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To cite this article: Matteo Fadel *et al* 2024 *New J. Phys.* **26** 103037

IOP Publishing New J. Phys. 26 (2024) 103037 https://doi.org/10.1088/1367-2630/ad8953

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Deutsche Physikalische Gesellschaft DPG
IOP Institute of Physics

Published in partnership with: Deutsche Physikalische Gesellschaft and the Institute of Physics

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RECEIVED
6 May 2024

REVISED
23 September 2024

ACCEPTED FOR PUBLICATION
21 October 2024

PUBLISHED
30 October 2024

PAPER

Effective Faraday interaction between light and nuclear spins of helium-3 in its ground state: a semiclassical study

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Keywords: helium-3, atom–light interaction, atomic physics

Abstract

We derive the semiclassical evolution equations for a system consisting of helium-3 atoms in the 2^3S metastable state interacting with a light field far-detuned from the 2^3S-2^3P transition, in the presence of metastability exchange collisions with ground state helium atoms and a static magnetic field. For two configurations, each corresponding to a particular choice of atom–light detuning in which the contribution of either the metastable level $F=1/2$ or $F=3/2$ is dominant, we derive a simple model of three coupled collective spins from which we can analytically extract an effective coupling constant between the collective nuclear spin and light. In these two configurations, we compare the predictions of our simplified model with the full model.

1. Introduction

Helium-3 in its ground state has a purely nuclear spin 1/2. Protected by a complete electronic shell and separated from the first excited state by 20 eV, the nuclear spin of helium is a two-level quantum system that offers exceptionally long coherence times of hundreds of hours [1]. For this reason, helium-3 nuclear spins are used in ultra-sensitive magnetometry in fundamental physics experiments [2–4]. A further improvement in precision, when using helium nuclear spins as a sensor, is in principle possible using quantum entanglement and spin squeezing, especially in the case of small samples where the relative effect of quantum fluctuations is greater. In addition to sensing, the possibility of effectively coupling helium-3 nuclear spins with light, which carries information over long distances and can be measured at the quantum noise level, offers interesting prospects for example for quantum memories. Although the manipulation of the collective nuclear spin of a helium gas at the quantum limit has not yet been achieved experimentally, theoretical and experimental efforts in this direction are ongoing [5–10]. The Faraday interaction between the collective spin of an ensemble of atoms and the Stokes spin of light provides a light-matter quantum interface that has already been demonstrated in the laboratory in the case of alkaline atoms [11–13]. In the case of the purely nuclear spin of rare gases in their ground state, interaction with light requires an intermediate system. For helium-3, it is a small fraction of atoms brought into a metastable state that offers near-infrared transitions and interacts with atoms in the ground state via metastability exchange collisions [1]. In this paper we study in detail the interaction of light with a set of helium-3 atoms, a fraction of which is brought into the metastable state. At the semiclassical level, we explore the validity of the simplified model used in [9, 10], taking into account the full atomic structure in the metastable 2^3S and the excited 2^3P states. Moreover, we propose another possible configuration that should allow for a larger effective coupling between the nuclear spins and the light.

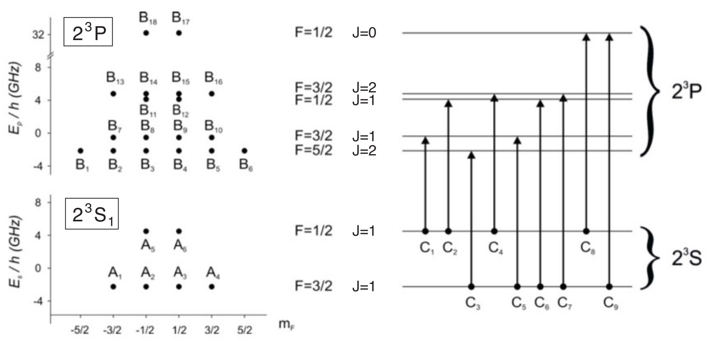

Figure 1. Left: hyperfine structure of states 2^3S_1 and 2^3P in ^3He. Right: allowed transitions at 1083 nm.

Table 1. Frequencies of transitions $C_1 - C_9$ shown in figure 1, and calculated with respect to transition $C_8 = 2\pi\,276\,726\,257$ MHz [14].

	Freq. offset (GHz)	F	J	F'	J'
$\Delta_1/2\pi$	-32.6045	1/2	1	3/2	1
$\Delta_2/2\pi$	-28.0929	1/2	1	1/2	1
$\Delta_3/2\pi$	-27.6453	3/2	1	5/2	2
$\Delta_4/2\pi$	-27.4238	1/2	1	3/2	2
$\Delta_5/2\pi$	-25.8648	3/2	1	3/2	1
$\Delta_6/2\pi$	-21.3532	3/2	1	1/2	1
$\Delta_7/2\pi$	-20.6841	3/2	1	3/2	2
$\Delta_8/2\pi$	0	1/2	1	1/2	0
$\Delta_9/2\pi$	$+6.7397$	3/2	1	1/2	0

2. Light–matter interaction for metastable helium atoms

While the ground state 1^1S_0 of helium-3 is purely nuclear, the metastable state 2^3S_1 has an electronic component and is the starting level for transitions at 1083 nm to the excited states 2^3P. Figure 1(Left) shows the hyperfine structure of the metastable and excited levels. The accessible transitions between levels are shown in figure 1(Right), and the relative frequencies in table 1.

For light propagating along the z-axis, we introduce the components of the Stokes spin in terms of the creation and annihilation operators of a photon polarized in the x or y direction, and in term of the circularly polarized photons creation and annihilation operators $a_1 = (a_x - i a_y)/\sqrt{2}$, $a_2 = (a_x + i a_y)/\sqrt{2}$

$$S_0 = \left(a_x^\dagger a_x + a_y^\dagger a_y\right)/2 = \left(a_1^\dagger a_2 + a_2^\dagger a_1\right)/2 \tag{1}$$

$$S_x = \left(a_x^\dagger a_x - a_y^\dagger a_y\right)/2 = \left(a_1^\dagger a_1 + a_2^\dagger a_2\right)/2 \tag{2}$$

$$S_y = \left(a_x^\dagger a_y + a_y^\dagger a_x\right)/2 = \left(a_1^\dagger a_2 - a_2^\dagger a_1\right)/2i \tag{3}$$

$$S_z = \left(a_x^\dagger a_y - a_y^\dagger a_x\right)/2i = \left(a_2^\dagger a_2 - a_1^\dagger a_1\right)/2. \tag{4}$$

In the weak saturation regime, the interaction of light with atoms in either one of the two states $F = 1/2$ or $F = 3/2$ of metastable helium can be described by an effective Hamiltonian obtained by adiabatically eliminating the optical coherences and the populations of the excited state [15–17]. The general form of the effective Hamiltonian for one atom of spin F with light a is recalled in appendix A.

2.1. Effective atom–light interaction in the metastable state

In the case of the metastable state 2^3S of helium-3, we first introduce the collective operators $\vec{K}$, $\vec{J}$ and T^l_m, respectively obtained by summing the single atom spin operators in the $F = 1/2$, and spin and tensor operators in the $F = 3/2$ manifolds of the metastable state. Considering the transitions to the excited states 2^3P, in the case of large detuning, the effective light–atoms Hamiltonian for the ensemble then takes the form

$$H_{\rm LA} = H_{1/2} + H^V_{3/2} + H^T_{3/2} \tag{5}$$

where the two vectorial contributions, of the $F = 1/2$ metastable state ($\vec{K}$) and of the $F = 3/2$ metastable state ($\vec{J}$) take the Faraday form

$$H_{1/2} = \hbar\chi K_z S_z \quad H^V_{3/2} = \hbar\eta J_z S_z, \tag{6}$$

and the tensorial contribution of the $F = 3/2$ metastable state takes the form

$$\begin{aligned} H^T_{3/2} &= \sum_{i=1}^{n} \hbar\mu \left[\left(\frac{F_i(F_i+1)}{3} - F^2_{i,z}\right) S_0 + \left(F^2_{i,x} - F^2_{i,y}\right) S_x + \left(F_{i,x}F_{i,y} + F_{i,y}F_{i,x}\right) S_y\right] \\ &= \hbar\mu \left[-2T^2_0 S_0 + \sqrt{12}\left(\Re T^2_2 S_x + \Im T^2_2 S_y\right)\right]. \end{aligned} \tag{7}$$

In the above, we used the collective irreducible tensor operators T^2_0, $\Re T^2_2$ and $\Im T^2_2$, that are obtained by summing the single-atom tensor operators defined in appendix A.

The constants χ, η and μ representing the strength of the different contributions have the form

$$\chi = \frac{\sigma_2}{4A}\Gamma\left(\frac{2}{9(\Delta_p - \Delta_1)} - \frac{8}{9(\Delta_p - \Delta_2)} + \frac{10}{9(\Delta_p - \Delta_4)} - \frac{4}{9\Delta_p}\right) \tag{8}$$

$$\eta = \frac{\sigma_2}{4A}\Gamma\left(\frac{3}{5(\Delta_p - \Delta_3)} - \frac{2}{9(\Delta_p - \Delta_5)} - \frac{1}{9(\Delta_p - \Delta_6)} - \frac{2}{45(\Delta_p - \Delta_7)} - \frac{2}{9(\Delta_p - \Delta_9)}\right) \tag{9}$$

$$\mu = \frac{\sigma_2}{10A}\Gamma\left(-\frac{1}{4(\Delta_p - \Delta_3)} + \frac{5}{9(\Delta_p - \Delta_5)} - \frac{5}{36(\Delta_p - \Delta_6)} + \frac{1}{9(\Delta_p - \Delta_7)} - \frac{5}{18(\Delta_p - \Delta_9)}\right). \tag{10}$$

In these equations, $\sigma_2 = 3\lambda^2/2\pi$, A is the cross sectional area of the light mode, $\Gamma \approx 10^7\,\mathrm{s}^{-1}$ is the excited-state spontaneous decay rate, and taking the C_8 transition as a reference, we have defined $\Delta_p = \omega_{\rm probe} - \omega_{C_8}$ and $\Delta_i = \omega_{FF'} - \omega_{C_8}$. In figure 2 we represent the three coupling constants χ (8), η (9) and μ (10), all divided by the constant $4A/(\sigma_2\Gamma)$, as a function of light frequency. For $\Delta_p/(2\pi) = -2\,\mathrm{GHz}$, which is the operating point considered in [9, 10] and marked as 'Config.1' in figure 2, the vector contribution of $F = 1/2$ is dominant. A second interesting operating point, marked as 'Config.2' in figure 2, is for $\Delta_p/(2\pi) = -31\,\mathrm{GHz}$, around the local minimum of the grey absorption curve where the tensor part of $F = 3/2$ is relatively small. The main advantage of this configuration is that one could work with a highly polarized state, $M \simeq 1$, for which the $F = 1/2$ spin manifold is empty and the initial state is effectively a spin coherent state [19].

2.2. Metastable atomic variables evolution due to the interaction with the light

Due to the Hamiltonian (5), we find from $\mathrm{d}O/\mathrm{d}t = i[H, O]/\hbar$ that the Stokes operators of the light and the collective atomic variable obey the following equation of motion

$$\left.\frac{\mathrm{d}S_x}{\mathrm{d}t}\right|_{\rm L} = -\chi K_z S_y - \eta J_z S_y + \sqrt{12}\mu \Im T^2_2 S_z \tag{11}$$

$$\left.\frac{\mathrm{d}S_y}{\mathrm{d}t}\right|_{\rm L} = \chi K_z S_x + \eta J_z S_x - \sqrt{12}\mu \Re T^2_2 S_z \tag{12}$$

$$\left.\frac{\mathrm{d}S_z}{\mathrm{d}t}\right|_{\rm L} = \sqrt{12}\mu\left(\Re T^2_2 S_y - \Im T^2_2 S_x\right) \tag{13}$$

$$\left.\frac{\mathrm{d}K_x}{\mathrm{d}t}\right|_{\rm L} = -\chi K_y S_z \tag{14}$$

$$\left.\frac{\mathrm{d}K_y}{\mathrm{d}t}\right|_{\rm L} = \chi K_x S_z \tag{15}$$

$$\left.\frac{\mathrm{d}K_z}{\mathrm{d}t}\right|_{\rm L} = 0 \tag{16}$$

$$\left.\frac{\mathrm{d}J_x}{\mathrm{d}t}\right|_{\rm L} = -\eta J_y S_z + \sqrt{12}\mu\left(\Im T^2_1 (S_x - S_0) - \Re T^2_1 S_y\right) \tag{17}$$

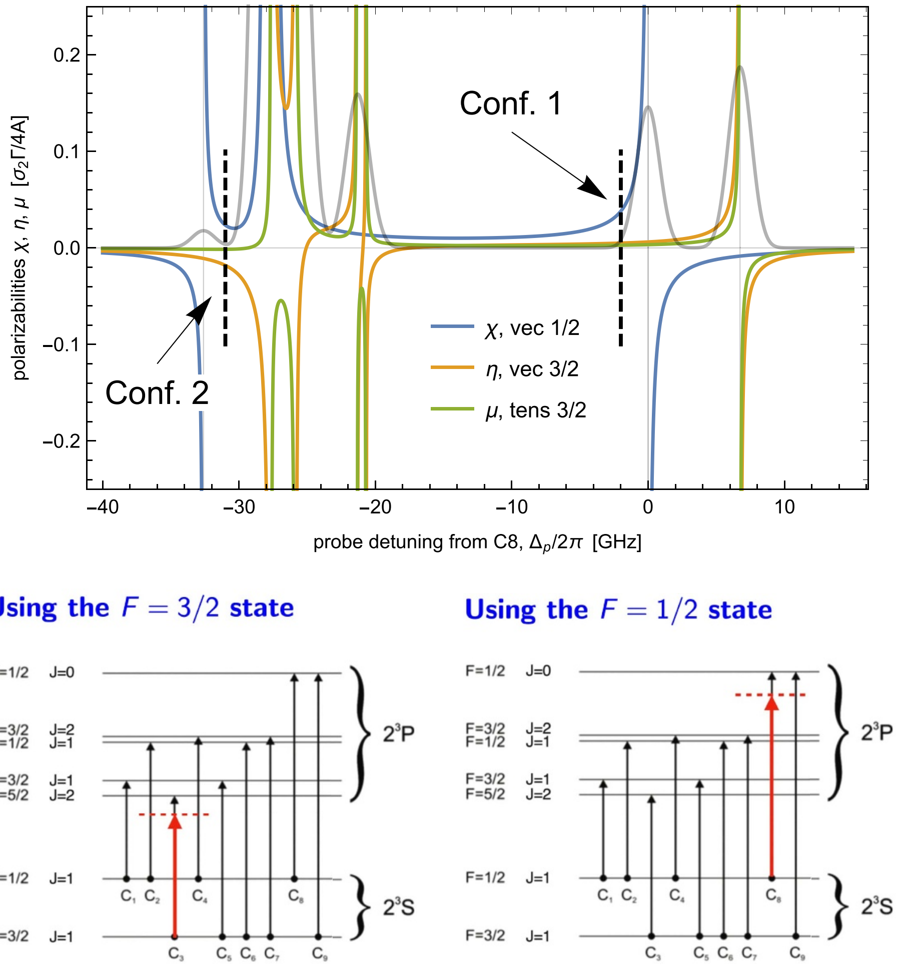

Figure 2. Top: coupling constants χ equation (8) (blue line), η equation (9) (orange line) and μ equation (10) (green line) for the $F = 1/2$ and $F = 3/2$ levels of the ^{3}He metastable, as a function of the light frequency detuning Δ_p taking the C_8 transition as the origin. All the constants are divided by the constant $4A/(\sigma_2\Gamma)$. The grey line shows the absorption spectrum taking into account the Doppler broadening for $T = 300$ K for a non polarized sample [18]. Two possible operating point marked as 'Config.2' at $\Delta_p/(2\pi) = -31$ GHz and 'Config.1' at $\Delta_p/(2\pi) = -2$ GHz are discussed in the text. Bottom: Level scheme depicting the two possible operating point marked as 'Config.2' and 'Config.1', respectively.

$$\left.\frac{\mathrm{d}J_y}{\mathrm{d}t}\right|_{\mathrm{L}} = \eta J_x S_z + \sqrt{12}\mu\left(\Re T_1^2\left(S_x + S_0\right) + \Im T_1^2 S_y\right) \tag{18}$$

$$\left.\frac{\mathrm{d}J_z}{\mathrm{d}t}\right|_{\mathrm{L}} = 2\sqrt{12}\mu\left(\Im T_2^2 S_x - \Re T_2^2 S_y\right) \tag{19}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\Re T_2^2\right|_{\mathrm{L}} = -2\eta S_z \Im T_2^2 + \sqrt{12}\mu\left(\frac{1}{\sqrt{5}}S_y\left(2T_0^1 - T_0^3\right) + \frac{1}{\sqrt{3}}S_0\Im T_2^3\right) \tag{20}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\Im T_2^2\right|_{\mathrm{L}} = 2\eta S_z \Re T_2^2 - \sqrt{12}\mu\left(\frac{1}{\sqrt{5}}S_x\left(2T_0^1 - T_0^3\right) + \frac{1}{\sqrt{3}}S_0\Re T_2^3\right) \tag{21}$$

$$\begin{aligned}\left.\frac{\mathrm{d}}{\mathrm{d}t}\Re T_1^2\right|_{\mathrm{L}} = &-\eta S_z \Im T_1^2 + \sqrt{6}\mu\left(S_x\left(-\sqrt{\frac{3}{5}}\Im T_1^3 - \frac{\sqrt{2}}{5}J_y + \Im T_3^3\right) + S_0\left(\frac{2}{\sqrt{15}}\Im T_1^2 - \frac{\sqrt{2}}{5}J_y\right)\right.\\ &\left. + S_y\left(\sqrt{\frac{3}{5}}\Re T_1^3 + \frac{\sqrt{2}}{5}J_x - \Re T_3^3\right)\right)\end{aligned} \tag{22}$$

$$\begin{aligned}\left.\frac{\mathrm{d}}{\mathrm{d}t}\Im T_1^2\right|_{\mathrm{L}} = &\,\eta S_z \Re T_1^2 + \sqrt{6}\mu\left(S_x\left(\sqrt{\frac{3}{5}}\Re T_1^3 + \frac{\sqrt{2}}{5}J_x + \Re T_3^3\right) + S_0\left(\frac{2}{\sqrt{15}}\Re T_1^2 - \frac{\sqrt{2}}{5}J_x\right)\right.\\ &\left. + S_y\left(\sqrt{\frac{3}{5}}\Im T_1^3 + \frac{\sqrt{2}}{5}J_y + \Im T_3^3\right)\right)\end{aligned} \tag{23}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}T_0^2\right|_{\mathrm{L}} = \sqrt{12}\mu\left(S_x\Im T_2^3 - S_y\Re T_2^3\right) \tag{24}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\Re T_3^3\right|_{\mathrm{L}} = -3\eta S_z \Im T_3^3 + \sqrt{6}\mu\left(S_x \Im T_1^2 + S_y \Re T_1^2\right) \tag{25}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\Im T_3^3\right|_{\mathrm{L}} = 3\eta S_z \Re T_3^3 - \sqrt{6}\mu\left(S_x \Re T_1^2 - S_y \Im T_1^2\right) \tag{26}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\Re T_2^3\right|_{\mathrm{L}} = -2\eta S_z \Im T_2^3 + 2\mu\left(\sqrt{3}S_y T_0^2 + S_0 \Im T_2^2\right) \tag{27}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\Im T_2^3\right|_{\mathrm{L}} = 2\eta S_z \Re T_2^3 - 2\mu\left(\sqrt{3}S_x T_0^2 + S_0 \Re T_2^2\right) \tag{28}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\Re T_1^3\right|_{\mathrm{L}} = -\eta S_z \Im T_1^3 + \sqrt{\frac{2}{5}}\mu\left(\left(2S_0 + 3S_x\right)\Im T_1^2 - 3S_y \Re T_1^2\right) \tag{29}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\Im T_1^3\right|_{\mathrm{L}} = \eta S_z \Re T_1^3 - \sqrt{\frac{2}{5}}\mu\left(\left(2S_0 - 3S_x\right)\Re T_1^2 - 3S_y \Im T_1^2\right) \tag{30}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}T_0^3\right|_{\mathrm{L}} = -\frac{2}{5}\sqrt{15}\mu\left(S_x \Im T_2^2 - S_y \Re T_2^2\right) \tag{31}$$

3. External magnetic field

In the presence of a static magnetic field $\vec{B}$, the system evolves according to the Hamiltonian

$$H_B = -\hbar\left(\gamma_{1/2}\vec{B}\cdot\vec{K} + \gamma_{3/2}\vec{B}\cdot\vec{J} + \gamma_{\mathrm{nuc}}\vec{B}\cdot\vec{I}\right), \qquad \text{where} \tag{32}$$

$$\gamma_{1/2} = \frac{4}{3}\gamma_{\mathrm{ms}}; \quad \gamma_{3/2} = \frac{2}{3}\gamma_{\mathrm{ms}}; \quad \gamma_{\mathrm{ms}} = -2\pi\, 2.802\,\mathrm{MHz\,G^{-1}}; \quad \gamma_{\mathrm{nuc}} = -2\pi\, 3.243\,\mathrm{kHz\,G^{-1}}; \tag{33}$$

are the gyromagnetic ratios [20].

3.1. Metastable atomic variables evolution due to an external magnetic field

The corresponding equations of motion for atomic variables are given by

$$\left.\frac{\mathrm{d}\vec{I}}{\mathrm{d}t}\right|_{B} = \gamma_{\mathrm{nuc}}\,\vec{I}\times\vec{B} \tag{34}$$

$$\left.\frac{\mathrm{d}\vec{K}}{\mathrm{d}t}\right|_{B} = \gamma_{1/2}\,\vec{K}\times\vec{B} \tag{35}$$

$$\left.\frac{\mathrm{d}\vec{J}}{\mathrm{d}t}\right|_{B} = \gamma_{3/2}\,\vec{J}\times\vec{B} \tag{36}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\Re T_2^2\right|_{B} = \gamma_{3/2}\left(B_x \Im T_1^2 + B_y \Re T_1^2 + 2B_z \Im T_2^2\right) \tag{37}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\Im T_2^2\right|_{B} = \gamma_{3/2}\left(-B_x \Re T_1^2 + B_y \Im T_1^2 - 2B_z \Re T_2^2\right) \tag{38}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\Re T_1^2\right|_{B} = \gamma_{3/2}\left(B_x \Im T_2^2 + B_y\left(\sqrt{3}T_0^2 - \Re T_2^2\right) + B_z \Im T_1^2\right) \tag{39}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\Im T_1^2\right|_{B} = \gamma_{3/2}\left(-B_x\left(\sqrt{3}T_0^2 + \Re T_2^2\right) - B_y \Im T_2^2 - B_z \Re T_1^2\right) \tag{40}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}T_0^2\right|_{B} = \gamma_{3/2}\sqrt{3}\left(B_x \Im T_1^2 - B_y \Re T_1^2\right) \tag{41}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\Re T_3^3\right|_{B} = \gamma_{3/2}\left(B_x\sqrt{\frac{3}{2}}\Im T_2^3 + B_y\sqrt{\frac{3}{2}}\Re T_2^3 + B_z 3\Im T_3^3\right) \tag{42}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\Im T_3^3\right|_{B} = \gamma_{3/2}\left(-B_x\sqrt{\frac{3}{2}}\Re T_2^3 + B_y\sqrt{\frac{3}{2}}\Im T_2^3 - B_z 3\Re T_3^3\right) \tag{43}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\Re T_2^3\right|_{B} = \gamma_{3/2}\left(B_x\left(\sqrt{\frac{10}{4}}\Im T_1^3 + \sqrt{\frac{3}{2}}\Im T_3^3\right) + B_y\left(\sqrt{\frac{10}{4}}\Re T_1^3 - \sqrt{\frac{3}{2}}\Re T_3^3\right) + B_z 2\Im T_2^3\right) \tag{44}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\Im T_2^3\right|_{B} = \gamma_{3/2}\left(B_x\left(-\sqrt{\frac{10}{4}}\Re T_1^3 - \sqrt{\frac{3}{2}}\Re T_3^3\right) + B_y\left(\sqrt{\frac{10}{4}}\Im T_1^3 - \sqrt{\frac{3}{2}}\Im T_3^3\right) - B_z 2\Re T_2^3\right) \tag{45}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\mathfrak{R}T_1^3\right|_B = \gamma_{3/2}\left(B_x\sqrt{\frac{5}{2}}\mathfrak{I}T_2^3 + B_y\left(\sqrt{6}T_0^3 - \sqrt{\frac{5}{2}}\mathfrak{R}T_2^3\right) + B_z\mathfrak{I}T_1^3\right) \tag{46}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\mathfrak{I}T_1^3\right|_B = \gamma_{3/2}\left(-B_x\left(\sqrt{6}T_0^3 - \sqrt{\frac{5}{2}}\mathfrak{R}T_2^3\right) - B_y\sqrt{\frac{5}{2}}\mathfrak{I}T_2^3 - B_z\mathfrak{R}T_1^3\right) \tag{47}$$

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}T_0^3\right|_B = \gamma_{3/2}\sqrt{6}\left(B_x\mathfrak{I}T_1^2 - B_y\mathfrak{R}T_1^2\right). \tag{48}$$

Note that in this work we are interested in a weak magnetic field regime, where the metastability exchange collision rates are weakly affected.

4. Metastability exchange collisions

4.1. Evolution of the one-body density matrix

Metastability exchange collisions (MEC) are fast processes in which the electronic excitation is transferred from a metastable to a ground state helium atom, without affecting the electronic and nuclear spin variables [1, 21]. They are usually described in terms of the one-body density matrix ρ which is assumed to be block-diagonal, with the 2×2 matrix ρ_f describing the ground state and the 6×6 matrix ρ_m describing the metastable state. Following a collision, ρ transforms according to $\rho \xrightarrow{\mathrm{MEC}} \rho'$ with [6, 20]

$$\rho_\mathrm{f}' = \mathrm{Tr}_\mathrm{e}\left[\rho_\mathrm{m}\right] \tag{49}$$

$$\rho_\mathrm{m}' = \rho_\mathrm{f}\otimes \mathrm{Tr}_\mathrm{n}\left[\rho_\mathrm{m}\right] \tag{50}$$

where Tr_e and Tr_n denote the trace on the electronic and nuclear degrees of freedom respectively. Note that the electronic degrees of freedom do not appear in ρ_f since the ground state is a singlet state, *i.e.* $S = 0$.

Considering a set of N_cell atoms in the ground state and n_cell in the metastable state, the equations of motion of the one-body density matrix are written

$$\frac{\mathrm{d}}{\mathrm{d}t}\rho_\mathrm{f} = \frac{1}{T}\left(-\rho_\mathrm{f} + \mathrm{Tr}_\mathrm{e}\left[\rho_\mathrm{m}\right]\right) \tag{51}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\rho_\mathrm{m} = \frac{1}{\tau}\left(-\rho_\mathrm{m} + \rho_\mathrm{f}\otimes\mathrm{Tr}_\mathrm{n}\left[\rho_\mathrm{m}\right]\right) \tag{52}$$

where the two collision rates $\gamma_\mathrm{f} = 1/T$ et $\gamma_\mathrm{m} = 1/\tau$ for an atom in the ground and metastable states, respectively, satisfy the relation

$$\frac{\gamma_\mathrm{m}}{\gamma_\mathrm{f}} = \frac{T}{\tau} = \frac{N_\mathrm{cell}}{n_\mathrm{cell}}. \tag{53}$$

Such equations of motion, expressed in the $\{|i\rangle\}$ basis of the Zeeman sublevels of helium-3, are found in [6]. They allow us to calculate the evolution due to the exchange of any one-body atomic operator O

$$\left.\frac{\mathrm{d}\langle O\rangle}{\mathrm{d}t}\right|_\mathrm{MEC} = \mathrm{Tr}\left[O\left.\frac{\mathrm{d}\rho}{\mathrm{d}t}\right|_\mathrm{MEC}\right]. \tag{54}$$

A detailed example explaining how we proceed to obtain the equations in given in appendix C.

4.2. Atomic variables evolution due to metastability exchange

For the three spins: ground I, metastable $(F = 1/2)$ K and metastable $(F = 3/2)$ J, we obtain the semi-classical equations

$$\left.\frac{\mathrm{d}\left\langle\vec{I}\right\rangle}{\mathrm{d}t}\right|_\mathrm{MEC} = -\frac{1}{T}\left\langle\vec{I}\right\rangle + \frac{1}{3T}\frac{N}{n}\left(\left\langle\vec{J}\right\rangle - \left\langle\vec{K}\right\rangle\right) \tag{55}$$

$$\left.\frac{\mathrm{d}\left\langle\vec{K}\right\rangle}{\mathrm{d}t}\right|_\mathrm{MEC} = -\frac{7}{9\tau}\left\langle\vec{K}\right\rangle + \frac{1}{9\tau}\left\langle\vec{J}\right\rangle - \frac{1}{9\tau}\frac{n}{N}\left\langle\vec{I}\right\rangle - \frac{4}{3\tau}\frac{1}{N}\left\langle\vec{\vec{Q}}\right\rangle\cdot\left\langle\vec{I}\right\rangle \tag{56}$$

$$\left.\frac{\mathrm{d}\left\langle\vec{J}\right\rangle}{\mathrm{d}t}\right|_\mathrm{MEC} = -\frac{4}{9\tau}\left\langle\vec{J}\right\rangle + \frac{10}{9\tau}\left\langle\vec{K}\right\rangle + \frac{10}{9\tau}\frac{n}{N}\left\langle\vec{I}\right\rangle + \frac{4}{3\tau}\frac{1}{N}\left\langle\vec{\vec{Q}}\right\rangle\cdot\left\langle\vec{I}\right\rangle, \tag{57}$$

where we have introduced the collective alignment tensor $\langle Q_{\alpha\beta}\rangle = \sum_{i=1}^{N} \frac{1}{3}\frac{1}{6}\left(\frac{3}{2}\langle F_{i,\alpha}F_{i,\beta} + F_{i,\beta}F_{i,\alpha}\rangle - 2\delta_{\alpha\beta}\right)$ [20]

$$Q_{xx} = \frac{1}{6}\left(\sqrt{3}\Re T_2^2 - T_0^2\right) \tag{58a}$$

$$Q_{yy} = -\frac{1}{6}\left(\sqrt{3}\Re T_2^2 + T_0^2\right) \tag{58b}$$

$$Q_{zz} = \frac{1}{3}T_0^2 \tag{58c}$$

$$Q_{xy} = \frac{1}{2\sqrt{3}}\Im T_2^2 \tag{58d}$$

$$Q_{xz} = -\frac{1}{2\sqrt{3}}\Re T_1^2 \tag{58e}$$

$$Q_{yz} = -\frac{1}{2\sqrt{3}}\Im T_1^2. \tag{58f}$$

The rank-2 tensors of state $F = 3/2$ evolve according to equations

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\langle \Re T_2^2\right\rangle\bigg|_{\mathrm{MEC}} = -\frac{2}{3\tau}\left\langle \Re T_2^2\right\rangle + \frac{1}{\sqrt{3}\tau}\frac{1}{N}\left(\langle I_x\rangle\langle \Sigma_x\rangle - \langle I_y\rangle\langle \Sigma_y\rangle\right) \tag{59}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\langle \Im T_2^2\right\rangle\bigg|_{\mathrm{MEC}} = -\frac{2}{3\tau}\left\langle \Im T_2^2\right\rangle + \frac{1}{\sqrt{3}\tau}\frac{1}{N}\left(\langle I_x\rangle\langle \Sigma_y\rangle + \langle I_y\rangle\langle \Sigma_x\rangle\right) \tag{60}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\langle \Re T_1^2\right\rangle\bigg|_{\mathrm{MEC}} = -\frac{2}{3\tau}\left\langle \Re T_1^2\right\rangle + \frac{1}{\sqrt{3}\tau}\frac{1}{N}\left(\langle I_x\rangle\langle \Sigma_z\rangle + \langle I_z\rangle\langle \Sigma_x\rangle\right) \tag{61}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\langle \Im T_1^2\right\rangle\bigg|_{\mathrm{MEC}} = -\frac{2}{3\tau}\left\langle \Im T_1^2\right\rangle + \frac{1}{\sqrt{3}\tau}\frac{1}{N}\left(\langle I_y\rangle\langle \Sigma_z\rangle + \langle I_z\rangle\langle \Sigma_y\rangle\right) \tag{62}$$

$$\frac{\mathrm{d}\left\langle T_0^2\right\rangle}{\mathrm{d}t}\bigg|_{\mathrm{MEC}} = -\frac{2}{3\tau}\left\langle T_0^2\right\rangle + \frac{1}{3\tau}\frac{1}{N}\left(3\langle I_z\rangle\langle \Sigma_z\rangle - \left\langle \vec{I}\right\rangle\cdot\left\langle \vec{\Sigma}\right\rangle\right) \tag{63}$$

where we defined the electron spin operator expectation value in the metastable state $\langle\vec{\Sigma}\rangle = \frac{2}{3}(\langle\vec{J}\rangle + 2\langle\vec{K}\rangle)$.

The rank-3 tensors of state $F = 3/2$ evolve according to equations

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\langle \Re T_3^3\right\rangle\bigg|_{\mathrm{MEC}} = -\frac{1}{\tau}\left\langle \Re T_3^3\right\rangle - \frac{\sqrt{6}}{3\tau}\frac{1}{N}\left(\langle I_x\rangle\left\langle \Re T_2^2\right\rangle - \langle I_y\rangle\left\langle \Im T_2^2\right\rangle\right) \tag{64}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\langle \Im T_3^3\right\rangle\bigg|_{\mathrm{MEC}} = -\frac{1}{\tau}\left\langle \Im T_3^3\right\rangle - \frac{\sqrt{6}}{3\tau}\frac{1}{N}\left(\langle I_x\rangle\left\langle \Im T_2^2\right\rangle - \langle I_y\rangle\left\langle \Re T_2^2\right\rangle\right) \tag{65}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\langle \Re T_2^3\right\rangle\bigg|_{\mathrm{MEC}} = -\frac{1}{\tau}\left\langle \Re T_2^3\right\rangle - \frac{2}{3\tau}\frac{1}{N}\left(\langle I_x\rangle\left\langle \Re T_1^2\right\rangle - \langle I_y\rangle\left\langle \Im T_1^2\right\rangle - \langle I_z\rangle\left\langle \Re T_2^2\right\rangle\right) \tag{66}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\langle \Im T_2^3\right\rangle\bigg|_{\mathrm{MEC}} = -\frac{1}{\tau}\left\langle \Im T_2^3\right\rangle - \frac{2}{3\tau}\frac{1}{N}\left(\langle I_x\rangle\left\langle \Im T_1^2\right\rangle + \langle I_y\rangle\left\langle \Re T_1^2\right\rangle - \langle I_z\rangle\left\langle \Im T_2^2\right\rangle\right) \tag{67}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\langle \Re T_1^3\right\rangle\bigg|_{\mathrm{MEC}} = -\frac{1}{\tau}\left\langle \Re T_1^3\right\rangle + \frac{2}{3\sqrt{10}\tau}\frac{1}{N}\left(\langle I_x\rangle\left(\left\langle \Re T_2^2\right\rangle - 2\sqrt{3}\left\langle T_0^2\right\rangle\right) + \langle I_y\rangle\left\langle \Im T_2^2\right\rangle + 4\langle I_z\rangle\left\langle \Re T_1^2\right\rangle\right) \tag{68}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\langle \Im T_1^3\right\rangle\bigg|_{\mathrm{MEC}} = -\frac{1}{\tau}\left\langle \Im T_1^3\right\rangle + \frac{2}{3\sqrt{10}\tau}\frac{1}{N}\left(\langle I_x\rangle\left\langle \Im T_2^2\right\rangle - \langle I_y\rangle\left(\left\langle \Re T_2^2\right\rangle - 2\sqrt{3}\left\langle T_0^2\right\rangle\right) + 4\langle I_z\rangle\left\langle \Im T_1^2\right\rangle\right) \tag{69}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\langle T_0^3\right\rangle\bigg|_{\mathrm{MEC}} = -\frac{1}{\tau}\left\langle T_0^3\right\rangle + \frac{2}{\sqrt{5}\tau}\frac{1}{N}\left(\frac{2\sqrt{3}}{6}\left(\langle I_x\rangle\left\langle \Re T_1^2\right\rangle + \langle I_y\rangle\left\langle \Im T_1^2\right\rangle\right) + \langle I_z\rangle\left\langle T_0^2\right\rangle\right) \tag{70}$$

5. Semiclassical equations of motion

5.1. Semiclassical equations for the atomic and field variables

Using the results of sections 2.2, 3 and 4.2, we can write the semiclassical equations of motion for the averages of the Stokes and atomic collective spin components operators, under the influence of (i) the light–atom interaction in the metastable state, (ii) a uniform external magnetic field, and (iii) metastability exchange collisions. The term 'semiclassical' means here that all the operators, including those in equations of sections 2.2 and 3, are replaced by their expectation values. The time derivative of a semiclassical variable $\langle O\rangle$ has three contributions:

$$\frac{\mathrm{d}\langle O\rangle}{\mathrm{d}t} = \left.\frac{\mathrm{d}\langle O\rangle}{\mathrm{d}t}\right|_{\mathrm{L}} + \left.\frac{\mathrm{d}\langle O\rangle}{\mathrm{d}t}\right|_{B} + \left.\frac{\mathrm{d}\langle O\rangle}{\mathrm{d}t}\right|_{\mathrm{MEC}}. \tag{71}$$

5.2. Stationary solution

For a nuclear polarization $M \in [-1,1]$, a fixed magnetic field along the x-direction, $\vec{B} = B_x\vec{e}_x$, and a fixed light intensity and polarization, the semi-classical equations of motion have a stationary solution that is found by setting the time derivatives to zero. Here we consider the Stokes spin and the nuclear spin polarized along the static magnetic field in the x-direction,

$$\langle S_x\rangle_s = \frac{n_{\mathrm{ph}}}{2}, \qquad \langle S_y\rangle_s = \langle S_z\rangle_s = 0, \qquad \langle I_x\rangle_s = M\frac{N}{2}, \qquad \langle I_y\rangle_s = \langle I_z\rangle_s = 0. \tag{72}$$

The stationary solution for the atomic variables in the metastable state and its small polarization expansion is then:

$$\langle K_x\rangle_s = \frac{M}{2}\left(\frac{1-M^2}{3+M^2}\right)n_{\mathrm{cell}} \overset{M\to 0}{\simeq} \left(\frac{M}{6} - \frac{2M^3}{9} + \mathcal{O}\left(M^5\right)\right)n_{\mathrm{cell}} \tag{73a}$$

$$\langle J_x\rangle_s = M\left(\frac{5+M^2}{3+M^2}\right)n_{\mathrm{cell}} \overset{M\to 0}{\simeq} \left(\frac{5M}{3} - \frac{2M^3}{9} + \mathcal{O}\left(M^5\right)\right)n_{\mathrm{cell}} \tag{73b}$$

$$\langle T_0^2\rangle_s = -\left(\frac{M^2}{3+M^2}\right)n_{\mathrm{cell}} \overset{M\to 0}{\simeq} \left(-\frac{M^2}{3} + \mathcal{O}\left(M^4\right)\right)n_{\mathrm{cell}} \tag{73c}$$

$$\langle \mathfrak{R}T_2^2\rangle_s = \sqrt{3}\left(\frac{M^2}{3+M^2}\right)n_{\mathrm{cell}} \overset{M\to 0}{\simeq} \left(\frac{M^2}{\sqrt{3}} + \mathcal{O}\left(M^4\right)\right)n_{\mathrm{cell}} \tag{73d}$$

$$\langle \mathfrak{R}T_1^3\rangle_s = \sqrt{\frac{3}{10}}\left(\frac{M^3}{3+M^2}\right)n_{\mathrm{cell}} \overset{M\to 0}{\simeq} \left(\frac{M^3}{\sqrt{30}} + \mathcal{O}\left(M^5\right)\right)n_{\mathrm{cell}} \tag{73e}$$

$$\langle \mathfrak{R}T_3^3\rangle_s = -\frac{1}{\sqrt{2}}\left(\frac{M^3}{3+M^2}\right)n_{\mathrm{cell}} \overset{M\to 0}{\simeq} \left(-\frac{M^3}{3\sqrt{2}} + \mathcal{O}\left(M^5\right)\right)n_{\mathrm{cell}}. \tag{73f}$$

For $M \to 0$, the quantities (73a), (73b), (73c), (73d) and (73e), (73f) are respectively of order one, two and three in M, indicating that the tensor contributions can be neglected for a small polarization.

Moreover, we note that the stationary solutions equation (73a)–(73d) are identical to those found in [9], although the light–matter interaction for the $F = 3/2$ level of the metastable state was not included in that work.

5.3. Linearized equations of motion

The linearized semiclassical equations around the stationary solution (73) are obtained by substituting $\langle O\rangle \to \langle O\rangle_s + \delta O$, where δO is the small variation of $\langle O\rangle$ from its steady state value, and keeping only linear terms in the variations. Introducing the fluctuation vector $\vec{c} = (\vec{a},\vec{b})$ with

$$\vec{a} = \left(\delta S_y, \delta S_z, \delta I_y, \delta I_z, \delta K_y, \delta K_z, \delta J_y, \delta J_z, \delta\mathfrak{R}T_1^2, \delta\mathfrak{I}T_2^2, \delta T_0^3, \delta\mathfrak{I}T_1^3, \delta\mathfrak{R}T_2^3, \delta\mathfrak{I}T_3^3\right) \tag{74}$$

$$\vec{b} = \left(\delta S_0, \delta S_x, \delta I_x, \delta K_x, \delta J_x, \delta T_0^2, \delta\mathfrak{I}T_1^2, \mathfrak{R}T_2^2, \delta\mathfrak{R}T_1^3, \delta\mathfrak{I}T_2^3, \delta\mathfrak{R}T_3^3\right), \tag{75}$$

the linearized equations of motion take the block-diagonal form

$$\dot{\vec{c}} = \left[\begin{array}{c|c} A & 0 \\ \hline 0 & B \end{array}\right]\vec{c}, \tag{76}$$

where the first block represents fluctuations in the yz-plane, namely the plane perpendicular to the spin polarization. In the expression of the matrices A and B given below, the lines isolate the sub-blocks for the light, nuclear and metastable states respectively.

$$A=\left(\begin{array}{cc|cc|cccccccccc}
0 & -\frac{6\mu M^2 n}{M^2+3} & 0 & 0 & 0 & \frac{\chi n_{\mathrm{ph}}}{2} & 0 & \frac{\eta n_{\mathrm{ph}}}{2} & 0 & 0 & 0 & 0 & 0 & 0\\
\frac{6\mu M^2 n}{M^2+3} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -\sqrt{3}\mu n_{\mathrm{ph}} & 0 & 0 & 0 & 0\\
\hline
0 & 0 & -\frac{1}{T} & B_x\gamma_{\mathrm{nuc}} & -\frac{N}{3nT} & 0 & \frac{N}{3nT} & 0 & 0 & 0 & 0 & 0 & 0 & 0\\
0 & 0 & -B_x\gamma_{\mathrm{nuc}} & -\frac{1}{T} & 0 & -\frac{N}{3nT} & 0 & \frac{N}{3nT} & 0 & 0 & 0 & 0 & 0 & 0\\
\hline
0 & \frac{1}{2}M\left(\frac{4}{M^2+3}-1\right)n\chi & -\frac{n-M^2 n}{3M^2N\tau+9N\tau} & 0 & -\frac{7}{9\tau} & \gamma_{1/2}B_x & \frac{1}{9\tau} & 0 & 0 & -\frac{M}{3\sqrt{3}\tau} & 0 & 0 & 0 & 0\\
0 & 0 & 0 & -\frac{n-M^2 n}{3M^2N\tau+9N\tau} & \gamma_{1/2}\left(-B_x\right) & -\frac{7}{9\tau} & 0 & \frac{1}{9\tau} & \frac{M}{3\sqrt{3}\tau} & 0 & 0 & 0 & 0 & 0\\
0 & \frac{\eta M\left(M^2+5\right)n}{M^2+3} & \frac{2\left(M^2+5\right)n}{3(M^2+3)N\tau} & 0 & \frac{10}{9\tau} & 0 & -\frac{4}{9\tau} & \gamma_{3/2}B_x & 2\sqrt{3}\mu n_{\mathrm{ph}} & \frac{M}{3\sqrt{3}\tau} & 0 & 0 & 0 & 0\\
-\frac{12\mu M^2 n}{M^2+3} & 0 & 0 & \frac{2\left(M^2+5\right)n}{3(M^2+3)N\tau} & 0 & \frac{10}{9\tau} & \gamma_{3/2}\left(-B_x\right) & -\frac{4}{9\tau} & -\frac{M}{3\sqrt{3}\tau} & 2\sqrt{3}\mu n_{\mathrm{ph}} & 0 & 0 & 0 & 0\\
\frac{2\sqrt{3}\mu M\left(M^2+1\right)n}{M^2+3} & 0 & 0 & -\frac{4Mn}{\sqrt{3}(M^2N\tau+3N\tau)} & 0 & -\frac{2M}{3\sqrt{3}\tau} & -\frac{2}{5}\sqrt{3}\mu n_{\mathrm{ph}} & -\frac{M}{3\sqrt{3}\tau} & -\frac{2}{3\tau} & \gamma_{3/2}B_x & 0 & -\frac{\mu n_{\mathrm{ph}}}{\sqrt{10}} & 0 & \sqrt{\frac{3}{2}}\mu n_{\mathrm{ph}}\\
0 & \frac{2\sqrt{3}\eta M^2 n}{M^2+3} & \frac{4Mn}{\sqrt{3}(M^2N\tau+3N\tau)} & 0 & \frac{2M}{3\sqrt{3}\tau} & 0 & \frac{M}{3\sqrt{3}\tau} & -\frac{2}{5}\sqrt{3}\mu n_{\mathrm{ph}} & \gamma_{3/2}\left(-B_x\right) & -\frac{2}{3\tau} & \sqrt{\frac{3}{5}}\mu n_{\mathrm{ph}} & 0 & -\mu n_{\mathrm{ph}} & 0\\
\frac{6\mu M^2 n}{\sqrt{5}(M^2+3)} & 0 & 0 & -\frac{2M^2 n}{\sqrt{5}(M^2N\tau+3N\tau)} & 0 & 0 & 0 & 0 & \frac{M}{\sqrt{15}\tau} & -\sqrt{\frac{3}{5}}\mu n_{\mathrm{ph}} & -\frac{1}{\tau} & \sqrt{6}\gamma_{3/2}B_x & 0 & 0\\
0 & \frac{\sqrt{\frac{3}{10}}\eta M^3 n}{M^2+3} & \frac{\sqrt{\frac{2}{15}}M^2 n}{M^2N\tau+3N\tau} & 0 & 0 & 0 & 0 & 0 & \frac{\mu n_{\mathrm{ph}}}{\sqrt{10}} & \frac{M}{3\sqrt{10}\tau} & -\sqrt{6}\gamma_{3/2}B_x & -\frac{1}{\tau} & -\sqrt{\frac{5}{2}}\gamma_{3/2}B_x & 0\\
-\frac{2\sqrt{3}\mu M^2 n}{M^2+3} & 0 & 0 & \frac{2M^2 n}{\sqrt{3}(M^2N\tau+3N\tau)} & 0 & 0 & 0 & 0 & -\frac{M}{3\tau} & \mu n_{\mathrm{ph}} & 0 & \sqrt{\frac{5}{2}}\gamma_{3/2}B_x & -\frac{1}{\tau} & \sqrt{\frac{3}{2}}\gamma_{3/2}B_x\\
0 & -\frac{3\eta M^3 n}{\sqrt{2}(M^2+3)} & -\frac{\sqrt{2}M^2 n}{M^2N\tau+3N\tau} & 0 & 0 & 0 & 0 & 0 & -\sqrt{\frac{3}{2}}\mu n_{\mathrm{ph}} & -\frac{M}{\sqrt{6}\tau} & 0 & 0 & -\sqrt{\frac{3}{2}}\gamma_{3/2}B_x & -\frac{1}{\tau}
\end{array}\right) \tag{77}$$

$$B=\left(\begin{array}{cc|cc|ccccccc}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0\\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0\\
\hline
0 & 0 & -\frac{1}{T} & -\frac{N}{3nT} & \frac{N}{3nT} & 0 & 0 & 0 & 0 & 0 & 0\\
0 & 0 & -\frac{3M^2n+n}{3M^2N\tau+9N\tau} & -\frac{7}{9\tau} & \frac{1}{9\tau} & \frac{M}{9\tau} & 0 & -\frac{M}{3\sqrt{3}\tau} & 0 & 0 & 0\\
\hline
0 & 0 & \frac{6M^2n+10n}{3M^2N\tau+9N\tau} & \frac{10}{9\tau} & -\frac{4}{9\tau} & -\frac{M}{9\tau} & 0 & \frac{M}{3\sqrt{3}\tau} & 0 & 0 & 0\\
0 & 0 & -\frac{4Mn}{3M^2N\tau+9N\tau} & -\frac{2M}{9\tau} & -\frac{M}{9\tau} & -\frac{2}{3\tau} & \sqrt{3}\gamma_{3/2}B_x & 0 & 0 & \sqrt{3}\mu n_{\mathrm{ph}} & 0\\
\frac{2\sqrt{3}\mu Mn}{M^2+3} & -\frac{2\sqrt{3}\mu Mn}{M^2+3} & 0 & 0 & 0 & -\sqrt{3}\gamma_{3/2}B_x & -\frac{2}{3\tau} & \gamma_{3/2}\left(-B_x\right) & -\sqrt{\frac{5}{2}}\mu n_{\mathrm{ph}} & 0 & -\sqrt{\frac{3}{2}}\mu n_{\mathrm{ph}}\\
0 & 0 & \frac{4Mn}{\sqrt{3}(M^2N\tau+3N\tau)} & \frac{2M}{3\sqrt{3}\tau} & \frac{M}{3\sqrt{3}\tau} & 0 & \gamma_{3/2}B_x & -\frac{2}{3\tau} & 0 & \mu n_{\mathrm{ph}} & 0\\
0 & 0 & \frac{\sqrt{\frac{6}{5}}M^2n}{M^2N\tau+3N\tau} & 0 & 0 & -\frac{\sqrt{\frac{2}{15}}M}{\tau} & \sqrt{\frac{5}{2}}\mu n_{\mathrm{ph}} & \frac{M}{3\sqrt{10}\tau} & -\frac{1}{\tau} & \sqrt{\frac{5}{2}}\gamma_{3/2}B_x & 0\\
-\frac{2\sqrt{3}\mu M^2n}{M^2+3} & \frac{2\sqrt{3}\mu M^2n}{M^2+3} & 0 & 0 & 0 & -\sqrt{3}\mu n_{\mathrm{ph}} & -\frac{M}{3\tau} & -\mu n_{\mathrm{ph}} & -\sqrt{\frac{5}{2}}\gamma_{3/2}B_x & -\frac{1}{\tau} & -\sqrt{\frac{3}{2}}\gamma_{3/2}B_x\\
0 & 0 & -\frac{\sqrt{2}M^2n}{M^2N\tau+3N\tau} & 0 & 0 & 0 & \sqrt{\frac{3}{2}}\mu n_{\mathrm{ph}} & -\frac{M}{\sqrt{6}\tau} & 0 & \sqrt{\frac{3}{2}}\gamma_{3/2}B_x & -\frac{1}{\tau}
\end{array}\right) \tag{78}$$

6. Effective coupling between the nuclear spin and the light in two configurations

A simplified model involving only three coupled spins (one for the light field, one for the metastable state and one for the fundamental state) can be derived when focusing on one of the two choices of the light frequency detuning shown in figure 2: 'Config.1' or 'Config.2', where the vector Hamiltonian of the metastable level $F = 1/2$ or the metastable level $F = 3/2$ dominates. For this purpose, for a given light detuning, we set the non-dominant interaction terms in H_{LA} equation (5) to zero and, among the degrees of freedom of the metastable state, we adiabatically eliminate those that evolve only under the influence of the magnetic field and the metastable exchange collisions.

6.1. Configuration 1: Exploiting the interaction with the $F = 1/2$ manifold

We start from the linearized equations (76), and neglect the coupling of the light with the $F = 3/2$ manifold by setting $\eta = \mu = 0$. Then, we adiabatically eliminate the δJ_α and $\delta Q_{\alpha x}$ degrees of freedom by solving the algebraic equations $\mathrm{d}(\delta J_\alpha)/\mathrm{d}t = 0$ and $\mathrm{d}(\delta Q_{\alpha x})/\mathrm{d}t = 0$, and inserting the solution in the equations of motion for the remaining variables. In terms of the complex variables

$$I_+ = I_y + iI_z, \qquad K_+ = K_y + iK_z, \tag{79}$$

we obtain

$$\frac{\mathrm{d}}{\mathrm{d}t}\delta S_y = \langle S_x\rangle \chi\, \delta K_z \tag{80a}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\delta S_z = 0 \tag{80b}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\delta I_+ = -\gamma_{\mathrm{f}}^{(1/2)}\left(\frac{a_1^{(1/2)}}{c^{(1/2)}} + i\frac{B_x\gamma_{\mathrm{nuc}}}{\gamma_{\mathrm{f}}^{(1/2)}}\right)\delta I_+ + \gamma_{\mathrm{m}}^{(1/2)}\frac{a_2^{(1/2)}}{c^{(1/2)}}\delta K_+ \tag{80c}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\delta K_+ = -\gamma_{\mathrm{m}}^{(1/2)}\left(\frac{b_1^{(1/2)}}{c^{(1/2)}} + i\frac{B_x\gamma_{1/2}}{\gamma_{\mathrm{f}}^{(1/2)}}\right)\delta K_+ + \gamma_{\mathrm{f}}^{(1/2)}\frac{b_2^{(1/2)}}{c^{(1/2)}}\delta I_+ + \langle K_x\rangle \chi\, \delta S_z \tag{80d}$$

where we introduced the rescaled polarization-dependent metastability exchange rates

$$\gamma_{\mathrm{f}}^{(1/2)} = \frac{1}{T}\frac{(4+M^2)(1-M^2)}{(8-M^2)(3+M^2)}, \qquad \gamma_{\mathrm{m}}^{(1/2)} = \frac{1}{\tau}\frac{(4+M^2)}{(8-M^2)}, \tag{81}$$

and the dimensionless coefficients $a_i^{(1/2)}$, $b_i^{(1/2)}$ and $c^{(1/2)}$, that can be found in appendix D. To first order in the product $B_x\gamma_{\mathrm{ms}}/\gamma_{\mathrm{m}}$, where γ_{ms} equation (33) is the gyromagnetic factor in the metastable state and γ_{m} (53) is the metastability exchange rate for a metastable atom, one has

$$\frac{a_1^{(1/2)}}{c^{(1/2)}} \overset{B_x\to 0}{\simeq} 1 - i\frac{6(M^4+37M^2+60)}{(M^2-8)^2(M^2-1)}\frac{B_x\gamma_{3/2}}{\gamma_{\mathrm{m}}} \tag{82}$$

$$\frac{a_2^{(1/2)}}{c^{(1/2)}} \overset{B_x\to 0}{\simeq} 1 - i\frac{30(M^2+4)}{(M^2-8)^2}\frac{B_x\gamma_{3/2}}{\gamma_{\mathrm{m}}} \tag{83}$$

$$\frac{b_1^{(1/2)}}{c^{(1/2)}} \overset{B_x\to 0}{\simeq} 1 - i\frac{2(M^4+17M^2-20)}{(M^2-8)^2}\frac{B_x\gamma_{3/2}}{\gamma_{\mathrm{m}}} \tag{84}$$

$$\frac{b_2^{(1/2)}}{c^{(1/2)}} \overset{B_x\to 0}{\simeq} 1 - i\frac{30(M^2+4)}{(M^2-8)^2}\frac{B_x\gamma_{3/2}}{\gamma_{\mathrm{m}}}. \tag{85}$$

We show in figure 3 the comparison between the numerical solution of equation (80) and the full systems of equations equation (71) for two values of the nuclear spin polarization, $M = 0.02$ and $M = 0.1$. While these two models are in good agreement for the atomic variables, which shows the validity of adiabatic elimination there is a discrepancy for the variables S_y and S_z representing the light field. Such a discrepancy is due to the fact that the interaction of light with the $F = 3/2$ level, although detuned, is never completely negligible. This is especially visible for the larger polarizations, where the $F = 3/2$ manifold is more populated, and for the evolution of the S_z quadrature equation (13), which depends exclusively on the tensorial interaction.

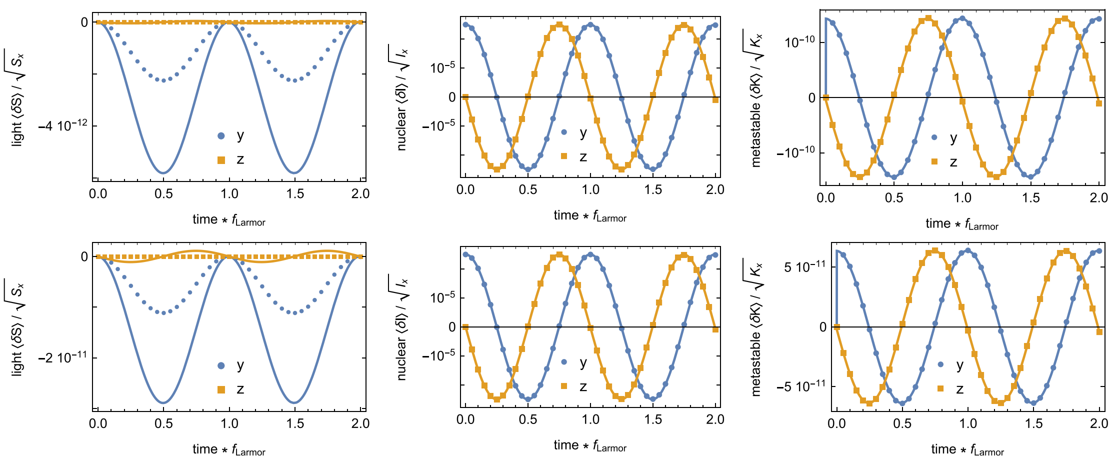

Figure 3. Comparison between the full model equation (71) and the simplified model equation (80) in 'Config.1'. Top row is for a nuclear magnetization of $M = 2\%$, while the bottom row is for $M = 10\%$. Blue indicates the y spin component, while yellow the z spin component. Solid lines are the full model, while round and square markers indicate the numerical solution of equation (80). Simulation parameters: $n_{\mathrm{ph}}/N_{\mathrm{cell}} = 10^{-3}$, $N_{\mathrm{cell}}/n_{\mathrm{cell}} = T/\tau = 10^6$, and an initial state resulting from tilting the nuclear spin by 0.01 rad. Time is in units of the nuclear spin Larmor frequency.

In the spirit of [9, 10], we are now interested in extracting the effective coupling constant between light and nuclear spin in the limit $B_x \to 0$. This can be done by adiabatically eliminating also the equations for δK, and then introducing the bosonic quadratures

$$\frac{\delta S_y}{\sqrt{\langle S_x\rangle_s}} \simeq X_S \qquad \frac{\delta I_y}{\sqrt{\langle I_x\rangle_s}} \simeq X_I \tag{86a}$$

$$\frac{\delta S_z}{\sqrt{\langle S_x\rangle_s}} \simeq P_S \qquad \frac{\delta I_z}{\sqrt{\langle I_x\rangle_s}} \simeq P_I, \tag{86b}$$

satisfying the canonical commutation relations $[X_S, P_S] = [X_I, P_I] = i\hbar$. The resulting equation of motion for the light field is

$$\frac{\mathrm{d}}{\mathrm{d}t} X_S = \chi \frac{\langle K_x\rangle_s}{\langle I_x\rangle_s} \sqrt{\langle S_x\rangle_s \langle I_x\rangle_s} P_S, \tag{87}$$

from which we obtain the effective Hamiltonian describing an interaction between light and nuclear spin

$$H_{\mathrm{eff}} = \hbar\Omega^{(1/2)} P_S P_I, \tag{88}$$

with the effective coupling rate

$$\Omega^{(1/2)} = \chi \frac{\langle K_x\rangle_s}{\langle I_x\rangle_s} \sqrt{\langle S_x\rangle_s \langle I_x\rangle_s} \tag{89}$$

$$= \chi \frac{n_{\mathrm{cell}}}{N_{\mathrm{cell}}} \sqrt{n_{\mathrm{ph}} N_{\mathrm{cell}}} f^{(1/2)}(M), \tag{90}$$

where the second line is obtained by inserting the stationary values equations (72) and (73), and in the last line we defined the polarization-dependent function

$$f^{(1/2)}(M) = \left(\frac{1 - M^2}{3 + M^2}\right) \sqrt{M}. \tag{91}$$

6.2. Configuration 2: Exploiting the interaction with the $F = 3/2$ manifold

For this configuration we want to exploit the interaction of the light with the $F = 3/2$ metastable manifold. Therefore, for a large nuclear spin polarization, we neglect the coupling of the light with the $F = 1/2$ manifold by setting $\chi = 0$ in the linearized equations (76). In addition, we see from figure 2 that in 'Config.2' the tensor polarizability is small, which motivates us to set $\mu = 0$ as well. Then we adiabatically eliminate the δK_α and $\delta Q_{\alpha x}$ degrees of freedom by solving the algebraic equations $\mathrm{d}(\delta K_\alpha)/\mathrm{d}t = 0$ and $\mathrm{d}(\delta Q_{\alpha x})/\mathrm{d}t = 0$

and inserting the solution into the equations of motion for the remaining variables. In terms of the complex variables

$$I_+ = I_y + iI_z, \qquad J_+ = J_y + iJ_z, \tag{92}$$

we obtain

$$\frac{\mathrm{d}}{\mathrm{d}t}\delta S_y = \langle S_x\rangle \eta \delta J_z \tag{93a}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\delta S_z = 0 \tag{93b}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\delta I_+ = -\gamma_{\mathrm{f}}^{(3/2)}\left(\frac{a_1^{(3/2)}}{c^{(3/2)}} + i\frac{B_x\gamma_{\mathrm{nuc}}}{\gamma_{\mathrm{f}}^{(3/2)}}\right)\delta I_+ + \gamma_{\mathrm{m}}^{(3/2)}\frac{a_2^{(3/2)}}{c^{(3/2)}}\delta J_+ + \frac{a_3^{(3/2)}}{c^{(3/2)}}\langle J_x\rangle \eta\delta S_z \tag{93c}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\delta J_+ = -\gamma_{\mathrm{m}}^{(3/2)}\left(\frac{b_1^{(3/2)}}{c^{(3/2)}} + i\frac{B_x\gamma_{3/2}}{\gamma_{\mathrm{f}}^{(3/2)}}\right)\delta J_+ + \gamma_{\mathrm{f}}^{(3/2)}\frac{b_2^{(3/2)}}{c^{(3/2)}}\delta I_+ + \left(\frac{b_3^{(3/2)}}{c^{(3/2)}} + 1\right)\langle J_x\rangle \eta\delta S_z \tag{93d}$$

where we introduced the rescaled polarization-dependent metastability exchange rates

$$\gamma_{\mathrm{f}}^{(3/2)} = \frac{1}{T}\frac{(4+M^2)(5+M^2)}{(7+M^2)(3+M^2)}, \qquad \gamma_{\mathrm{m}}^{(3/2)} = \frac{1}{\tau}\frac{(4+M^2)}{2(7+M^2)} \tag{94}$$

and the dimensionless coefficients $a_i^{(3/2)}$, $b_i^{(3/2)}$ and $c^{(3/2)}$, that can be found in appendix D. To first order in the product $B_x\gamma_{\mathrm{ms}}\tau$, where γ_{ms} equation (33) is the gyromagnetic factor in the metastable state and τ is the inverse of the metastability exchange rate, one has

$$\frac{a_1^{(3/2)}}{c^{(3/2)}} \overset{B_x\to 0}{\simeq} 1 + i\frac{3B_x\left(6\left(M^2+1\right)\gamma_{1/2} + \left(M^2+13\right)M^2\gamma_{3/2}\right)}{4\left(M^2+5\right)\left(M^2+7\right)^2\gamma_{\mathrm{m}}} \tag{95}$$

$$\frac{a_2^{(3/2)}}{c^{(3/2)}} \overset{B_x\to 0}{\simeq} 1 - i\frac{3B_x\left(2\left(M^2-2\right)\gamma_{1/2} + 3M^2\gamma_{3/2}\right)}{4\left(M^2+7\right)^2\gamma_{\mathrm{m}}} \tag{96}$$

$$\frac{a_3^{(3/2)}}{c^{(3/2)}} \overset{B_x\to 0}{\simeq} \frac{3M^2}{4\left(M^2+5\right)\left(M^2+7\right)^3}\left(4\left(M^2+7\right)^2 - 3i\left(M^2+4\right)\frac{B_x}{\gamma_{\mathrm{m}}}\left(6\gamma_{1/2} + 7\gamma_{3/2}\right)\right) \tag{97}$$

$$\frac{b_1^{(3/2)}}{c^{(3/2)}} \overset{B_x\to 0}{\simeq} 1 - i\frac{B_x\left(2\left(M^4+8M^2-20\right)\gamma_{1/2} + 9M^2\gamma_{3/2}\right)}{4\left(M^2+7\right)^2\gamma_{\mathrm{m}}} \tag{98}$$

$$\frac{b_2^{(3/2)}}{c^{(3/2)}} \overset{B_x\to 0}{\simeq} 1 + i\frac{3B_x\left(2\left(M^2+1\right)\left(M^2+10\right)\gamma_{1/2} + \left(M^2+13\right)M^2\gamma_{3/2}\right)}{4\left(M^2+5\right)\left(M^2+7\right)^2\gamma_{\mathrm{m}}} \tag{99}$$

$$\frac{b_3^{(3/2)}}{c^{(3/2)}} \overset{B_x\to 0}{\simeq} \frac{3M^2}{4\left(M^2+5\right)\left(M^2+7\right)^3}\left(i3\left(M^2+4\right)\frac{B_x}{\gamma_{\mathrm{m}}}\left(2\left(M^2+10\right)\gamma_{1/2} + 7\gamma_{3/2}\right) - 4\left(M^2+7\right)^2\right). \tag{100}$$

We show in figure 4 the comparison between the numerical solution of equation (93) and the full systems of equations equation (71) for a nuclear spin polarization of $M = 98\%$. While these two models are in good agreement for the atomic and S_y variables, there is a discrepancy for the light S_z variable. As in the previous case, such a discrepancy is due to the residual tensor interaction, as it can be noted by the oscillation of S_z at twice the Larmor frequency and it is more pronounced for large nuclear polarizations due to the M^3 scaling of the tensor components (73). On the other hand, the adiabatic elimination of the $F = 1/2$ and tensor degrees of freedom (without setting μ and χ to zero) gives a very good approximation of the dynamics, as we show numerically in section 7.

The effective coupling constant between light and nuclear spin in the $B_x = 0$ limit can be extracted similarly to the previous case, now adiabatically eliminating also the equations for δJ. The effective Hamiltonian is then

$$H_{\mathrm{eff}} = \hbar\Omega^{(3/2)} P_S P_I, \tag{101}$$

with the effective coupling rate between the light field and the nuclear spin given by

$$\Omega^{(3/2)} = \eta\frac{\langle J_x\rangle_s}{\langle I_x\rangle_s}\sqrt{\langle S_x\rangle_s\langle I_x\rangle_s} \tag{102}$$

$$= \eta\frac{n_{\mathrm{cell}}}{N_{\mathrm{cell}}}\sqrt{n_{\mathrm{ph}}N_{\mathrm{cell}}}f^{(3/2)}(M). \tag{103}$$

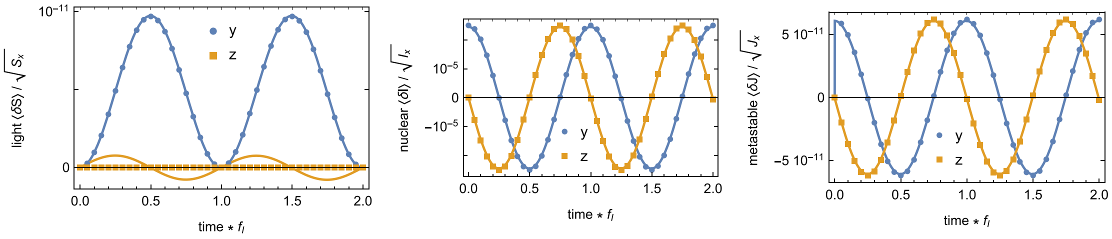

Figure 4. Comparison between the full model equation (71) and the simplified model equation (93) in 'Config.2'. The nuclear magnetization is set to $M = 98\%$. Blue indicates the y spin component, while yellow the z spin component. Solid lines are the full model, while round and square markers indicate the numerical solution of equation (93). Simulation parameters: $n_{\mathrm{ph}}/N_{\mathrm{cell}} = 10^{-3}$, $N_{\mathrm{cell}}/n_{\mathrm{cell}} = T/\tau = 10^{6}$, and an initial state resulting from tilting the nuclear spin by 0.01 rad. Time is in units of the nuclear spin Larmor frequency.

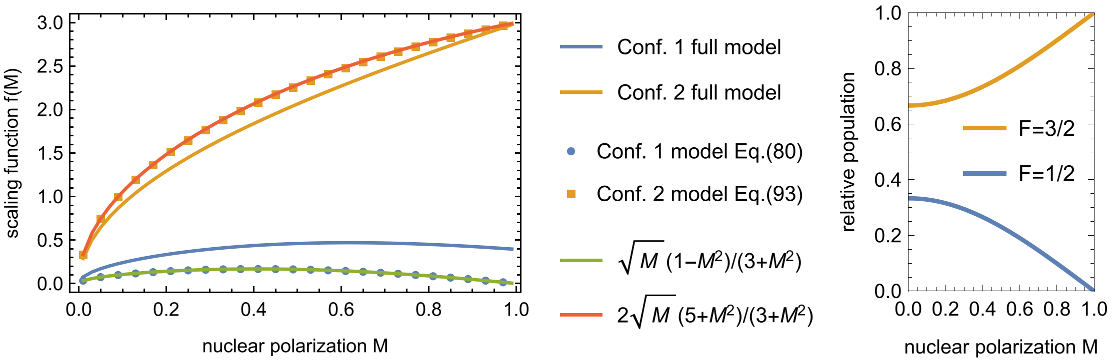

Figure 5. Comparison between the effective light-nuclear spin coupling factor in 'Config.1' and 'Config.2' as a function of the nuclear spin magnetization **M**, and associated metastable manifolds population. Left: Blue indicates 'Config.1', while yellow 'Config.2'. Solid lines refer to the coupling extracted from the full model, while round and square markers refer to the coupling extracted from equations (80) and (93), respectively. Red and green lines are the analytical expressions obtained for $B_x = 0$, namely equations (91) and (104) respectively. Simulation parameters: $n_{\mathrm{ph}}/N_{\mathrm{cell}} = 10^{-3}$ and $N_{\mathrm{cell}}/n_{\mathrm{cell}} = T/\tau = 10^{6}$. Right: relative population in the metastable manifolds $F = 1/2$ and $F = 3/2$ as a function of the nuclear polarization along the x-direction.

Here we used $T/\tau = N_{\mathrm{cell}}/n_{\mathrm{cell}}$, together with the stationary solution for the spins equations (72) and (73), and we defined the polarization-dependent scaling function

$$f^{(3/2)}(M) = 2\left(\frac{5+M^2}{3+M^2}\right)\sqrt{M}. \tag{104}$$

In the next section we will compare this result with the one obtained for 'Config.1'.

6.3. Effective coupling in the two configurations and comparison with the full model

Equations (89) and (102) for the rates $\Omega^{(1/2)}$ and $\Omega^{(3/2)}$ describing the effective coupling between the collective nuclear spin and the light in 'Config.1' and 'Config.2' respectively, were obtained from approximate models. In this section, we extract such coupling constants from numerical simulations of the full semiclassical equations, and compare the results with the analytical expressions.

From the evolution of the Stokes spin fluctuation X_S, equations (86) and (87), we see that an oscillation of the collective nuclear spin fluctuation $P_I = P_I(0)\cos(\omega_I t + \phi)$ results in a light signal $X_S = (\Omega P_I(0)/\omega_I)\sin(\omega_I t + \phi)$. Computing the ratio between the oscillation amplitude of the light and nuclear spin gives us Ω/ω_I, from which we extract the effective coupling for different nuclear polarizations.

We plot in figure 5 the polarization dependent part of the coupling as obtained from the solution of the full set equations of motions (71), for a small initial tilt of the collective nuclear spin in the linear response regime in the two configurations (solid lines), and from the solution of the simplified models (80) in 'Config.1' (circles) and (93) in 'Config.2' (squares). On the same plot, we show the analytic expressions of the functions (91) and (104).

Even accounting for the difference in the coupling constants in the two configurations, η being approximately 0.48 times χ, due to the large difference in the scaling factors $f^{(3/2)}$ and $f^{(1/2)}$ the effective coupling between nuclear spin and light is significantly larger in 'Config.2' than in 'Config.1'.

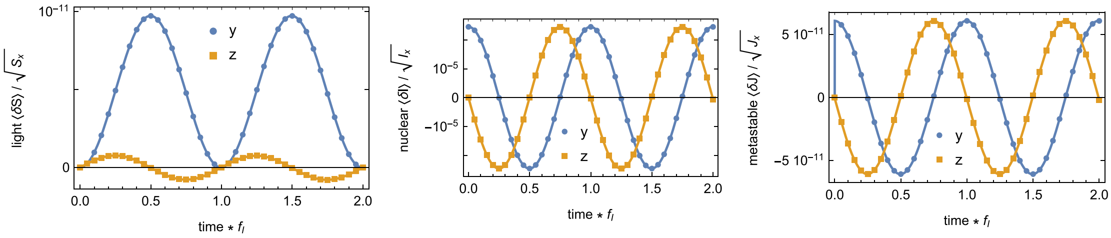

Figure 6. Comparison between the full model equation (71) and the same model after adiabatic elimination of the $F = 1/2$ and tensor degrees of freedom in 'Config.2'. The nuclear magnetization is set to $M = 98\%$. Blue indicates the y spin component, while yellow the z spin component. Solid lines are the full model, while round and square markers indicate the numerical solution of the same model after adiabatic elimination of the $F = 1/2$ and tensor degrees of freedom (without setting $\mu = \chi = 0$, as done in figure 4). Simulation parameters: $n_{\mathrm{ph}}/N_{\mathrm{cell}} = 10^{-3}$, $N_{\mathrm{cell}}/n_{\mathrm{cell}} = T/\tau = 10^6$, and an initial state resulting from tilting the nuclear spin by 0.01 rad. Time is in units of the nuclear spin Larmor frequency.

7. Discussion on the validity of the analytical models

In sections 6.1 and 6.2 we obtain for 'Config.1' and 'Config.2' a set of simple equations of motion for three collective variables (light, nuclear and metastable spins) that can be treated analytically. For example, for 'Config.2' we obtain a closed set of equations for the variables S, I and J, by first setting the coupling coefficients $\chi = \mu = 0$ in equation (76), and then adiabatically eliminating the δK_α and $\delta Q_{\alpha x}$ degrees of freedom.

When we compare the predictions of the full numerical model including all the atomic transitions and those of the analytical models in 'Config.1' or 'Config.2', we find good agreement for the nuclear and metastable variables, see figures 3 and 4, but less good agreement for the light variables. This is because in the analytical models we have neglected from the start part of the light–atoms interaction, and in particular the tensor part from the $F = 3/2$ manifold that, although subdominant, is always present. This is also the reason for the difference between the curves from the full numerical model and the analytical models in figure 5(left). In particular, the simplified model for 'Config.1' fails for large polarizations, because the $F = 1/2$ manifold is empty when $M = 1$, see figure 5(right).

A more accurate (but non analytic) effective model involving three collective spins can be obtained by still performing the adiabatic elimination, but keeping all the terms in the light–atom Hamiltonian. For example, for 'Config.2', we perform the adiabatic elimination of δK_α and $\delta Q_{\alpha x}$ directly on equation (76), keeping all the coupling coefficients. This leads to a set of expressions that are too complex to be treated analytically, but can be solved numerically showing good agreement with the full model, see figure 6.

8. Conclusions

In conclusion, we have derived the full set of semiclassical equation of motion describing the interaction between light and metastable helium-3, taking into account metastability-exchange collisions with helium-3 atoms in the ground state as well as a static external weak magnetic field. We then explored two particular choices of detunings between light and metastable helium-3 $2^3S - 2^3P$ transition, for which the dominant interaction, in the $F = 1/2$ or $F = 3/2$ manifolds, is vectorial of the Faraday form. For these configurations we were able to extract analytically the effective coupling constant between light and helium-3 nuclear spin as a function of the experimental parameters, and conclude that this quantity is considerably larger in the configuration dominated by the $F = 3/2$ manifold with a large nuclear spin polarization. A comparison between the numerical solution of the full set of equations of motion and the analytical model shows a fairly good agreement. The observed discrepancies come from the coupling of the light with tensor spin components that was neglected in the analytical models. In the future, it will be important to explore how the presence of these tensor contributions might affect squeezing of the nuclear spin in a fully quantum treatment [9, 10].

Data availability statement

No new data were created or analyzed in this study.

Acknowledgments

MF was supported by the Swiss National Science Foundation Ambizione Grant No. 208886, and The Branco Weiss Fellowship—Society in Science, administered by the ETH Zürich.

Appendix A. Effective Hamiltonian in a state F in the large detuning limit

The Hamiltonian for a single-atom interacting with a light field is

$$h_F = \sum_{F'} \hbar \frac{\sigma_{F'}}{2A} \frac{\Gamma/2}{\Delta_{F'} + i\Gamma/2} \left\{ \alpha^V_{F'} F_z S_z + \frac{\alpha^T_{F'}}{(F+1)} \left[\left(\frac{F(F+1)}{3} - F_z^2 \right) S_0 + \left(F_x^2 - F_y^2 \right) S_x + \left(F_x F_y + F_y F_x \right) S_y \right] \right\}. \tag{A1}$$

In equation (A1), F (F') is the total angular momentum of the starting (target) state of the transition, $\sigma_{F'}$ is the resonant effective cross section of the transition $F \to F'$, and $\Delta_{F'} = \omega_{\text{probe}} - \omega_{FF'}$ is the detuning with respect to the resonance. In the expression $\Delta_{F'} + i\Gamma/2$ in the denominator, the imaginary part can be neglected for $\Delta_{F'} \gg \Gamma/2$.

The vector and tensor components of the polarization have the form [15, 22]

$$\alpha^V_{F'} = \frac{3(2J'+1)}{2(2F'+1)(2J+1)} \left(-\frac{2F-1}{F} \delta^{F'}_{F-1} - \frac{2F+1}{F(F+1)} \delta^{F'}_{F} + \frac{2F+3}{F+1} \delta^{F'}_{F+1} \right) \tag{A2}$$

$$\alpha^T_{F'} = -\frac{3(F+1)(2J'+1)}{2(2F'+1)(2J+1)} \left(\frac{1}{F} \delta^{F'}_{F-1} - \frac{2F+1}{F(F+1)} \delta^{F'}_{F} + \frac{1}{F+1} \delta^{F'}_{F+1} \right). \tag{A3}$$

Introducing $\sigma_2 = 3\lambda^2/2\pi$ and Wigner's $6j$ symbols $\{\}$, the resonant effective cross section $\sigma_{F'}$ between two levels F, J, I and F', J', I is given by

$$\sigma_{F'} = \sigma_2 \frac{2(2J+1)(2F'+1)}{3} \begin{Bmatrix} J' & 1 & J \\ F & I & F' \end{Bmatrix}^2. \tag{A4}$$

For a spin greater than $F = 1/2$, the irreducible tensor basis t^l_m should be used, with $l = 0, 1, .., 2F$ and $m = -l, \ldots, l$ defined as a function of the ladder operators $F_\pm = F_x \pm i F_y$, and given below for $l \leqslant 3$.

$$t^0_0 = n^0_0\, \mathbb{I} \tag{A5a}$$

$$t^1_0 = n^1_0\, F_z \tag{A5b}$$

$$t^1_{\pm 1} = n^1_{\pm 1}\, F_\pm \tag{A5c}$$

$$t^2_0 = n^2_0\, \left(3F_z^2 - \mathbf{F}^2\right) \tag{A5d}$$

$$t^2_{\pm 1} = n^2_{\pm 1}\, \left(F_\pm F_z + F_z F_\pm\right) \tag{A5e}$$

$$t^2_{\pm 2} = n^2_{\pm 2}\, F_\pm^2 \tag{A5f}$$

$$t^3_0 = n^3_0\, \left(5F_z^2 - 3\mathbf{F}^2 + 1\right) F_z \tag{A5g}$$

$$t^3_{\pm 1} = n^3_{\pm 1}\, \left[\left(5F_z^2 - \mathbf{F}^2 - 1/2\right) F_\pm + F_\pm \left(5F_z^2 - \mathbf{F}^2 - 1/2\right)\right] \tag{A5h}$$

$$t^3_{\pm 2} = n^3_{\pm 2}\, \left(F_\pm^2 F_z + F_\pm F_z F_\pm + F_z F_\pm^2\right) \tag{A5i}$$

$$t^3_{\pm 3} = n^3_{\pm 3}\, F_\pm^3. \tag{A5j}$$

As expected, for a spin F operators of rank $l > 2F$ are null. The operators t^l_m satisfy property $(t^l_m)^\dagger = (-1)^m t^l_{-m}$ and are of null trace except $t^{l=0}_0$. Other properties and commutation relations are given in appendix B. Prefactors n^l_m are chosen to ensure the normalization condition $\mathrm{Tr}[t^l_m (t^l_m)^\dagger] = 1$, and for a spin $F = 3/2$ they read

n^0_0	n^1_0	$n^1_{\pm 1}$	n^2_0	$n^2_{\pm 1}$	$n^2_{\pm 2}$	n^3_0	$n^3_{\pm 1}$	$n^3_{\pm 2}$	$n^3_{\pm 3}$
$\frac{1}{2}$	$\frac{1}{\sqrt{5}}$	$\mp \frac{1}{\sqrt{10}}$	$\frac{1}{6}$	$\mp \frac{1}{2\sqrt{6}}$	$\frac{1}{2\sqrt{6}}$	$\frac{1}{3\sqrt{5}}$	$\mp \frac{1}{4\sqrt{15}}$	$\frac{1}{3\sqrt{6}}$	$\mp \frac{1}{6}$.

Finally, we introduce the symmetric and antisymmetric combinations

$$\mathfrak{R} t^l_m \equiv \frac{t^l_m + \left(t^l_m\right)^\dagger}{\sqrt{2}} \qquad \mathfrak{I} t^l_m \equiv \frac{t^l_m - \left(t^l_m\right)^\dagger}{i\sqrt{2}}. \tag{A6}$$

The associated collective operators are then defined by summing over all particles as *e.g.* $\Re T^l_m = \sum_{i=1}^N (\Re t^l_m)_i$. We note that T^1_0 is proportional to the longitudinal magnetization, $\Re T^1_1$ and $\Im T^1_1$ to the magnetizations according to x and y, T^2_0 is the quadrupolar spin polarization (called alignment), $\Re T^2_1$ and $\Im T^2_1$ imply coherences between levels $\Delta m = 1$, $\Re T^2_2$ and $\Im T^2_2$ between levels $\Delta m = 2$, T^3_0 is the octopolar spin polarization, etc.

Assuming $\Delta \gg \Gamma$, and summing over all atoms, we can rewrite the collective Hamiltonian

$$H_F = \sum_{F'} \hbar \frac{\sigma_{F'}}{4A} \frac{\Gamma}{\Delta_{F'}} \left\{ \alpha^V_{F'} F_z S_z + \frac{\alpha^T_{F'}}{(F+1)} \left[-2T^2_0 S_0 + \sqrt{12} \left(\Re T^2_2 S_x + \Im T^2_2 S_y \right) \right] \right\}, \tag{A7}$$

where we have preferred the notation F_z rather than T^1_0.

Appendix B. Commutation relations for the atomic operators

Let $\vec{F} = \{F_x, F_y, F_z\}$ be the components of a spin operator, $F_\pm = F_x \pm i F_y$ the corresponding ladder operators, and t^l_m the irreducible tensors explicit in (A5) for $l \leqslant 3$. The operators satisfy the following commutator rules [15, 23]:

$$[F_x, F_y] = iF_z \qquad \text{(and cyclic permutations)} \tag{B1}$$

$$[F_z, F_\pm] = \pm F_\pm \tag{B2}$$

$$[F_+, F_-] = 2F_z \tag{B3}$$

$$[F_z, t^l_m] = m t^l_m \tag{B4}$$

$$[F_\pm, t^l_m] = \sqrt{(l \pm m + 1)(l \mp m)}\, t^l_{m\pm1} \tag{B5}$$

$$[t^{l_1}_{m_1}, t^{l_2}_{m_2}] = \sum_{L,M} (-1)^{L+2F} \sqrt{(2l_1+1)(2l_2+1)} \begin{Bmatrix} l_1 & l_2 & L \\ F & F & F \end{Bmatrix} \langle l_1 m_1 l_2 m_2, LM \rangle \left[1 - (-1)^{l_1+l_2+L} \right] t^L_M, \tag{B6}$$

where $\{\}$ denotes Wigner's 6j symbols, and $\langle , \rangle$ the Clebsch–Gordan coefficients.

Appendix C. Derivation of metastability exchange equations

In this appendix we explain how the metastability exchange equations presented in section 4.2 can be derived in practice. First, the density matrix ρ can be written as $\rho = \sum_{i,j} \rho_{i,j} |i\rangle \langle j|$, where the indices i, j label the basis states

$$|5\rangle = \sqrt{\frac{1}{3}} \left|0, -\frac{1}{2}\right\rangle - \sqrt{\frac{2}{3}} \left|-1, \frac{1}{2}\right\rangle \quad |6\rangle = -\sqrt{\frac{1}{3}} \left|0, \frac{1}{2}\right\rangle + \sqrt{\frac{2}{3}} \left|1, -\frac{1}{2}\right\rangle \tag{C1a}$$

$$|1\rangle = \left|-1, -\frac{1}{2}\right\rangle \quad |2\rangle = \sqrt{\frac{2}{3}} \left|0, -\frac{1}{2}\right\rangle + \sqrt{\frac{1}{3}} \left|-1, \frac{1}{2}\right\rangle \quad |3\rangle = \sqrt{\frac{2}{3}} \left|0, \frac{1}{2}\right\rangle + \sqrt{\frac{1}{3}} \left|1, -\frac{1}{2}\right\rangle \quad |4\rangle = \left|1, \frac{1}{2}\right\rangle \tag{C1b}$$

$$|9\rangle = \left|-\frac{1}{2}\right\rangle \quad |0\rangle = \left|\frac{1}{2}\right\rangle. \tag{C1c}$$

Here, note that $|9\rangle$ and $|0\rangle$ are purely nuclear states, while the others are hyperfine states of total spin $F = 3/2$ and $F = 1/2$ expressed in the decoupled basis of the electronic and nuclear spin. In practice, we neglect coherences between metastable and ground states, as well as coherences between the $3/2$ and $1/2$ states. This gives us

$$\rho = \left[\begin{array}{c|c} \rho_\mathrm{m} & 0 \\ \hline 0 & \rho_\mathrm{f} \end{array} \right] = \left[\begin{array}{cccccc|cc} \rho_{1,1} & \rho_{1,2} & \rho_{1,3} & \rho_{1,4} & 0 & 0 & 0 & 0 \\ \rho_{2,1} & \rho_{2,2} & \rho_{2,3} & \rho_{2,4} & 0 & 0 & 0 & 0 \\ \rho_{3,1} & \rho_{3,2} & \rho_{3,3} & \rho_{3,4} & 0 & 0 & 0 & 0 \\ \rho_{4,1} & \rho_{4,2} & \rho_{4,3} & \rho_{4,4} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & \rho_{5,5} & \rho_{5,6} & 0 & 0 \\ 0 & 0 & 0 & 0 & \rho_{6,5} & \rho_{6,6} & 0 & 0 \\ \hline 0 & 0 & 0 & 0 & 0 & 0 & \rho_{9,9} & \rho_{9,0} \\ 0 & 0 & 0 & 0 & 0 & 0 & \rho_{0,9} & \rho_{0,0} \end{array} \right]. \tag{C2}$$

Using now equations (51) and (52), with equations (49) and (50), allows us to derive the equations of motion of ρ due to metastability exchange collisions. These can be found in the appendix of [6]. Finally, the evolution due to metastability exchange collisions of any one-body atomic operator O can be calculated using equation (54). To this end, it is convenient to express the spin operators in the basis equation (C1). For the $F = 1/2$ metastable state and the nuclear ground state, the spin operators in the relevant 2×2 subspace are proportional to the Pauli matrices, *e.g.* $k_z = \frac{1}{2}\begin{pmatrix}-1 & 0\\ 0 & 1\end{pmatrix}$. For the $F = 3/2$ metastable state the spin operators in the relevant 4×4 subspace are

$$j_x = \begin{pmatrix} 0 & \frac{\sqrt{3}}{2} & 0 & 0 \\ \frac{\sqrt{3}}{2} & 0 & 1 & 0 \\ 0 & 1 & 0 & \frac{\sqrt{3}}{2} \\ 0 & 0 & \frac{\sqrt{3}}{2} & 0 \end{pmatrix}, \quad j_y = \begin{pmatrix} 0 & \frac{i\sqrt{3}}{2} & 0 & 0 \\ -\frac{i\sqrt{3}}{2} & 0 & i & 0 \\ 0 & -i & 0 & \frac{i\sqrt{3}}{2} \\ 0 & 0 & -\frac{i\sqrt{3}}{2} & 0 \end{pmatrix}, \quad j_z = \begin{pmatrix} -\frac{3}{2} & 0 & 0 & 0 \\ 0 & -\frac{1}{2} & 0 & 0 \\ 0 & 0 & \frac{1}{2} & 0 \\ 0 & 0 & 0 & \frac{3}{2} \end{pmatrix}, \tag{C3}$$

$$t_0^2 = \begin{pmatrix} \frac{1}{2} & 0 & 0 & 0 \\ 0 & -\frac{1}{2} & 0 & 0 \\ 0 & 0 & -\frac{1}{2} & 0 \\ 0 & 0 & 0 & \frac{1}{2} \end{pmatrix}, \quad \Re t_1^2 = \begin{pmatrix} 0 & \frac{1}{2} & 0 & 0 \\ \frac{1}{2} & 0 & 0 & 0 \\ 0 & 0 & 0 & -\frac{1}{2} \\ 0 & 0 & -\frac{1}{2} & 0 \end{pmatrix}, \quad \Re t_2^2 = \begin{pmatrix} 0 & 0 & \frac{1}{2} & 0 \\ 0 & 0 & 0 & \frac{1}{2} \\ \frac{1}{2} & 0 & 0 & 0 \\ 0 & \frac{1}{2} & 0 & 0 \end{pmatrix}, \tag{C4}$$

$$\Im t_1^2 = \begin{pmatrix} 0 & \frac{i}{2} & 0 & 0 \\ -\frac{i}{2} & 0 & 0 & 0 \\ 0 & 0 & 0 & -\frac{i}{2} \\ 0 & 0 & \frac{i}{2} & 0 \end{pmatrix}, \quad \Im t_2^2 = \begin{pmatrix} 0 & 0 & \frac{i}{2} & 0 \\ 0 & 0 & 0 & \frac{i}{2} \\ -\frac{i}{2} & 0 & 0 & 0 \\ 0 & -\frac{i}{2} & 0 & 0 \end{pmatrix}, \quad t_0^3 = \begin{pmatrix} -\frac{1}{2\sqrt{5}} & 0 & 0 & 0 \\ 0 & \frac{3}{2\sqrt{5}} & 0 & 0 \\ 0 & 0 & -\frac{3}{2\sqrt{5}} & 0 \\ 0 & 0 & 0 & \frac{1}{2\sqrt{5}} \end{pmatrix}, \tag{C5}$$

$$\Re t_1^3 = \begin{pmatrix} 0 & -\frac{1}{\sqrt{10}} & 0 & 0 \\ -\frac{1}{\sqrt{10}} & 0 & \sqrt{\frac{3}{10}} & 0 \\ 0 & \sqrt{\frac{3}{10}} & 0 & -\frac{1}{\sqrt{10}} \\ 0 & 0 & -\frac{1}{\sqrt{10}} & 0 \end{pmatrix}, \quad \Re t_2^3 = \begin{pmatrix} 0 & 0 & -\frac{1}{2} & 0 \\ 0 & 0 & 0 & \frac{1}{2} \\ -\frac{1}{2} & 0 & 0 & 0 \\ 0 & \frac{1}{2} & 0 & 0 \end{pmatrix}, \quad \Re t_3^3 = \begin{pmatrix} 0 & 0 & 0 & -\frac{1}{\sqrt{2}} \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ -\frac{1}{\sqrt{2}} & 0 & 0 & 0 \end{pmatrix}, \tag{C6}$$

$$\Im t_1^3 = \begin{pmatrix} 0 & -\frac{i}{\sqrt{10}} & 0 & 0 \\ \frac{i}{\sqrt{10}} & 0 & i\sqrt{\frac{3}{10}} & 0 \\ 0 & -i\sqrt{\frac{3}{10}} & 0 & -\frac{i}{\sqrt{10}} \\ 0 & 0 & \frac{i}{\sqrt{10}} & 0 \end{pmatrix}, \quad \Im t_2^3 = \begin{pmatrix} 0 & 0 & -\frac{i}{2} & 0 \\ 0 & 0 & 0 & \frac{i}{2} \\ \frac{i}{2} & 0 & 0 & 0 \\ 0 & -\frac{i}{2} & 0 & 0 \end{pmatrix}, \quad \Im t_3^3 = \begin{pmatrix} 0 & 0 & 0 & -\frac{i}{\sqrt{2}} \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ \frac{i}{\sqrt{2}} & 0 & 0 & 0 \end{pmatrix}. \tag{C7}$$

The equations of motion for the collective spin operators are then readily found by taking

$$\left\langle \vec{I} \right\rangle = N \left\langle \vec{i} \right\rangle, \qquad \left\langle \vec{K} \right\rangle = n \left\langle \vec{k} \right\rangle, \qquad \left\langle \vec{J} \right\rangle = n \left\langle \vec{j} \right\rangle, \qquad \langle \Re T_2^2 \rangle = n \langle \Re t_2^2 \rangle, \qquad \text{etc} \tag{C8}$$

and are the one presented in section 4.2.

Appendix D. Coefficients of the linearized equations of the simplified models

In this appendix we give the expression of the coefficients appearing in the linearized equations of the simplified models of section 6.

The coefficients appearing in equation (80) read

$$c^{(1/2)} = \left(\frac{30i\left(M^2+4\right)\gamma_{3/2}B_x}{\gamma_{\mathrm{m}}} + \frac{27\left(M^2+4\right)^2 \gamma^2_{3/2(M^2-8)} B_x^2}{\gamma_{\mathrm{m}}^2} + \left(M^2-8\right)^2 \right) \tag{D1}$$

$$a_1^{(1/2)} = \frac{\left(M^2-8\right)}{\left(M^2+4\right)\left(M^2-1\right)} \left(\left(M^2+3\right)c^{(1/2)} + 4\left(\left(M^2-8\right)\left(2M^2+5\right) - \frac{3i\left(M^2+4\right)\left(M^2+5\right)\gamma_{3/2}B_x}{2\gamma_{\mathrm{m}}} \right) \right) \tag{D2}$$

$$a_2^{(1/2)} = \left(\frac{9\left(M^2+4\right)\gamma_{3/2}^2 B_x^2}{\gamma_{\mathrm{m}}^2} + \left(M^2-8\right)^2 \right) \tag{D3}$$

$$b_1^{(1/2)} = \left(-\frac{21\left(M^2+4\right)\gamma_{3/2}^2 B_x^2}{\gamma_{\mathrm{m}}^2} - \frac{2i\left(M^4+2M^2-80\right)\gamma_{3/2}B_x}{\gamma_{\mathrm{m}}} + \left(M^2-8\right)^2 \right) \tag{D4}$$

$$b_2^{(1/2)} = \left(\frac{9\left(M^2+4\right)\gamma_{3/2}^2 B_x^2}{\gamma_{\mathrm{m}}^2} + \left(M^2-8\right)^2 \right). \tag{D5}$$

The coefficients appearing in equation (93) read

$$c^{(3/2)} = \frac{6i\left(M^2+4\right)B_x\left(6\gamma_{1/2}+7\gamma_{3/2}\right)}{\gamma_{\mathrm{m}}} - \frac{27\left(M^2+4\right)^2\gamma_{1/2}\gamma_{3/2}B_x^2}{\left(M^2+7\right)\gamma_{\mathrm{m}}^2} + 8\left(M^2+7\right)^2 \tag{D6}$$

$$a_1^{(3/2)} = \frac{\left(M^2+7\right)}{\left(M^2+4\right)\left(M^2+5\right)}\left(\left(M^2+3\right)c^{(3/2)} - 8\left(\left(M^2+1\right)\left(M^2+7\right) - \frac{3i\left(M^2-1\right)\left(M^2+4\right)\gamma_{3/2}B_x}{4\gamma_{\mathrm{m}}}\right)\right) \tag{D7}$$

$$a_2^{(3/2)} = 2\left(\frac{12i\left(M^2+7\right)B_x\left(\gamma_{1/2}+\gamma_{3/2}\right)}{\gamma_{\mathrm{m}}} - \frac{9\left(M^2+4\right)\gamma_{1/2}\gamma_{3/2}B_x^2}{\gamma_{\mathrm{m}}^2} + 4\left(M^2+7\right)^2\right) \tag{D8}$$

$$a_3^{(3/2)} = \frac{24M^2\left(M^2+7\right)}{\left(M^2+5\right)} \tag{D9}$$

$$b_1^{(3/2)} = -4\left(\frac{i\left(M^2+7\right)B_x\left(\left(M^2-8\right)\gamma_{1/2}-6\gamma_{3/2}\right)}{\gamma_{\mathrm{m}}} + \frac{6\left(M^2+4\right)\gamma_{1/2}\gamma_{3/2}B_x^2}{\gamma_{\mathrm{m}}^2} - 2\left(M^2+7\right)^2\right) \tag{D10}$$

$$b_2^{(3/2)} = a_2^{(3/2)} + a_3^{(3/2)}\frac{iB_x\left(\gamma_{1/2}+\gamma_{3/2}\right)}{\gamma_{\mathrm{m}}} \tag{D11}$$

$$b_3^{(3/2)} = \frac{12M^2}{\left(M^2+5\right)}\left(-2\left(M^2+7\right) + \frac{3i\left(M^2+4\right)\gamma_{1/2}B_x}{\gamma_{\mathrm{m}}}\right). \tag{D12}$$

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