

Atomic coherent-state representation of the spectrum of scattered light from a cooperative system and numerical results

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(Received 29 August 1978; revised manuscript received 25 June 1979)

The problem of light scattering from an atomic system under cooperative conditions is analyzed. The atomic correlation function is constructed using the atomic coherent-state representation in a form that is ideally suited for numerical computations. Explicit results are reported for a system of five atoms and the question of the existence of additional side bands is analyzed.

I. INTRODUCTION

The influence of cooperative atomic effects on the statistical and spectral properties of resonance fluorescence has been the subject of considerable recent interest.¹⁻⁵

The properties of the scattered light by a cooperative system are conveniently studied by solving the master equation for the reduced density operator of the atomic system. The master-equation approach treats the interaction with the coherent driving field to all orders in the perturbation. The approach is fully quantum-mechanical and takes into account various quantum-mechanical correlations properly. However, an analytical solution of this master equation has been provided only for the one-⁶ and two-atom case.⁷ The two- and three-atom problems were studied by Agarwal *et al.*² numerically, using the master equation in the usual representation in terms of the angular momentum operator eigenstates. However, as the number of atoms increases, the representation based on the angular momentum eigenstates becomes too cumbersome even for numerical computations. It is clear that alternative methods have to be developed to handle larger numbers of atoms.

In an early discussion of this problem, Senitzky¹ proposed that a large number of externally driven two-level systems would behave in an essentially classical way^{8,9} provided that the atoms are initially removed from a state of complete inversion and that the external driving field is sufficiently strong.

To be sure, there are drastic qualitative differ-

ences between the solution of the classical and quantum equations of motion. For example, under the action of a strong external field, the atomic population difference undergoes damped oscillations until a nonequilibrium steady state is reached with nearly zero population difference between excited and ground atomic states.^{10,11} The classical equations,¹² on the contrary, predict undamped periodic (but not sinusoidal) oscillations in the same strong-field limit. If one allows for the possibility of stochastic amplitude and phase fluctuations in an ensemble of classical solutions, the average population difference will also exhibit a damped character, presumably in good qualitative agreement with the quantum behavior.

Another consequence of this point of view is the appearance of additional sidebands, in the spectrum of resonance fluorescence⁵ beside the well-known pair which have been predicted¹³ and accurately analyzed¹⁴ with low-density atomic systems. Qualitatively, it is easy to see how the spectrum of a strongly driven collection of atoms should display more than the usual pair of sidebands in this classical limit. If the collective atomic polarization oscillates as a classical periodic but not sinusoidal function, the radiated field is bound to contain an infinite number of harmonic components. If, in addition, the classical oscillation is damped, the spectral components will be broadened and slightly shifted. The exact numerical analysis² of the spectrum and intensity correlation function of resonance fluorescence for two- and three-atom systems shows that in the high-field limit, only the usual single-atom pair of sidebands appears in the spectrum. Agarwal *et al.*¹²

also pointed out that, while this result was mathematically surprising, it was not a consequence of numerical round-off errors.

More recently, the two-atom resonance fluorescence spectrum has also been investigated analytically in the strong-field limit.^{3,4} In particular, in Ref. 3, the collective behavior has been analyzed as a function of the atomic separation. In Ref. 7, the master equation has also been solved analytically for the two-atom problem. Thus, little has been done to explore exactly the quantum-mechanical resonance fluorescence problem for large samples.

The behavior of the expectation values of collective atomic operators, on the other hand, has been studied successfully for fairly large numbers of atoms¹¹ using a method based on the atomic coherent-state representation.¹⁵ In this paper we extend this procedure to calculate the spectrum of resonance fluorescence from a collection of atoms driven by an external field of arbitrary strength.

The calculation of the spectrum in the atomic coherent-state representation is reduced to the solution of a set of linear equations. The method of calculation represents a significant improvement with respect to the scheme discussed in Ref. 2 and can easily be used to study larger systems, given sufficient computer time.

In Sec. II we review the model and compare it with the one discussed in Ref. 1. In Sec. III we develop a formula for the atomic correlation function of interest and for the fluorescence spectrum using the atomic coherent-state representation. We demonstrate the use of our atomic coherent-state representation for the spectrum by calculating it for a system of five atoms. Even though we handle a system consisting of a small number of atoms, we believe that our results strongly suggest that no more than two-shifted sidebands will be produced by a sufficiently strong external field.

II. DESCRIPTION OF MODEL

We consider an ensemble of N identical two-level atoms driven by a resonant external field which couples the ground and excited states into a coherent superposition. The model emphasizes only the cooperative decay mechanism so that competition between the collective and single-atom decay modes is not present.

The time evolution of the system is described by the reduced atomic density operator W , solution of the master equation

$$\partial W / \partial t = -i\Omega_f [S^+ + S^-, W] + 2\gamma (S^- W S^+ - \frac{1}{2} W S^+ S^- - \frac{1}{2} S^+ S^- W). \quad (2.1)$$

The parameters Ω_f and 2γ represent the Rabi frequency of the external field and the single-atom

decay rate, respectively. The operators $S^\pm = \sum_i s_i^\pm$ are the collective dipole operators. Together with the atomic energy operator $S_z = \sum_i s_{zi}$, they close under the usual angular momentum commutation rules

$$[S^+, S^-] = 2S_z, [S_z, S^\pm] = \pm S^\pm \quad (\hbar = 1). \quad (2.2)$$

The atoms are initially in the ground state prior to turning on the external field. This circumstance, together with the conserved nature of the total angular momentum $S^2 = S_z^2 + \frac{1}{2}(S^+ S^- + S^- S^+)$, implies that the atoms will evolve in the manifold of collective states of maximum cooperation number.¹⁶ The density operator W is then a $(2S+1) \times (2S+1)$ matrix with $2S = N$, where N is the number of atoms.

In the analysis of Ref. 1, the starting point was a system comprised of N two-level atoms (described, as usual, in the framework of the angular momentum algebra), an infinite denumerable number of field modes for the vacuum field, and a c -number external field. After adiabatic elimination of the quantum field variables, the resulting Heisenberg equations of motion for the expectation values of the atomic collective operators are formally identical to the equations of motion for $\langle S^\pm \rangle$ and $\langle S_z \rangle$ and the quantum-mechanical Langevin equations¹⁷ which follow directly from the master equation (2.1).

In the high-field limit ($\Omega_f \gg N/2$), the Heisenberg equations have been reduced to the following second-order differential equation for the atomic population difference:

$$\frac{d^2}{dt^2} \langle S_z \rangle + 3\gamma \frac{d}{dt} \langle S_z \rangle + 4\Omega_f^2 \langle S_z \rangle - 3\gamma \left\langle \left\{ \frac{dS_z}{dt}, S_z \right\} \right\rangle = 0, \quad (2.3)$$

where the brackets $\{ \}$ indicate the equal-time anticommutator. Equation (2.3) is not a closed differential equation. If one approximates the term $\langle \{ \dot{S}_z, S_z \} \rangle$ by $2\langle \dot{S}_z \rangle \langle S_z \rangle$, a numerical solution can be obtained in a straightforward way.

This approximation does not appear to be too drastic for large values of N (and correspondingly large value of Ω_f). Still, it is clear that certain quantum fluctuations are neglected; the effect of the error on the spectrum of atomic fluctuations is not easy to assess.

In Fig. 1 we show a comparison between the numerical solution of Eq. (2.3) after factorization and the corresponding exact solution for $\langle S_z \rangle$ derived with the method of Ref. 11 for the same choice of parameters. It is worth pointing out that, while the agreement between the exact and approximate solutions improves for increasing values of N , we have no convincing evidence that the atomic fluctuations are described with sufficient accuracy by the above factorization scheme.

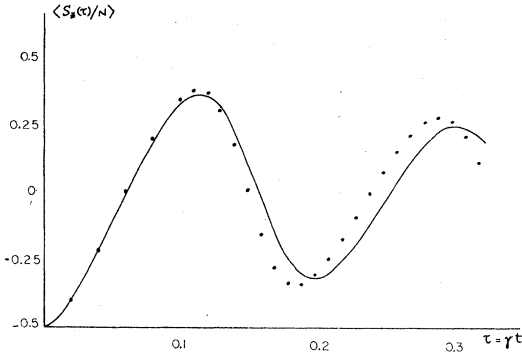


FIG. 1. Time evolution of the atomic population difference per atom $\langle S_z \rangle / N$ for $N=15$ and $\Omega_I/\gamma=18$. Solid line represents the exact solution from the master equation (2.1). Dots represent the solution of the nonlinear differential equation (2.3) after factorization.

III. ATOMIC CORRELATIONS AND SPECTRUM OF FLUCTUATIONS

A very convenient way to analyze the master equation (2.1) is by means of its c -number representative in the atomic coherent-state representation. It was pointed out in Ref. 11 that if $|\Omega^{N/2}\rangle$ is an atomic coherent state in the subspace of maximum cooperation number $S=N/2$, a c -number representative function $P(\Omega, t)$ can be associated to the atomic density operator W as follows:

$$W = \int d\Omega |\Omega^{N/2}\rangle P(\Omega, t) \langle \Omega^{N/2}|, \quad (3.1)$$

$$d\Omega = \sin\theta d\theta d\varphi.$$

The quasi-probability distribution $\tilde{P}(\Omega, t) \equiv \sin\theta P(\Omega, t)$ is found to satisfy the Fokker-Planck equation¹¹

$$\begin{aligned} \frac{d}{d\tau} p_{l,m}(\tau) = & - \left(\frac{(l-m+1)(l+m+1)}{(2l+1)(2l+3)} \right)^{1/2} l(N-l) p_{l+1,m} - \left(\frac{\Omega_I}{\gamma} \right) [(l+m)(l-m+1)]^{1/2} p_{l,m-1} \\ & - [l(l+1) - m^2] p_{l,m} + \left(\frac{\Omega_I}{\gamma} \right) [(l-m)(l+m+1)]^{1/2} p_{l,m+1} + \left(\frac{(l-m)(l+m)}{(2l-1)(2l+1)} \right)^{1/2} (l+1)(N+l+1) p_{l-1,m}, \end{aligned} \quad (3.5)$$

where $\tau = t/\gamma$.

While an infinite set of coefficients $p_{l,m}$ is required for a complete specification of the density function $P(\Omega, t)$, we have shown in Ref. 11 that only the first $(N+1)^2$ coefficients for $0 \leq l \leq N$ and $|m| < l$ are required for an exact description of the time-dependent atomic expectation values. The initial conditions for the expansion amplitudes $p_{l,m}$ in Eq. (3.4) are

$$\begin{aligned} \frac{\partial \tilde{P}}{\partial t} = & - 2\Omega_I \left(\frac{\partial}{\partial \theta} (\sin\theta \tilde{P}) + \frac{\partial}{\partial \varphi} (\cot\theta \cos\varphi \tilde{P}) \right) \\ & + 2\gamma \frac{\partial}{\partial \theta} \left[\left(\frac{N}{2} \sin\theta + \frac{1 - \cos\theta}{2\sin\theta} \right) \tilde{P} \right] \\ & + 2\gamma \frac{\partial^2}{\partial \theta^2} \left(\frac{1 - \cos\theta}{2} \tilde{P} \right) - 2\gamma \frac{\partial^2}{\partial \varphi^2} \left(\frac{1}{2} \frac{\cos\theta}{1 + \cos\theta} \tilde{P} \right). \end{aligned} \quad (3.2)$$

The advantage of the mapping technique, of course, is that this diffusion equation can be treated by standard methods of Fourier analysis on the Bloch sphere surface and by exact numerical techniques.

Once the c -number representative $\tilde{P}(\Omega, t)$ is known, the expectation values of arbitrary atomic operators can be calculated directly by simple quadratures.¹⁵ For example, the collective atomic polarization and population inversion are given by

$$\langle S^{\pm} \rangle = \int \left(\frac{1}{2} N \sin\theta e^{\pm i\varphi} \right) P(\Omega, t) d\Omega, \quad (3.3)$$

$$\langle S_z \rangle = - \int \left[\frac{1}{2} N \cos\theta \right] P(\Omega, t) d\Omega.$$

As it turns out, the Fourier-expansion method developed below leads to the atomic expectation values much more directly, and without the need for evaluating the integrals over the quasi-probability distribution function.

It was stated in Ref. 11 that, if one expands the function $P(\Omega, t)$ in terms of spherical harmonics on the surface of the Bloch sphere

$$P(\Omega, t) = \sum_{l=0}^{\infty} \sum_{|m| \leq l} i^m p_{l,m}(t) Y_l^m(\Omega), \quad (3.4)$$

the Fokker-Planck equation (3.2) reduces to the linear set of coupled equations

$$\begin{aligned} |m| \leq l \leq N: & p_{l,m}(0) = \int P(\Omega, 0) (i^m Y_l^m(\Omega))^* d\Omega, \\ |m| > l: & p_{l,m}(0) = 0, \\ l > N: & p_{l,m}(0) = \text{arbitrary}. \end{aligned} \quad (3.6)$$

In particular, if the atomic system is initially in its ground state, the relevant expansion amplitudes at $t=0$ are given by

$$|m| \leq l \leq N: p_{l,m}(0) = \left(\frac{2l+1}{4\pi}\right)^{1/2} \delta_{m,0}. \quad (3.7)$$

It is also easy to verify that as a result of the orthogonality properties of the spherical harmonics on the unit sphere, the expectation values of the collective atomic operators can all be expressed in terms of the expansion amplitudes. For example, we have

$$\begin{aligned} \langle S^+(\tau)/N \rangle &= \pm \frac{1}{2}i (8\pi/3)^{1/2} p_{1,1}(\tau), \\ \langle S_z(\tau)/N \rangle &= -\frac{1}{2}(4\pi/3)^{1/2} p_{1,0}(\tau). \end{aligned} \quad (3.8)$$

This of course eliminates the need for calculating the integrals (3.3) or the corresponding generalizations for arbitrary operators.

We now turn our attention to the main purpose of this paper, the calculation of the incoherent part of the spectrum of resonance fluorescence. As a result of the linear dependence of the source-field operators and the atomic polarization, the spectrum of the fluorescence light is given by the Fourier transform of the steady-state correlation function

$$\chi(\tau) = \lim_{t \rightarrow \infty} \langle S^+(t+\tau)S^-(t) \rangle, \quad (3.9)$$

to within a frequency independent scale factor.

A convenient way to calculate the atomic correlation function $\chi(\tau)$ has been proposed in Ref. 18 using the atomic coherent-state representation and certain differential rules of mapping (D -operator calculus).¹⁹ For the sake of brevity, we refer to the reader to Ref. 18 for details. Here we only need to point out that if $P_s(\Omega_0)$ and $P(\Omega\tau|\Omega_0)$ represent the steady-state solution of the Fokker-

Planck equation (3.2) and its Green's function, respectively, then the required atomic correlation function is given by

$$\chi(\tau) = \int d\Omega_0 d\Omega P_s(\Omega_0) \mathfrak{D}_s^-(\Omega_0) [P(\Omega, \tau|\Omega_0)] \langle \Omega | S^+ | \Omega \rangle. \quad (3.10)$$

In Eq. (3.10) the differential operator \mathfrak{D}_s^- , which acts only on the angular variables θ_0, φ_0 of the Green's function; is given by

$$\mathfrak{D}_s^-(\Omega_0) = e^{-i\varphi_0} \left(\frac{N}{2} \sin\theta_0 - \sin^2 \frac{\theta_0}{2} \frac{\partial}{\partial \theta_0} - \frac{i}{2} \tan \frac{\theta_0}{2} \frac{\partial}{\partial \varphi_0} \right), \quad (3.11)$$

and the diagonal matrix element of S^+ has the form

$$\langle \Omega | S^+ | \Omega \rangle = \frac{1}{2}N e^{i\varphi} \sin\theta = -\frac{1}{2}N(8\pi/3)^{1/2} Y_1^1(\Omega). \quad (3.12)$$

The integration in Eq. (3.10) is to be carried out twice independently over the surface of the unit sphere. In practice, the calculation of $\chi(\tau)$ is simplified considerably by the Fourier decomposition of the density functions $P_s(\Omega_0)$ and $P(\Omega\tau|\Omega_0)$. The steady-state solution of the Fokker-Planck equation is given by Eqs. (3.4) and (3.5) in the limit $\tau \rightarrow \infty$. The Green's function, instead, is expanded in the biorthogonal series

$$P(\Omega\tau|\Omega_i) = \sum_{l,m} \sum_{l',m'} i^{m+m'} p_{l',m'}^{l',m'}(\tau) Y_{l',m'}^{m'+*}(\Omega_0) Y_l^m(\Omega). \quad (3.13)$$

After substitution of both Fourier expansions (3.4) and (3.13) in Eq. (3.10), the required atomic correlation function takes the form

$$\begin{aligned} \chi(\tau) = \frac{N}{2} \left(\frac{8\pi}{3}\right)^{1/2} \sum_{l,m} (-1)^m p_{l',m'}^{l',m'}(\tau) \left[-\frac{1}{2} [(l-m)(l+m+1)]^{1/2} p_{l,m+1}(\infty) + \frac{1}{2} (N+l+1) \left(\frac{(l-m)(l-m-1)}{(2l-1)(2l+1)}\right)^{1/2} p_{l-1,m+1}(\infty) \right. \\ \left. - \frac{1}{2} (N-l) \left(\frac{(l+m)(l+m+2)}{(2l+1)(2l+3)}\right)^{1/2} p_{l+1,m+1}(\infty) \right]. \end{aligned} \quad (3.14)$$

The time-dependent coefficients $p_{l',m'}^{l',m'}(\tau)$ for fixed l',m' satisfy the same set of differential equations (3.5) as the expansion coefficients $p_{l,m}(\tau)$ (Appendix A). Of course, the initial conditions are different:

$$p_{l',m'}^{l',m'}(0) = \delta_{l',l} \delta_{m',m} (-i)^{m+m'}. \quad (3.15)$$

Some details of the lengthy calculation leading to Eq. (3.14) are summarized in Appendix B.

Equation (3.14) is our final expression for the atomic correlation function in terms of the atomic coherent-state representation. We only need to know the coefficients $p_{l',m'}^{l',m'}(\tau)$ and $p_{l,m}(\infty)$ which can

be obtained from the solution of Eq. (3.5). The spectrum of the scattered light is related to $\chi(\tau)$ by

$$S(\omega) = \text{Re} \int_0^\infty d\tau \chi(\tau) e^{-i\omega\tau}. \quad (3.16)$$

IV. NUMERICAL RESULTS

The spectrum of resonance fluorescence has been constructed in three steps: First, we have solved the set of Eqs. (3.5) for the $(N+1)^2$ coefficients $p_{l,m}(\tau)$ [$l \leq N, |m| \leq l$] and $\tau \rightarrow \infty$. As a second

step, we have solved the coupled set of Eqs. (3.5) for the expansion coefficients $p_{i_m}^{l'm'}(\tau)$ subject to the initial conditions (3.5). This step has been repeated independently for every set of indices l' , m' .

After completion of the second step we have extracted the set of coefficients $p_{1,-1}^{l'm}$, which are required for the construction of $\chi(\tau)$. Two typical correlation functions for a sample of five atoms driven by fields of different strength are shown in Fig. 2.

As a final step we have calculated the numerical Fourier transform of the numerical correlation function $\chi(\tau)$. Two typical results are plotted in Fig. 3.

In order to ensure numerical accuracy, we have used different grid sizes for both the time-dependent solution of Eq. (3.5) and for its numerical Fourier transform. In both cases, we have found no significant variation upon changing the time and frequency step. As an additional check, we have tested our computer codes against the well-known analytic expressions for the single-atom case.

On the basis of our calculations, it is well established that, at least for the limited sample size which we have been able to analyze, the fluorescence spectrum shows no evidence of additional sidebands beside the usual Rabi-shifted pair over a frequency range equal to approximately 15–20 times the scaled Rabi frequency Ω_I/γ . In addition the cooperative spectrum approaches the single-atom spectrum to within a scale factor for large values of the driving field.

Our recent analytical results for the two-atom problem have established that the spectrum of the resonance fluorescence in the neighborhood of $2\Omega_I/\gamma$ is a monotonically decreasing function of ω .

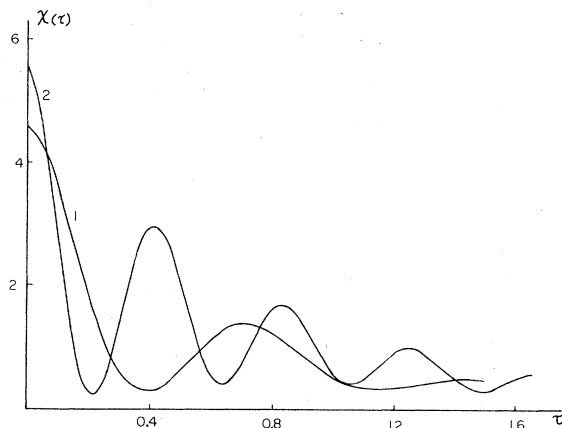


FIG. 2. Time dependence of the atomic correlation function $\chi(\tau)$ for a sample of five atoms. The scaled Rabi frequencies are (1) $\Omega_I/\gamma=5$ and (2) $\Omega_I/\gamma=8$.

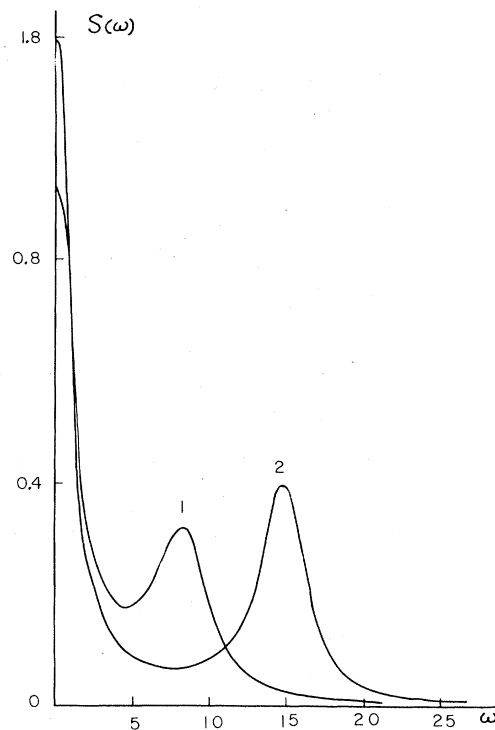


FIG. 3. Numerical Fourier transforms of the atomic correlation function $\chi(\tau)$ for a sample of five atoms. The scaled Rabi frequencies are (1) $\Omega_I/\gamma=5$ (2) $\Omega_I/\gamma=8$.

Our analytical results also establish that any additional structures lie in the tail of the spectrum and are not resolvable. In view of our numerical work, and of the analytical results of Ref. 7, we conclude that the additional resonant structures as discussed by Senitzky are not ordinarily resolvable.

We conclude with a comment on a recent suggestion by Amin and Cordes⁴ regarding the limiting form of the multi-atom spectrum for large external fields. These authors have speculated that for $\Omega_I \rightarrow \infty$, the N -atom and the single-atom spectrum appear to differ only by the scale factor $N(N+2)/3$. The origin of the proportionality factor is easily traced from Eq. (3.14). The initial value of the atomic correlation function is given by

$$\chi(0) = \frac{N}{2} \left(\frac{8\pi}{3} \right)^{1/2} \left[-\frac{\sqrt{2}}{2} p_{1,0}(\infty) + \frac{N+2}{2} \left(\frac{2}{3} \right)^{1/2} p_{0,0}(\infty) \right]. \quad (4.1)$$

Since, however, $p_{1,0}$ is proportional to the atomic population difference, while $p_{0,0}$ is a constant of motion [cf. Eq. (3.5)] it follows that

$$\chi(0) \xrightarrow{\Omega_I \rightarrow \infty} \frac{N(N+2)}{3}. \quad (4.2)$$

Thus, at least as far as the integrated spectrum is concerned, Amin and Cordes' speculation is well substantiated.

APPENDIX A: EQUATIONS OF MOTION OF EXPANSION COEFFICIENTS $P_{lm}^{l'm'}(\tau)$

The biorthogonal expansion of the Green's function $P(\Omega, \tau | \Omega_0)$ can be cast into the form

$$P(\Omega, \tau | \Omega_0) = \sum_{lm} i^m p_{lm}(\tau, \Omega_0) Y_l^m(\Omega), \quad (\text{A1})$$

where

$$p_{lm}(\tau, \Omega_0) = \sum_{l'm'} i^{m'} p_{l'm'}^{l'm'}(\tau) Y_{l'}^{m'*}(\Omega_0). \quad (\text{A2})$$

By definition, the coefficients $p_{lm}(\tau, \Omega_0)$ satisfy the set of equations (3.5) which we can write in the symbolic form

$$\frac{\partial}{\partial \tau} p_{lm}(\tau, \Omega_0) = \sum_{p,q} \mathcal{L}_{lm}^{pq} p_{pq}(\tau, \Omega_0). \quad (\text{A3})$$

After multiplication of Eq. (A3) by $Y_r^s(\Omega_0)$ and integration over Ω_0 , one finds

$$\frac{\partial}{\partial \tau} p_{lm}^r(\tau) = \sum_{p,q} \mathcal{L}_{lm}^{pq} p_{pq}^r(\tau) \quad (\text{A4})$$

for every pair of indices r, s .

APPENDIX B: DERIVATION OF EQ. (3.14)

The starting point of our analysis is the correlation function²⁰

$$\chi(\tau) = \int d\Omega_0 d\Omega P_s(\Omega_0) \mathfrak{D}_s(\Omega_0) [P(\Omega, \tau | \Omega_0)] \langle \Omega | S^+ | \Omega \rangle, \quad (\text{B1})$$

where

$$\langle \Omega | S^+ | \Omega \rangle = \frac{N}{2} e^{i\phi} \sin\theta = -\frac{N}{2} \left(\frac{8\pi}{3}\right)^{1/2} Y_1^1(\Omega), \quad (\text{B2})$$

$$P_s(\Omega_0) = \sum_{lm} i^m p_{lm}(\infty) Y_l^m(\Omega_0), \quad (\Omega_0 \equiv \theta_0, \phi_0), \quad (\text{B3})$$

$$P(\Omega, \tau | \Omega_0) = \sum_{lm} \sum_{l'm'} i^{m+m'} p_{l'm'}^{l'm'}(\tau) Y_{l'}^{m'*}(\Omega_0) Y_l^m(\Omega), \quad (\text{B4})$$

and

$$\mathfrak{D}_s(\Omega_0) = e^{-i\phi_0} \left(\frac{N}{2} \sin\theta_0 - \sin^2 \frac{\theta_0}{2} \frac{\partial}{\partial \theta_0} - \frac{i}{2} \tan \frac{\theta_0}{2} \frac{\partial}{\partial \phi_0} \right). \quad (\text{B5})$$

After substituting Eqs. (B2)–(B5) in Eq. (B1) and carrying out the Ω integration, we arrive at

$$\chi(\tau) = \sum_{l'm'} i^{m+m'-1} p_{l'm'}^{l'm'}(\tau) \frac{N}{2} \left(\frac{8\pi}{3}\right)^{1/2} \sum_{lm} p_{lm}(\infty) \times \int d\Omega_0 Y_l^m(\Omega_0) \mathfrak{D}_s(\Omega_0) Y_{l'}^{m'*}(\Omega_0). \quad (\text{B6})$$

The problem is now reduced to the calculation of the integral

$$I_{l'm'}^{l'm'} = \int d\Omega Y_l^m(\Omega) \mathfrak{D}_s(\Omega) Y_{l'}^{m'*}(\Omega) = A + B, \quad (\text{B7})$$

where

$$A = \frac{N}{2} \int d\Omega Y_l^m(\Omega) e^{-i\phi} \sin\theta Y_{l'}^{m'*}(\Omega), \quad (\text{B8})$$

$$B = -\frac{1}{2} \int d\Omega Y_l^m(\Omega) e^{-i\phi} \left((1 - \cos\theta) \frac{\partial}{\partial \theta} + i \frac{1 - \cos\theta}{\sin\theta} \frac{\partial}{\partial \phi} \right) Y_{l'}^{m'*}(\Omega). \quad (\text{B9})$$

The A contribution can be calculated trivially from the identity

$$e^{-i\phi} \sin\theta Y_{l'}^{m'*} = -\left(\frac{(l'+m'+1)(l'+m'+2)}{(2l'+1)(2l'+3)} \right)^{1/2} Y_{l'+1}^{m'+1*} + \left(\frac{(l'-m')(l'-m'-1)}{(2l'-1)(2l'+1)} \right)^{1/2} Y_{l'-1}^{m'+1*}. \quad (\text{B10})$$

The result is

$$A = \frac{N}{2} \left[-\left(\frac{(l'+m'+1)(l'+m'+2)}{(2l'+1)(2l'+3)} \right)^{1/2} \delta_{l,l'+1} \delta_{m,m'+1} + \left(\frac{(l'-m')(l'-m'-1)}{(2l'-1)(2l'+1)} \right)^{1/2} \delta_{l,l'-1} \delta_{m,m'+1} \right]. \quad (\text{B11})$$

The B contribution is a bit more involved and it appears to require explicit consideration of the properties of the associated Legendre functions. If we let

$$Y_l^m(\Omega) = N_{lm} e^{im\phi} p_l^m(\theta), \quad (\text{B12})$$

$$N_{lm} = (-1)^m \left(\frac{2l+1}{4\pi} \frac{(l-m)!}{(l+m)!} \right)^{1/2},$$

and carry out the ϕ integration, we are left with

$$B = -\pi N_{lm} N_{l',m'} \delta_{m',m-1} \int_{-1}^1 dz p_l^m(z) \frac{1-z}{(1-z^2)^{1/2}} \times \left((1-z^2) \frac{d}{dz} p_{l'}^{m-1} + (m-1) p_{l'}^{m-1} \right), \quad (\text{B13})$$

where we have set $z = \cos\theta$.

The successive use of the identities

$$(1-z^2) \frac{d}{dz} p_\nu^\mu = (1-z^2)^{1/2} p_{\nu+1}^{\mu+1} - \mu z p_\nu^\mu, \quad (\text{B14})$$

$$(1-z^2)^{1/2} p_\nu^{\mu-1} = \frac{1}{2\nu+1} (p_{\nu-1}^\mu - p_{\nu+1}^\mu), \quad (\text{B15})$$

$$z p_\nu^\mu = \frac{\nu-\mu+1}{2\nu+1} p_{\nu+1}^\mu + \frac{\nu+\mu}{2\nu+1} p_{\nu-1}^\mu, \quad (\text{B16})$$

leads to a linear combination of integrals that can be solved immediately from the orthogonality relation of the associated Legendre functions.

The result is

$$B = \pi N_{lm} N_{l'm'} \delta_{m',m-1} \left(\delta_{ll'} - \frac{l-1}{2l-1} \delta_{l',l-1} - \frac{l+2}{2l+3} \delta_{l',l+1} \right) \frac{2}{2l+1} \frac{(l+m)!}{(l-m)!}. \quad (\text{B17})$$

The remaining steps leading to Eq. (3.14) involve only trivial algebraic manipulations.

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