Two-atom resonance fluorescence including the dipole-dipole interaction*

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The spectrum of resonance fluorescence and the second-order correlation function are calculated for a system of two identical two-level cooperative systems interacting via the dipole-dipole interaction. The results are valid for arbitrary incident-field strengths and for atomic separations much less than the resonant wavelength. The intensity of the sidebands at the Rabi frequency is diminished by the dipole interaction, whereas that of the sidebands at twice the Rabi frequency is enhanced. Both the cooperative behavior and the dipole-dipole interaction act to diminish the photon antibunching, as shown by the second-order correlation function.

I. INTRODUCTION

Resonance fluorescence, or the resonant interaction of electromagnetic radiation with an atomic system, has received considerable attention in recent years. Mollow's original prediction[1] of a three-peaked spectrum for one atom at a suitably high excitation-field strength has been experimentally verified,[2-4] and several papers dealing with the collective behavior of several two-level systems (TLS's) have predicted additional structure in the spectrum. For the special case of two TLS's, a second pair of peaks in the scattered spectrum has been predicted,[5-7] and in four papers[8-11] the effect of the first-order dispersion force,[12] or dipole-dipole interaction, has also been considered. We refer the reader to the papers of Mavroyannis[9] and Agarwal et al.[6] for a more complete list of references.

The purpose of this paper is to present analytic results for the spectrum of resonance fluorescence for two atoms interacting via the dipole-dipole interaction, which are valid for arbitrary incident-field strengths and for atomic separations much less than a wavelength. In our model we assume two identical TLS's, separated by a distance R, which may be described as coupled spin-$\frac{1}{2}$ systems interacting with an external radiation field and with each other. Any state of the system may be written as a superposition of states which are symmetric and antisymmetric under interchange of the atoms. The master equation[13] for the reduced atomic-density operator for such a system leads to a set of 15 coupled equations.[14] The spatial phase factor of the driving field couples the symmetric

operators with the antisymmetric. Since we are interested in the effect of the dipole-dipole interaction, which increases strongly with decreasing atomic separation,[15] we assume that atomic separations are much less than the resonant wavelength. This has the added advantage that symmetric and antisymmetric operators no longer appear in the same equations; we must now solve a set of eight coupled equations containing only symmetric operators. From these symmetric operators, the first- and second-order field correlation functions[16]

$$\langle E^{-}(t+\tau)E^{+}(t)\rangle$$

and

$$\langle E^{-}(t)E^{-}(t+\tau)E^{+}(t+\tau)E^{+}(t)\rangle$$

may be calculated; the Fourier transform of the first yields the scattered spectrum, and the second yields information about the so-called antibunching phenomenon.

The master equation for this system is described in Sec. II and its solution is presented in Appendix A. The atomic correlation functions, which are proportional to the field correlation functions,[17] are derived in Appendix B. Several plots of the scattered spectrum are displayed and discussed in Sec. III. The effect of the dipole-dipole interaction is to broaden and flatten the central peak and the first pair of sidebands, while enhancing the height of the second pair of sidebands. The Laplace transform of the second-order correlation function is presented in Sec. IV, along with its value at $t=0$ and as $t\rightarrow\infty$. These results are compared with those of Agarwal et al.[6] The antibunching phenomenon is reduced by the interaction between

the atoms.

II. DESCRIPTION OF THE MODEL

Each TLS may be described as a spin-$\frac{1}{2}$ system, characterized by operators $S_i^{\pm}$ and S_i^z, which are the ordinary spin angular momentum operators. We assume that one TLS is at the origin $\vec{r}_1 = 0$, while the other is located at $\vec{r}_2 = \vec{R}$. The incident EM field is given by

$$\vec{E}(\vec{r},t) = \vec{E}_0 \cos(\omega_0 t) , \qquad (1)$$

with frequency ω_0 equal to the resonant frequency of one TLS. It is assumed that the atomic separation is much less than the resonant wavelength so that the spatial dependence of the field may be neglected.

The master equation for the reduced density operator in the rotating-wave, Born, and Markov approximations, in a frame rotating with the frequency of the applied laser field, is[13]

$$\frac{\partial \rho}{\partial t} = -i\Omega_d[S_1^+S_2^- + S_2^+S_1^-, \rho]$$

$$-\tfrac{1}{2}i\Omega_R[S^+ + S^-, \rho]$$

$$-\gamma(\rho S^+ S^- - 2S^- \rho S^+ + S^+ S^- \rho) . \qquad (2)$$

$S^{\pm} = S_1^{\pm} + S_2^{\pm}$ are the collective atomic dipole operators, 2γ is the Einstein A coefficient and represents the natural linewidth of the atom, $\Omega_R = \vec{d} \cdot \vec{E}_0 \equiv \gamma \omega_R$ is the Rabi frequency, and

$$\Omega_d = \frac{3\gamma}{2(k_0 R)^3}(1 - 3\cos^2\theta) \equiv \gamma \omega_d \qquad (3)$$

represents the dipole-dipole interaction, where $k_0 = \omega_0/c$, and $\cos\theta = \hat{d} \cdot \hat{R}$. The dipole moments $\vec{d}$ of the atoms are assumed identical. In Eq. (2), the first term represents the coupling of the atoms through the dipole-dipole interaction, while the second represents the coupling between the driving field and the atoms. The final term arises from the atom-field coupling and the atom-atom radiative coupling.

From Eq. (2), a set of eight equations for the time dependence of the symmetric atomic operators is derived. The solution of this set of equations is simplified by using the Laplace-transformed equations and is presented in Appendix A. As shown in Appendix B, the Laplace transform of the first- and second-order atomic correlation functions $\langle S^+(t+\tau)S^-(t)\rangle$ and $\langle S^+(t)S^+(t+\tau)S^-(t+\tau)S^-(t)\rangle$ may be calculat-

ed from the Laplace transform of the solution of Eq. (2),

$$\langle S^+(z)\rangle = \sum_{i=1}^{9} f_i(z)\langle J_i(\tau=0)\rangle , \qquad (4a)$$

and

$$\langle S^+S^-(z)\rangle = \sum_{i=1}^{9} g_i(z)\langle J_i(\tau=0)\rangle . \qquad (4b)$$

The $f_i(z)$ and $g_i(z)$ are polynomials in z, and the $\langle J_i(0)\rangle$ are the initial values of the atomic operators as defined in Appendix A.

III. THE SPECTRUM OF RESONANCE FLUORESCENCE

As shown in Appendix B, the steady-state first-order correlation function is given by

$$\Gamma_1(z) = \sum_i f_i(z)\left[\lim_{t\to\infty}\langle J_i(t)S^-(t)\rangle\right]$$

$$= \left[\frac{\omega_R^2 J_0}{zBD(z)}\right][n_1(z)x_0 + zn_2(z)v_0$$

$$+ \omega_d^2 z_2 v_2 + 4\omega_d^4 z_2^2 z_4^2] , \qquad (5)$$

where $z_n = z+n$, J_0 is a constant dependent upon the initial conditions,

$$B = 64 + 16\omega_R^2 + 3\omega_R^4 + 16\omega_d^2 , \qquad (6)$$

and $n_1(z)$, $n_2(z)$, x_0, v_0, v_2, and $D(z)$ are polynomials in z which are listed in Appendix B. Note that Eq. (5) contains no imaginary term. This means that the spectrum is symmetric, contrary to the results of Kuś and Wódkiewicz,[10] who perform a similar calculation.

In the limit $\omega_d \to 0$ (no dipole-dipole interaction), we find

$$\Gamma_1 = \frac{\omega_R^2 J_0}{B}\left[\frac{n_1(z)}{zv_0} + \frac{n_2(z)}{x_0}\right] , \qquad (7)$$

which agrees with Agarwal et al.[6] for the case of both atoms initially in the ground state ($J_0 = 4$), after we account for notational differences by making the substitutions $z \to 2z$ and $\omega_R \to 4\beta$.

The spectrum of resonance fluorescence $S(\omega,R,\theta)$ is given by

$$S(\omega,R,\theta) \equiv \mathrm{Re}\,\Gamma_1[i(\omega - \omega_0)] . \qquad (8)$$

In Fig. 1 the spectrum is shown for various strengths of the dipole-dipole interaction and ω_R = 14. This may be compared with similar figures in the papers by Ja Kilin and Freedhoff.[18] The central peak is flattened by the dipole term, while the sidebands are broadened; additional peaks appear displaced from resonance by frequencies other than the Rabi frequency and may be ascribed to the presence of the dipole-dipole interaction.

The experiments cited[2-4] have used atomic-beam methods to reduce the Doppler broadening. This means that the atomic separation distance R and the atomic orientation are random variables; the spectrum should be averaged over ω_d,

$$\bar{S}(\omega)=\frac{1}{4\pi}\int d\Omega \int_0^\infty dR \; P(R)S(\omega,R,\theta) \;, \qquad (9)$$

where

$$P(R)=4\pi\langle N\rangle R^2 \exp(-4\pi\langle N\rangle R^3/3) \qquad (10)$$

is the probability that two nearest-neighbor atoms in an atomic beam are separated by a distance R, assuming that the atoms are randomly located. $\langle N\rangle$ is the atomic-number density. We assume that the most important collective effects arise from atomic pairs, and we ignore collective effects due to groups of three or more atoms. The integration over R must be consistent with the approximations made above; the integral is separated into two parts,

$$\bar{S}(\omega)\cong\frac{1}{4\pi}\int d\Omega \int_0^{R_0} dR \; P(R)S(\omega,R,\theta)$$

$$+ S(\omega)\int_{R_0}^\infty dR \; P(R) \;. \qquad (11)$$

The first term includes values of R for which the dipole-dipole interaction is greatest. The remaining contribution is significant only for the case of the smallest number density considered here, and even then, only at resonance and for $\omega_R \lesssim 1$. We evaluate this term by replacing $S(\omega,R,\theta)$ by the two-atom spectrum for large separations[14] which may be factored from the integral. This approach is most meaningful when R_0 is chosen to satisfy the condition

$$|\omega_d(R_0,\theta=0)| = 1 \;. \qquad (12)$$

Figures 2 − 5 display the averaged spectrum for various values of the Rabi frequency. Each figure shows the spectrum for $\omega_d = 0$, and for $\langle N\rangle = 10^{14}$ cm^{-3}, 10^{15} cm^{-3}, and 10^{16} cm^{-3}. For case (a) of Fig. 2, the second integral of Eq. (11) is dominant at resonance, and we feel that our calculation of that contribution is not to be trusted; consequently we have suppressed those points in our figure. Each spectrum is normalized by dividing the intensity by the value at resonance in the absence of the dipole term. One effect of the dipole interaction is to broaden and flatten the peaks. For large enough values of the Rabi frequency, the first pair of sidebands appears (Fig. 3); for still larger values, a second set of sidebands becomes
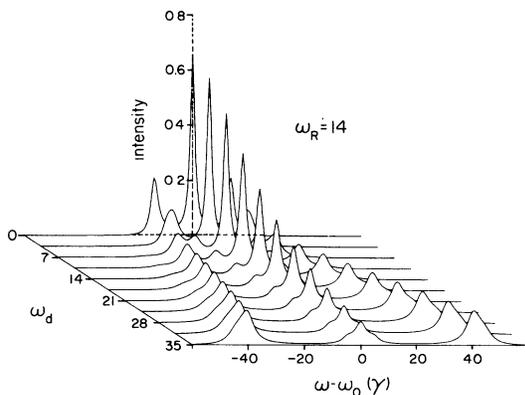

FIG. 1. Plot of Eq. (8) for a Rabi frequency of 14γ and for the dipole-dipole strength ω_d varying between 0 and 35.
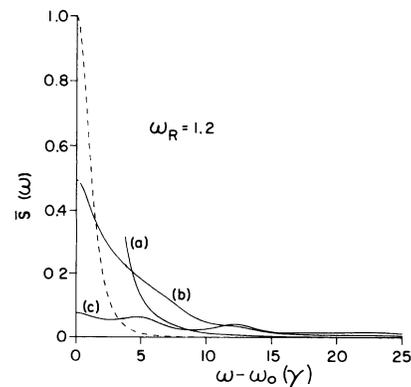

FIG. 2. Plot of Eq. (11) for a Rabi frequency 1.2γ and (a) $\langle N\rangle = 10^{14}$ cm^{-3}, (b) 10^{15} cm^{-3}, and (c) 10^{16} cm^{-3}. The dashed curve is the spectrum without the dipole-dipole interaction; its value at resonance is used to normalize the curves (a), (b), and (c).
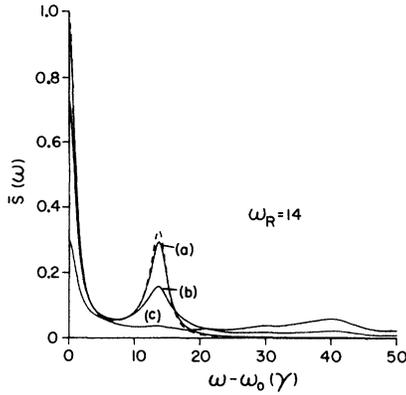
FIG. 3. As in Fig. 2, but for $\omega_R = 14$.

apparent (Fig. 5). It is interesting to notice that the amplitude of these extra peaks increases with increasing $\langle N \rangle$, while the amplitude of the first set of peaks decreases. This behavior for suitable

values of ω_d and ω_R has been noted in the paper of Ja Kilin.[11] As the number density $\langle N \rangle$ increases, the fluorescence energy is shifted increasingly into regions away from resonance.

IV. THE SECOND-ORDER CORRELATION FUNCTION

The Laplace-transformed steady-state second-order correlation function may be obtained from Eq. (B8) of Appendix B,

$$\Gamma_2(z) = \frac{\omega_R^2 J_0}{B}\left[\frac{(4+\omega_R^2)}{z}\left[1-\frac{t_0}{2D(z)}\right] -2\frac{t_1}{D(z)}\right] , \qquad (13)$$

where

$$t_0 = x_0[8z_2^2z_4^2+8\omega_R^2zz_3-\omega_R^4(3z+8)]+\omega_d^2z_2z_4[8z_2(z_2^2+z_4^2)+16\omega_R^2z_1-\omega_R^4]+8\omega_d^4z_2^2z_4 , \qquad (14a)$$

and

$$t_1 = x_0[2z_2^2z_4^2-2\omega_R^2(z^3+7z^2+19z+20)-5\omega_R^4z_3] + \omega_d^2z_2[2z_2z_4(z_2^2+z_4^2)-2\omega_R^2z_4(z^2-2)-\omega_R^4z_7]+2\omega_d^4z_2^2z_4 . \qquad (14b)$$

Its value as $\tau \to \infty$ is given by

$$\Gamma_2(\tau \to \infty) = \lim_{z \to 0} z\Gamma_2(z) = \frac{4\omega_R^4 J_0(4+\omega_R^2)^2}{B^2} . \qquad (15)$$

which reduces to $[\Gamma_1(\tau=0)]^2$ for the case of both atoms in the ground state, or both in the excited
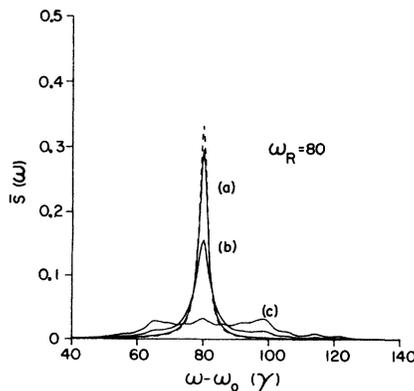
state $(J_0=4)$. The normalized second-order correlation function may be defined as

$$\gamma_2(z) \equiv \frac{\Gamma_2(z)}{[\Gamma_1(\tau=0)]^2} = \frac{B^2}{\omega_R^4 J_0^2(4+\omega_R^2)^2}\Gamma_2(z) . \qquad (16)$$
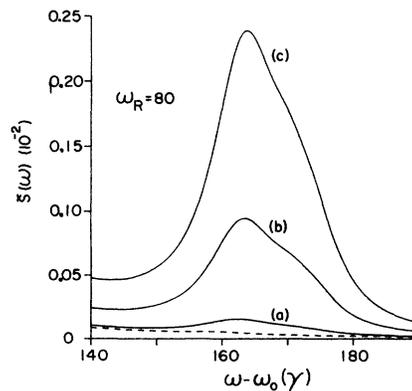
At $\tau=0$, the normalized function has the value

FIG. 4. As in Fig. 2, but for $\omega_R = 80$; only the portion of the spectrum around the first sideband is shown.

FIG. 5. As in Fig. 4, but for the portion of the spectrum around the second sideband.

$$\gamma_2(\tau=0) \equiv \lim_{z \to \infty} z\gamma_2(z)$$

$$= \frac{64 + 16\omega_R^2 + 3\omega_R^4 + 16\omega_d^2}{J_0(4 + \omega_R^2)^2} . \tag{17}$$

For $J_0 = 4$, and in the limit $\omega_R \gg 1$ and $\omega_d \ll \omega_R$, $\gamma_2(\tau=0) = 0.75$, which agrees with Agarwal et al.[6] For the case of a single atom, $\gamma_2(0) \equiv 0$,[19] which is complete antibunching. Thus, the antibunching effect is greatly reduced by the cooperative behavior of two atoms and by their dipole-dipole interaction.

In the limit of weak incident-field strength, the correlation function depends strongly on the dipole-dipole energy, and the antibunching is greatly reduced.

V. DISCUSSION

We have calculated the spectrum of resonance fluorescence and the second-order correlation function for a system of two two-level systems interacting via the dipole-dipole interaction and via radiative coupling. The scattered spectrum is symmetric about the resonant frequency and is altered significantly by the presence of the dipole-dipole term. Pairs of sidebands are displaced from resonance by $\pm\omega_R$ and $\pm 2\omega_R$, where ω_R is the Rabi frequency. The intensity of the first pair is diminished by the dipole interaction, whereas that of the second pair is enhanced. Perhaps this second set of sidebands would be observable in an atomic-beam experiment for suitably large atomic number densities.

The second-order correlation function predicts incomplete antibunching for strong incident-field strengths. At weak-field strengths the antibunching depends strongly on the dipole-dipole energy.

APPENDIX A: SOLUTION OF THE MASTER EQUATION

The master equation (2) is used to calculate the time dependence of the expectation values of the atomic operators. The solution of the resulting eight equations is simplified by using the following symmetric operators:

$$S_H \equiv \tfrac{1}{2}(S^+ + S^-) ,$$

$$S_A \equiv \frac{1}{2i}(S^+ - S^-) ,$$

$$S_z \equiv S_1^z + S_2^z ,$$

$$Q_H \equiv \tfrac{1}{2}(S_1^+ S_2^+ + S_1^- S_2^-) , \tag{A1}$$

$$Q_A \equiv \frac{1}{2i}(S_1^+ S_2^+ - S_1^- S_2^-) ,$$

$$T_H \equiv \tfrac{1}{2}[(S_1^+ + S_1^-)S_2^z + (S_2^+ + S_2^-)S_1^z] ,$$

$$T_A \equiv \frac{1}{2i}[(S_1^+ - S_1^-)S_2^z + (S_2^+ - S_2^-)S_1^z] ,$$

$$R \equiv S_1^+ S_2^- + S_2^+ S_1^- .$$

The Laplace transform of an operator $O(\tau)$ is defined by

$$O \equiv \langle O(z) \rangle = L\{\langle O(\tau) \rangle\} = \int_0^\infty d\tau\, e^{-z\tau}\langle O(\tau) \rangle , \tag{A2}$$

where $\tau = \gamma t$ is used to simplify the notation. The transformed equations are

$$z_1 S_H = I_1 + 2T_H + 2\omega_d T_A ,$$

$$z_1 S_A = I_2 - 2\omega_d T_H + 2T_A - \omega_R S_z ,$$

$$z_2 S_z = \left(I_3 - \frac{2}{z}\right) - 2R + \omega_R S_A ,$$

$$z_2 Q_H = I_4 + \omega_R T_A ,$$

$$z_2 Q_A = I_5 - \omega_R T_H ,$$

$$z_5 T_H = I_6 - \tfrac{3}{2}S_H + \tfrac{1}{2}\omega_d S_A + \omega_R Q_A , \tag{A3}$$

$$z_5 T_A = \left(I_7 - \omega_R \frac{I_0}{z}\right) - \tfrac{1}{2}\omega_d S_H - \tfrac{3}{2}S_A$$

$$+ \omega_R(\tfrac{3}{2}R - Q_H) ,$$

$$z_6 R = \left(I_8 + 4\frac{I_0}{z}\right) + 2S_z - 2\omega_R T_A ,$$

where $z_n = z + n$, the I_i are the initial values of the operators, e.g., $I_1 = \langle S_H(\tau=0) \rangle$, $I_2 = \langle S_A(\tau=0) \rangle$, etc., and

$$I_0 = \langle R(0) + 2S_1^z S_2^z(0) \rangle$$

$$= s(s+1) - \tfrac{3}{2} ,$$

where s is the total spin quantum number for the two-atom system. The notation for the Laplace-transformed operators is the same as that for the time-dependent operators except for argument.

These equations when solved give for S^+ and $S^+ S^-$

$$\langle S^+(z)\rangle = \sum_{i=1}^{9} f_i(z)\langle J_i(\tau=0)\rangle \ ,\tag{A4}$$

and

$$\langle S^+ S^-(z)\rangle = \sum_{i=1}^{9} g_i(z)\langle J_i(\tau=0)\rangle \ ,\tag{A5}$$

where $J_1 = S_H(\tau)$, $J_2 = S_A(\tau)$, etc., $J_9 = 1$ and the $f_i(z)$ and $g_i(z)$ are polynomials in z of order 7 or less.[14] Equations (A4) and (A5) are used in Appendix B to calculate the correlation functions.

APPENDIX B: THE CORRELATION FUNCTIONS

The quantum-regression theorem[20] states that if B and J_i are system operators such that

$$\langle B(t+\tau)\rangle = \sum_i f_i(\tau)\langle J_i(t)\rangle \ ,\tag{B1}$$

then

$$\langle A(t)B(t+\tau)C(t)\rangle$$
$$= \sum_i f_i(\tau)\langle A(t)J_i(t)C(t)\rangle \ .\tag{B2}$$

The field correlation functions are proportional to the atomic correlation functions,[17]

$$\langle E^-(t+\tau)E^+(t)\rangle \propto \langle S^+(t+\tau)S^-(t)\rangle$$

and

$$\langle E^-(t)E^-(t+\tau)E^+(t+\tau)E^+(t)\rangle$$
$$\propto \langle S^+(t)S^+(t+\tau)S^-(t+\tau)S^-(t)\rangle \ .$$

Since we are interested in the steady-state functions, we define the correlation functions by

$$\Gamma_i(z) = \lim_{t\to\infty} \int_0^{\infty} d\tau\, e^{-z\tau}\Gamma_i(t,\tau) \ ,\tag{B3}$$

where

$$\Gamma_1(t,\tau) = \langle S^+(t+\tau)S^-(t)\rangle \ ,\tag{B4}$$

and

$$\Gamma_2(t,\tau) = \langle S^+(t)S^+(t+\tau)S^-(t+\tau)S^-(t)\rangle \ .\tag{B5}$$

Equations (A4), (A5), (B1), (B2), and (B3) together imply

$$\Gamma_1(z) = \sum_i f_i(z)[\lim_{t\to\infty} \langle J_i(t)S^-(t)\rangle] \ ,\tag{B6}$$

and

$$\Gamma_2(z) = \sum_i g_i(z)[\lim_{t\to\infty} \langle S^+(t)J_i(t)S^-(t)\rangle] \ .\tag{B7}$$

Upon simplification of Eq. (B6), we find

$$\Gamma_1(z) = \left[\frac{\omega_R^2 J_0}{zBD(z)}\right][\, n_1(z)x_0 + zn_2(z)v_0$$
$$+ \omega_d^2 z_2 v_2 + 4\omega_d^4 z_2^2 z_4^2] \ ,$$

where

$$J_0 = 3 + 2I_0 \ ,$$

$$B = 64 + 16\omega_R^2 + 3\omega_R^4 + 16\omega_d^2 \ ,$$

$$D(z) = x_0 v_0 + \omega_d^2 z_2 v_1 + \omega_d^4 z_2^2 z_4^2 \ ,$$

$$x_0 = z_2^2 z_4 + \omega_R^2 z_1 \ ,$$

$$v_0 = z_2^2 z_4^3 + \omega_R^2(5z^3 + 37z^2 + 88z + 64) + 4\omega_R^4 z_3 \ ,$$

$$v_1 = z_2 z_4^2(z_2^2 + z_4^2) + 2\omega_R^2 z_1 z_4(3z + 8) + \omega_R^4 z_6 \ ,$$

$$v_2 = 4z_2 z_4^2(z_2^2 + z_4^2) + \omega_R^2 z_4(z^4 + 10z^3 + 60z^2 + 144z + 96) + \tfrac{1}{2}\omega_R^4(5z^3 + 32z^2 + 60z + 64) \ ,$$

$$n_1(z) = 4z_2^2 z_4^3 + \tfrac{1}{2}\omega_R^2(z^5 + 17z^4 + 156z^3 + 696z^2 + 1408z + 1024) + 2\omega_R^4(z^3 + 7z^2 + 23z + 32) \ ,$$

and

$$n_2(z) = \tfrac{1}{2}\omega_R^2(z_3 z_4 + \omega_R^2) \ .$$

I_0 is defined in Appendix A, and $z_n = z + n$. The solution of Eq. (B7) is Eq. (13) in Sec. IV.

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