi}\sin^2\tfrac{1}{2}\theta - s_z e^{i\varphi}\sin\theta , \\ S'_z &\equiv R^{-1}_{\theta,\varphi}S_zR_{\theta,\varphi} \\ &= S_z\cos\theta + \tfrac{1}{2}\sin\theta(S_+e^{-i\varphi} + S_-e^{i\varphi}) . \end{aligned} \quad \text{(A2)}$$

Hence the average values of raising and lowering operators on $|\theta,\varphi\rangle$ are

$$\langle S_-\rangle = Se^{-i\varphi}\sin\theta ; \quad \langle S_+\rangle = Se^{i\varphi}\sin\theta , \quad \text{(A3)}$$

while

$$\langle S_x\rangle = S\sin\theta\cos\varphi ; \quad \langle S_y\rangle = S\sin\theta\sin\varphi ;$$
$$\langle S_z\rangle = -S\cos\theta . \quad \text{(A4)}$$

The variances in state $|\theta,\varphi\rangle$ can be calculated as follows:

(i) $$(S_z - \langle S_z\rangle)R_{\theta,\varphi}|-S\rangle = R_{\theta,\varphi}(S'_z - \langle S_z\rangle)|-S\rangle = R_{\theta,\varphi}\tfrac{1}{2}\sin\theta e^{-i\varphi}S_+|-S\rangle .$$

From this we have

$$(\Delta S_z)^2 = \|(S_z - \langle S_z\rangle)R_{\theta,\varphi}|-S\rangle\| = \tfrac{1}{2}S\sin^2\theta , \quad \text{(A5)}$$

where $\||\xi\rangle\|$ means $|\langle\xi|\xi\rangle|^2$.

(ii) $$(S_- - \langle S_-\rangle)R_{\theta,\varphi}|-S\rangle = R_{\theta,\varphi}(S'_- - \langle S_-\rangle)|-S\rangle = R_{\theta,\varphi}(e^{-i2\varphi}\sin^2\theta)S_+|-S\rangle , \quad \text{(A6)}$$

from which

$$(\Delta S_-)^2 = \|(S_- - \langle S_-\rangle)R_{\theta,\varphi}|-S\rangle\| = 2S\sin^4\tfrac{1}{2}\theta . \quad \text{(A7)}$$

(iii) $$(S_+ - \langle S_+\rangle)R_{\theta,\varphi}|-S\rangle = R_{\theta,\varphi}(S'_+ - \langle S_+\rangle)|-S\rangle = R_{\theta,\varphi}\cos^2\tfrac{1}{2}\theta S_+|-S\rangle , \quad \text{(A8)}$$

so that

$$(\Delta S_+)^2 = \|(S_+ - \langle S_+\rangle)R_{\theta,\varphi}|-S\rangle\| = 2S\cos^4\tfrac{1}{2}\theta . \quad \text{(A9)}$$

(iv) Summing (A6) to (A8) and dividing by 2 we get

$$(S_x - \langle S_x\rangle)R_{\theta,\varphi}|-S\rangle = R_{\theta,\varphi}(S'_x - \langle S_x\rangle)|-S\rangle = \tfrac{1}{2}R_{\theta,\varphi}(\cos^2\tfrac{1}{2}\theta - e^{-i2\varphi}\sin^2\tfrac{1}{2}\theta)S_+|-S\rangle$$

and

$$(\Delta S_x)^2 = \|(S_x - \langle S_x\rangle)R_{\theta,\varphi}|-S\rangle\| = \tfrac{1}{2}S(\cos^4\tfrac{1}{2}\theta + \sin^4\tfrac{1}{2}\theta - \tfrac{1}{2}\cos2\varphi\sin^2\theta) . \quad \text{(A10)}$$

(v) Subtracting (A6) from (A8) and proceeding as in (iv) we find

$$(\Delta S_y)^2 = \tfrac{1}{2}S(\cos^4\tfrac{1}{2}\theta + \sin^4\tfrac{1}{2}\theta + \tfrac{1}{2}\cos2\varphi\sin^2\theta) . \quad \text{(A11)}$$

It is interesting to consider the product

$$\Delta S_x \Delta S_y = \tfrac{1}{2}S\left[(\cos^4\tfrac{1}{2}\theta + \sin^4\tfrac{1}{2}\theta)^2 - 4\cos^2 2\varphi \sin^4\tfrac{1}{2}\theta\cos^4\tfrac{1}{2}\theta\right]^{1/2},$$

which has minima for $\varphi = \pm\frac{1}{2}n\pi$ $(n=0,1,2\ldots)$. These values of φ characterize the so-called intelligent spin states,[13] for which

$$\Delta S_x \Delta S_y = \tfrac{1}{2}S\cos\theta = \tfrac{1}{2}\left|\langle S_z\rangle\right|,$$

while for all other values of φ one has always

$$\Delta S_x \Delta S_y > \tfrac{1}{2}\left|\langle S_z\rangle\right|.$$

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