Nonlinear metrology with a quantum interface

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Abstract. We describe nonlinear quantum atom–light interfaces and nonlinear quantum metrology in the collective continuous variable formalism. We develop a nonlinear effective Hamiltonian in terms of spin and polarization collective variables and show that model Hamiltonians of interest for nonlinear quantum metrology can be produced in $^{87}$Rb ensembles. With these Hamiltonians, metrologically relevant atomic properties, e.g. the collective spin, can be measured better than the ‘Heisenberg limit’ $\propto 1/N$, where $N$ is the number of photons. In contrast to other proposed nonlinear metrology systems, the atom–light interface allows both linear and nonlinear estimation of the same atomic quantities.

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1. Introduction

In quantum metrology, a quantum state is prepared, evolves under the action of a Hamiltonian containing a parameter \( x \) of interest and is measured. The parameter is estimated from the measurement outcome and knowledge of the system and Hamiltonian. In most problems, the Hamiltonian is assumed to act in the same way on each of \( N \) systems (e.g. atoms or photons), and precision scales as \( \delta x \propto N^{-1/2} \) for product states, and down to \( \delta x \propto N^{-1} \) for entangled states (‘Heisenberg limit’ scaling) \([1]\). A number of studies \([2]–[4]\) have considered also nonlinear quantum metrology, in which the Hamiltonian describes a \( k \)-system coupling with strength \( x' \).

Remarkably, the scaling is \( \delta x' \propto N^{-k+1/2} \) or \( \delta x' \propto N^{-k} \) for independent or entangled states, respectively \([2]\). Because this improves upon the best possible scaling for the linear case, it has been called ‘super-Heisenberg’ (SH) scaling \([4]\). Proposed implementations include scattering in Bose condensates \([4]\), Duffing nonlinearity in nano-mechanical resonators \([5]\), two-pass effective nonlinearity with an atomic ensemble \([6]\) and Kerr nonlinearities \([7]–[9]\).

Here we describe nonlinear metrology applied to the measurement of collective spin variables of atomic ensembles. Atomic ensembles with long coherence-time internal degrees of freedom, e.g. nuclear spin, are essential elements of many quantum information and quantum metrology protocols including quantum memory \([10]\), quantum non-demolition (QND) measurement \([11]\), spin squeezing \([12]–[14]\) and magnetometry \([15]\). Systems to interact atomic ensembles with optical probes, if they achieve quantum-noise-limited performance in both the atomic and optical domains, have been called ‘quantum interfaces’ between light and matter \([16]\). The terminology comes from quantum memory, where the goal is controlled and bi-directional transfer of quantum information between the two systems.

We imagine a quantum interface consisting of a macroscopic number of cold, trapped alkali atoms, through which pass optical pulses containing a macroscopic number of photons. The light is tuned sufficiently far from the atomic resonances to experience little absorption but close enough that optical nonlinearities are resonantly enhanced. Fast nonlinearities such as ac-Stark shifts and stimulated Raman transitions create atomic-spin-state-dependent interactions among the photons. The polarization state of the photons evolves in response to this interaction and is measured, allowing the spin polarization to be estimated.

In the following, we first describe the structure of the problem, in terms of the collective atomic polarization \( \mathbf{F} \) and the collective polarization of the optical pulses, described by the Stokes operators \( \mathbf{S} \). A nonlinear effective Hamiltonian \( H_{\text{eff}} \) for the interaction of these two systems is then derived, using the degenerate perturbation theory. This Hamiltonian contains several terms with very different dependence on detuning, which allows a number of proposed quantum metrology scenarios to be implemented. A scenario employing interactions but not entanglement, previously considered by Boixo et al \([4]\) is analyzed using the continuous-variable quantum noise theory. The scenario gives SH scaling in the estimation of the atomic spin polarization.

We follow the approach of collective continuous variables (CCV), in which both light and atoms are described by macroscopic quantum variables. In the case of spinor atoms interacting with polarized light, the \( N_A \) atoms are described by the collective spin \( \mathbf{F} = \sum_i \mathbf{f}^{(i)} \), where \( \mathbf{f}^{(i)} \) is the spin of the \( i \)th atom. \( \mathbf{F} \) obeys the commutation relations \([F_x, F_y] = i\hbar F_z\) and cyclic permutations, and can itself be considered a macroscopic spin variable. The light is described by its electric field \( \mathbf{E} = \mathcal{E} + \mathcal{E}^\ast \), where \( \mathcal{E} \) is the positive-frequency part. The Stokes
vector $\mathbf{S}$ with components $S_i = (E_s^+, E_s^-)\sigma_i (E_s^+, E_s^-)^T$ where the subscript indicates plus/minus circular polarization, $\sigma_i$ are the Pauli matrices and $\sigma_0$ is the identity. As described by several authors [17]–[19], the electric dipole interaction $h_{\text{int}} = -\mathbf{E} \cdot \mathbf{d}$, taken in the second-order perturbation theory, gives rise to an effective (single-atom) Hamiltonian of the form

$$h_{\text{eff}} = \sum_k \frac{\mathcal{E}^* \cdot \mathbf{d}_\uparrow |\phi_k\rangle \langle \phi_k| \mathbf{d}_\downarrow \cdot \mathcal{E}}{\hbar \delta_k}$$

or

$$= \mathcal{E}^* \cdot \vec{\alpha} \cdot \mathcal{E},$$

where $d_\uparrow$ and $d_\downarrow$ are the parts of the dipole operator causing upward/downward transitions, $\delta_k$ is the detuning from resonance of the $k$th state, and $\vec{\alpha}$ is the tensor polarizability operator. The effective Hamiltonian for the ensemble $H_{\text{eff}} = \sum_i h_{\text{eff}}^{(i)}$ can then be decomposed into irreducible tensor components as

$$H_{\text{eff}}^{(2)} = \alpha^{(1)}(S_z J_z + \alpha^{(2)} (S_x J_x + S_y J_y),$$

plus terms in $S_0$ that do not alter the optical polarization. Here $\alpha^{(1)}$ and $\alpha^{(2)}$ describe the vectorial and tensorial components of the interaction, respectively, and the atomic collective variable is $J \equiv \sum_i j^{(i)}$ with $j_x \equiv (f_x^2 - f_y^2)/2$, $j_y \equiv (f_x f_y + f_y f_x)/2$, $j_z \equiv f_z/2$ and $j_0 \equiv f_z/2$. The ratio of the $\alpha^{(i)}$ can be tuned by adjusting the optical frequency $\omega$, giving a variety of Hamiltonians interesting for quantum information tasks [19].

To apply this formalism to nonlinear metrology, we generalize the CCV method to the nonlinear optics regime, i.e. we include higher-order processes in the effective Hamiltonian. For this purpose, naïve application of the higher-order perturbation theory fails due to the appearance of vanishing resonance denominators, and the degenerate perturbation theory [20] is required. Here we describe the method and its main results; in the appendix we show a fully worked example using the $D_2$ line of $^{87}$Rb, one of the most used transitions for atom–light interactions.

We consider the ground and excited states of a dipole-allowed transition in an alkali atom, which are indexed by their hyperfine ($F$) and magnetic ($m_F$) quantum numbers. The unperturbed energies are described by the (single-atom) atomic Hamiltonian $h_0$. In the alkali atoms, the ground state hyperfine splitting is large compared to the excited state hyperfine splittings, and we identify one $F$ manifold as a $(2F+1)$-fold degenerate subspace for the purposes of the perturbation theory. The perturbation theory will give the effective Hamiltonian describing interaction of atoms initially in these states with an applied optical field $\mathbf{E}$. Writing $h_0$ in a frame rotating at the optical frequency, we obtain a matrix representation of the unperturbed Hamiltonian in terms of the detunings $\delta_F$ from the various excited-state hyperfine levels $F'$. As the perturbation, we consider the electric dipole interaction $h_{\text{int}} = -\mathbf{E} \cdot \mathbf{d}$, in the rotating-wave approximation. This can be written as a matrix $V$ containing $\mathcal{E}_\pm$, the circular-plus-minus components of the envelope of $\mathbf{E}$. Degenerate perturbation theory provides a closed-form expression for the effective Hamiltonian, expressed as an expansion in orders of $V$ or equivalently of $\mathcal{E}_\pm$. As in the usual linear treatment, we express the result as a polynomial in Stokes operators and collective (pseudo-) spin operators. The second-order contribution $H_{\text{eff}}^{(2)}$ is given above, and for $F = 1$ the fourth-order contribution is

$$H_{\text{eff}}^{(4)} = \beta_j^{(0)} S_z^2 J_0 + \beta_N^{(0)} S_z^2 N_A + \beta^{(1)} S_0 S_z J_z + \beta^{(2)} S_0 (S_x J_x + S_y J_y).$$
Figure 1. Spectra of the terms of the effective Hamiltonian. The first two curves from the top, the left axis: continuous, $\alpha^{(1)}$; dashed, $\alpha^{(2)}$. Lower curves, the right axis: continuous, $\beta^{(1)}$; dotted, $\beta^{(0)}_N$; dashed, $\beta^{(2)}$; dot-dashed, $\beta^{(0)}_J$. Detuning (MHz) is relative to the transition $F = 1 \rightarrow F' = 0$ of $^{87}\text{Rb}$ $D_2$ transition. Points A and B indicate detunings at which $\alpha^{(1)}$ or $\beta^{(1)}$ vanish.

The coefficients $\beta$ depend strongly on the probe frequency due to the excited state hyperfine structure. The exact expressions for the $\alpha$ and $\beta$ coefficients for the $D_2$ line of $^{87}\text{Rb}$, from the $F = 1$ ground state, are calculated in the appendix and are shown graphically in figure 1.

2. Application to nonlinear metrology

Terms containing the parameters $\beta$ are nonlinear in $S$, indicating photon–photon interactions. We expect these terms to describe polarization effects of fast electronic nonlinearities including saturation and four-wave mixing. As in the linear case, the frequency dependence of the nonlinear terms provides considerable flexibility in designing a light–matter interaction. Applied to quantum metrology, these terms produce SH scaling, because they are nonlinear in the $S$ collective variables, while the atomic variables $J_i, N_A$ play the role of the parameter. The terms containing $\beta^{(0)}$ and $\beta^{(1)}$ are analogous to Hamiltonians considered by Boixo et al [2]. The term containing $\beta^{(1)}$, which is $\propto S_0 S_z$, in particular, achieves SH scaling without input or generated entanglement [4]. The term containing $\beta^{(2)}$ describes a nonlinear tensorial contribution, and does not appear to have been considered yet for nonlinear metrology.

3. Quantum noise

To understand the quantum noise in this system, we use standard techniques from the CCV literature [21, 22]. We define polarization operators $\hat{S}_i \equiv \frac{1}{2}(a_+^\dagger, a_-^\dagger)\sigma_i(a_+, a_-)^\dagger$, where $a_\pm$ are annihilation operators for the $\pm$ circular polarizations of a mode defined by the pulse shape.
These obey angular momentum commutation relations $[\hat{S}_i, \hat{S}_j] = i\varepsilon_{ijk}\hat{S}_k$ and are related to the Stokes parameters by $\hat{S}_i = S_i/2\gamma$, where $\gamma \equiv \hbar\omega Z_0/2TA$ is the single-photon intensity, $T$ is the pulse duration, $A$ is the beam area and $Z_0$ is the impedance of free space. The average number of photons is $N_L = 2\langle \hat{S}_0 \rangle$. For a typical input, a coherent state, $\langle (\hat{S}_x, \hat{S}_y, \hat{S}_z) \rangle = (N_L/2, 0, 0)$ and $\text{var}(\hat{S}_i) = N_L/4$.

Evolution under this effective Hamiltonian produces, to first order in the interaction time $\tau$,

$$\hat{S}_y^{(\text{out})} = \hat{S}_y^{(\text{in})} + \frac{\tau}{\hbar} (\alpha^{(1)} + \beta^{(1)} \gamma \hat{S}_0) \gamma \hat{S}_x J_z^{(\text{in})}$$

(5)

plus terms containing $\hat{S}_z^{(\text{in})} J_x^{(\text{in})}$ that are negligible for the given input coherent state of the light. This evolution physically corresponds to a paramagnetic Faraday rotation of the input linear polarization. In a metrological scheme, one would measure this polarization rotation and from it estimate the atomic variable $J_z$.

For small rotation on the Poincaré sphere, i.e. $\phi \equiv \hat{S}_y^{(\text{out})}/\hat{S}_x^{(\text{in})} = \gamma \tau \hbar^{-1} (\alpha^{(1)} + \beta^{(1)} \gamma \hat{S}_0) \times J_z^{(\text{in})} \ll 1$, we note that the input polarization noise dominates: $\text{var}(\hat{S}_y^{(\text{out})}) = \text{var}(\hat{S}_y^{(\text{in})}) + \phi^2 \text{var}(\hat{S}_z) \approx \text{var}(\hat{S}_y^{(\text{in})}) = N_L/4$, and that the signal-to-noise ratio equals one when $\langle \hat{S}_y^{(\text{out})} \rangle^2 = \text{var}(\hat{S}_y^{(\text{in})})$, i.e. when

$$\frac{\tau^2 \gamma^2 N_L^2}{4} \left( \alpha^{(1)} + \beta^{(1)} \gamma \frac{N_L}{2} \right)^2 (J_z^{(\text{in})})^2 = \frac{N_L}{4}.$$  

(6)

We can identify the value of $J_z^{(\text{in})}$ that solves equation (6) as the sensitivity, or precision of the estimation, $\delta J_z$. We find

$$\delta J_z = \hbar \left| \tau \gamma \left( \alpha^{(1)} N_L^{1/2} + \beta^{(1)} \gamma \frac{N_L^3}{2} \right) \right|^{-1}.$$  

(7)

Thus, the sensitivity will have a transition from shot noise to SH scaling with increasing $N_L$. As indicated in figure 1, there are points in the spectrum where either $\alpha^{(1)}$ or $\beta^{(1)}$ vanishes, allowing pure nonlinear or pure linear estimation of the same atomic variable.

In another scenario, an unpolarized input state $\langle (\hat{S}_x, \hat{S}_y, \hat{S}_z) \rangle = (0, 0, 0)$ gives rise to dynamics dominated by the $\beta^{(0)}$ terms $\propto \hat{S}_z^2$, sometimes called the ‘one-axis twisting Hamiltonian’. This describes a self-rotation of the optical polarization, and can be used to generate polarization squeezing and also to obtain sensitivity scaling as $N_L^{-3/2}$ in the estimation of $\beta^{(0)} J_0 + \beta^{(0)} N_A$, using an entanglement-generating strategy described in [4].

The physical mechanisms generating the nonlinear signal are similar to those already demonstrated for optical QND measurements [23]. In that experiment, near-resonant beams containing macroscopic numbers of photons experienced dispersive nonlinearities in cold atomic ensembles, due to light-induced level shifts, in good agreement with theory [24]. While the extrapolation of these techniques to the single-photon level may be impossible for fundamental reasons [25, 26], the nonlinear metrology we analyze here moves in the opposite direction, toward larger photon numbers.
4. Conclusion

We have generalized the formalism of continuous collective variables to the nonlinear regime. The resulting nonlinear effective Hamiltonian includes several distinct nonlinear couplings with strengths widely tunable via the probe light frequency. This allows the production of model Hamiltonians proposed for nonlinear metrology, including both models that generate entanglement and those which achieve SH scaling without entanglement. Similar nonlinear probing techniques could improve optical probing of atomic clocks [13, 27] and atomic magnetometers [15, 28, 29]. Unlike previous proposals, the atomic ensemble system allows both linear and nonlinear estimation of the same atomic variables.

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Appendix. \( F = 1 \) effective Hamiltonian for the D\(_2\) line of \( ^{87}\text{Rb} \)

We consider the \( 5^2\text{S}_{1/2} \rightarrow 5^2\text{P}_{3/2} \) transition (the \( D_2 \) transition at 780 nm). The ground states are \(|F, m_F\rangle\) with \((F, m_F) = (1, -1), (1, 0), (1, 1), (2, -2), \ldots, (2, 2)\). The excited states are \(|F', m_{F'}\rangle\) with \((F', m_{F'}) = (0, 0), (1, -1), (1, 1), \ldots, (3, 3)\). We use these states as a basis, with the ground states preceding the excited states. We calculate the single-atom Hamiltonian \( \hat{h}_{\text{eff}} \), and note that the ensemble Hamiltonian \( \hat{H}_{\text{eff}} = \sum_i \hat{h}_{\text{eff}}^{(i)} \) is found simply by replacing single-atom operators such as \( \mathbf{j} \) with collective operators \( \mathbf{J} \equiv \sum_i \mathbf{j}^{(i)} \). The unperturbed Hamiltonian is \( \hat{h}_0 = \hbar \sum_i \omega_i \langle l | l \rangle \), or in matrix notation

\[
\hat{h}_0 = \hbar (\omega_{F=1} I_3 \oplus \omega_{F=2} I_5 \oplus \omega_{F=0} I_1 \oplus \omega_{F=3} I_3) \oplus \omega_{F=2} I_5 \oplus \omega_{F=3} I_7),
\]

where \( \oplus \) indicates a direct sum, and \( I_d \) is the identity matrix of dimension \( d \). We choose the origin of energy such that \( \omega_{F=1} = 0 \), and define \( \Delta \equiv \omega_{F=2} \). We work in a frame rotating with the laser frequency \( \omega \) and define detunings \( \delta_{F'} \equiv \omega_{F'} - \omega \). In this frame, the Hamiltonian is

\[
\hat{h}_0 = \hbar (0 I_3 \oplus \Delta I_5 \oplus \delta_{0} I_5 \oplus \delta_{1} I_3 \oplus \delta_{2} I_5 \oplus \delta_{3} I_7)
\]

(A.2)

In the rotating wave approximation, the single-atom perturbation \( \nu = \hbar \nu_{\text{int}} = -\mathbf{E} \cdot \mathbf{d} \) is approximated as \( \nu \approx \mathbf{E} \cdot \mathbf{d} \). If \( \mathbf{E}_\pm \) are the amplitudes for the sigma-plus/minus components, respectively, of \( \mathbf{E} \), then

\[
\langle F', m_{F'} | V | F, m_{F} \rangle = \mathbf{E}_q \langle F', m_{F'} | \nu | F, m_{F} \rangle
\]

(A.3)

with \( q = m_{F'} - m_{F} \). Note that \( q = 0 \) transitions (\( \pi \)-transitions) are not considered because the \( z \)-propagating beam cannot contain this polarization. The dipole matrix elements are related to the ‘matrix element’ \( \langle J || \nu || J' \rangle \equiv D_{JJ'} \approx 3.58410^{-29} \text{C m} \) by angular-momentum addition rules. We follow the conventions given by Steck [30]. In this way, we arrive at the perturbation Hamiltonian

\[
V = \begin{pmatrix} 0 & \mathbf{V}^\top \\ \mathbf{V} & 0 \end{pmatrix},
\]

(A.4)
where $0_d = 0I_d$ and

$$V_\perp \equiv \sqrt{5} D_{J'J},$$

(A.5)

To obtain the effective Hamiltonian, we follow Klein [20]. The notation of that work is somewhat obscure, so for ease of understanding we repeat the main results. From equation (A7) of that work, we have the $t$-order contribution to the effective Hamiltonian

$$h_{\text{eff}}^{(t)} = \sum \{k\} A_{\{k\}} O_{\{k\}},$$

(A.6)

where $k_1, \ldots, k_{t-1}$ are non-negative integers, the $A$ are real coefficients, the $O$, denoted as \{$(k_1, k_2, \ldots, k_t)$\} by Klein [20], are operators and the sum is taken over all $\{k\}$ satisfying $\sum_{i=1}^{t-1} k_i = t - 1$. The $A$ are given in table 1 of that work and the $O$ are given in equation (A1) as

$$O_{\{k_1, \ldots, k_{t-1}\}} \equiv P_0 V R^{(k_1)} V R^{(k_2)} \ldots V R^{(k_{t-1})} V P_0,$$

(A.7)

with $P_0$ being the projector onto the degenerate subspace $F = 1$, $m_F = \{-1, 0, +1\}$ and by equation (II.A5),

$$R^{(k)} \equiv \begin{cases} P_0 & k = 0, \\ \left( \frac{1 - P_0}{E_0 - H^{(0)}} \right)^k & k > 0, \end{cases}$$

(A.8)

where $E_0$ is the energy of the degenerate subspace. In our case, we have chosen $E_0 = 0$. 

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We can then directly calculate the second- and fourth-order contributions. We are only concerned with $h_{\text{eff}}$ as it acts on the $F = 1$ subspace, that is, with a $3 \times 3$ matrix, and it is convenient to express it in terms of the pseudo-spin components $j_0, j_x, j_y$ and $j_z$ and the Stokes components $S_0, S_x, S_y$ and $S_z$ defined above. Summing the second-order contributions, we find $H_{\text{eff}}^{(2)}$ of equation (3). With $B \equiv -D_{JJ}^2/48\delta_0\delta_1\delta_2\delta_3h$,

\[
\alpha^{(1)} = B(5\delta_0\delta_1 - 5\delta_0\delta_2 - 4\delta_1\delta_2), \quad \text{(A.9)}
\]

\[
\alpha^{(2)} = B(\delta_0\delta_1 - 5\delta_0\delta_2 + 4\delta_1\delta_2). \quad \text{(A.10)}
\]

Similarly, the fourth-order contribution is, dropping terms in $S_0^2$,

\[
H_{\text{eff}}^{(4)} = \beta_j^{(0)} S_j^2 J_0 + \beta_N^{(0)} S_N^2 N_A + \beta_j^{(1)} S_0 S_j J_z + \beta_j^{(2)} S_0(S_z J_z + S_y J_y). \quad \text{(A.11)}
\]

Note that the term in $N_A$ arises because $h_{\text{eff}}^{(4)}$ contains a self-rotation term of the form $\beta_j^{(0)} S_j^2 P_{m=0}$, where $P_{m=0}$ is a projector onto the state $|F = 1, m_F = 0\rangle$. We express this in terms of $J_0$ and $N_A$ using $\sum_i P_m^{(i)} = 1$, $J_i^{(i)} - J_0^{(i)} = N_A - J_0$.

With $C \equiv D_{JJ}^4/1152\delta_0^3\delta_1^2\delta_2^3\Delta h^3$, the coefficients, shown graphically in figure 1, are

\[
\beta_j^{(0)} = C(12\delta_0^3\delta_1^2\delta_2^2 - 4\delta_0^3\delta_1^2\delta_2 + 12\delta_0^3\delta_1^3\Delta - 10\delta_0^3\delta_1^2\delta_2\Delta - 12\delta_0^3\delta_1\delta_2^2\Delta - 10\delta_0^3\delta_1^2\delta_2\Delta - 12\delta_0^3\delta_1^3\Delta - 12\delta_0^3\delta_1^2\delta_2\Delta - 20\delta_0^3\delta_1\delta_2^2\Delta + 20\delta_0^3\delta_1^2\delta_2\Delta), \quad \text{(A.12)}
\]

\[
\beta_N^{(0)} = C(-12\delta_0^3\delta_1\delta_2 - 24\delta_0^3\delta_1^2\delta_2 + 4\delta_0^3\delta_1\delta_2^2), \quad \text{(A.13)}
\]

\[
\beta^{(1)} = C(-9\delta_0^3\delta_1^3\delta_2 + 6\delta_0^3\delta_1^2\delta_2^2 + 3\delta_0^3\delta_1^2\delta_2^2 + 35\delta_0^3\delta_1^3\Delta - 5\delta_0^3\delta_1^2\delta_2\Delta - 4\delta_0^3\delta_1\delta_2^2\Delta - 5\delta_0^3\delta_1\delta_2^2\Delta - 4\delta_0^3\delta_1^2\delta_2^2 - 25\delta_0^3\delta_1^3\Delta - 20\delta_0^3\delta_1^2\delta_2^2 - 20\delta_0^3\delta_1\delta_2^2\Delta - 16\delta_1^3\delta_2^3\Delta), \quad \text{(A.14)}
\]

\[
\beta^{(2)} = C(3\delta_0^3\delta_1^3\delta_2 - 6\delta_0^3\delta_1^2\delta_2^2 - 25\delta_0^3\delta_1\delta_2^2\Delta + 7\delta_1^3\delta_2^3\Delta - 15\delta_0^3\delta_1^3\delta_2\Delta - 16\delta_0^3\delta_1^2\delta_2\Delta - 15\delta_0^3\delta_1\delta_2^2\Delta + 16\delta_1^3\delta_2^3\Delta - 25\delta_0^3\delta_1^3\Delta + 16\delta_1^3\delta_2^3\Delta). \quad \text{(A.15)}
\]

In order to make it easier for the reader to follow the calculations, the used script is provided as supplementary material, available at stacks.iop.org/NJP/12/093016/mmedia.

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