Spin-dressed relaxation and frequency shifts from field imperfections

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Critical dressing, the simultaneous dressing of two spin species to the same effective Larmor frequency, is a technique that can, in principle, improve the sensitivity to small frequency shifts. The benefits of spin dressing and thus critical dressing are achieved at the expense of generating a large (relative to the holding field B_0,) homogeneous oscillating field. Due to inevitable imperfections of the fields generated, the benefits of spin dressing may be lost from the additional relaxation and noise generated by the dressing field imperfections. In this analysis, the subject of relaxation and frequency shifts are approached with simulations and theory. Analytical predictions are made from a new quasiquantum model that includes gradients in the holding field $B_0 = \omega_0/\gamma$ and dressing field $B_1 = \omega_1/\gamma$ where B_1 is oscillating at frequency ω. It is found that irreversible DC gradient relaxation can be canceled by an AC spin-dressing gradient in the Redfield regime. Furthermore, it is shown that there is no linear in E frequency shift generated by gradients in the dressing field. The results are compared with a Monte Carlo simulation coupled with a fifth-order Runge-Kutta integrator. Comparisons of the two methods are presented as well as a set of optimized parameters that produce stable critical dressing for a range of oscillating frequencies ω, as well as pulsed frequency modulation parameters for maximum sensitivity.

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I. INTRODUCTION

Spin dressing is a technique that changes the effective Zeeman splitting of an atomic or nuclear spin system [1,2] and is found to have a wide range of applications. Notably, the manipulation of spin dynamics of ultracold quantum gases [3], and orbit interactions and its influence on dynamics, and enabling a new avenue for coupling optics to nanomaterials [4]. It is also a valuable tool in the field of fundamental physics.

A variation of spin dressing has been proposed as a technique to maximize sensitivity in the search for the time reversal and parity violating observable, the permanent electric dipole moment (EDM). In Ref. [5] they propose critical dressing of spin-polarized neutrons and ^{3}He. Critical dressing is the simultaneous spin dressing of two species so that they have the same effective gyromagnetic ratio.

Here we present a detailed framework of the technique of critical dressing with the aim of optimization of experimental sensitivity and mitigation of systematic effects. This is achieved by including the effects of the fluctuations in field as observed by the particle's trajectory through the field inhomogeneities, similar to the derivation in Ref. [6]. In Ref. [7] they approach the case of relaxation due to collisions of gas atoms; however, in the system presented here, scattering in the bulk does not directly affect the dynamics, it only modifies the field fluctuations observed by the particle by modification of the particle's trajectory. In Ref. [8] they reformulate the result in Ref. [2] and find solutions for nonharmonic dressing fields as well offset dressing fields. In Ref. [9] they consider the case of a chromatic dressing field, and in Ref. [10] they find the propagator for the polychromatic dressed Hamiltonian. Here we consider fluctuations in the field producing the spin dressing of a gas sample over a macroscopically large volume, where the particle's trajectory through the inhomogeneities of the DC holding field, as well as the spin-dressing field, are the largest sources of relaxation and frequency shifts. We use an approach similar to that found in Refs. [6], [11], and [12]. We show that the spectrum of the correlation functions that determine the relaxation and frequency shifts depend on the dressed energy, it is not enough to correct the result in the case of no oscillating field by a factor $J_0(\frac{\omega_1}{\omega})$, as would be found if one included the shift as an intrinsic energy splitting in the Hamiltonian.

The benefits of spin dressing and thus critical dressing are achieved at the expense of generating a large (relative to the holding field B_0,) homogeneous oscillating field. Due to inevitable imperfections of laboratory fields, the benefits of the spin-dressing technique may be lost from the additional relaxation and noise generated by an imperfect field. In this analysis the subject of relaxation and frequency shifts are approached with a quasiquantum model that includes gradients in the holding field $B_0 = \omega_0/\gamma$ and dressing field $B_1 = \omega_1/\gamma$ oscillating at frequency ω. We will present an analytic model that can determine the relaxation and frequency shifts given an inhomogeneity of the holding field B_0 and a uniform applied electric field. The model is considered quasiquantum due to the use of classical trajectories, presented in Ref. [13], to determine the field fluctuations in the rest frame of the particle. Furthermore, we compare the results to a Monte Carlo simulation coupled with a fifth-order Runge-Kutta integrator. This simulation is a modified version of the simulation outlined in Ref. [14] and used to produce the results in Ref. [15]. This code was modified to produce a set of parameters that produce critical dressing at a range of oscillating frequencies ω, as

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well as parameters for pulsed frequency modulation of the spin-dressing field.

II. SPIN-DRESSING MODEL

Starting from the Hamiltonian, according to Refs. [1,2], for a spin in a strong AC magnetic field along x (B_1) and weaker DC holding field (B_0) along z,

$$\frac{H}{\hbar} = \omega a^\dagger a + \frac{\gamma B_1}{2\lambda^{1/2}}\frac{\sigma_x}{2}(a + a^\dagger) + \omega_0\frac{\sigma_z}{2}, \tag{1}$$

where $\lambda = \langle n\rangle$ is the expected number of photons, which can be written in terms of a magnetic field amplitude B_1 oscillating at frequency ω,

$$\lambda = \frac{B_1^2 V}{8\pi\hbar\omega}. \tag{2}$$

The first term represents the energy in the oscillating field, the second term represents the energy of the interaction of the oscillating field with the spin, and the third term is the energy of the interaction of the spin with the holding field. The Pauli matrices in this system are chosen so that the diagonal state is in the direction of the strong oscillating field,

$$\sigma_x = \begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix} = |+\rangle\langle+| - |-\rangle\langle-|, \tag{3}$$

$$\sigma_y = \begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix} = |+\rangle\langle-| + |-\rangle\langle+|, \tag{4}$$

$$\sigma_z = \begin{pmatrix}0 & -i\\ i & 0\end{pmatrix} = -i|+\rangle\langle-| + i|-\rangle\langle+|. \tag{5}$$

An ensemble of photons of a harmonic field is described by Glauber states such that the coefficients of the probability amplitudes in terms of the average photon number $\lambda = \langle n\rangle$ are

$$a_n = \exp\left(-\frac{\lambda}{2}\right)\frac{\lambda^{\frac{n}{2}}}{(n!)^{\frac{1}{2}}}. \tag{6}$$

The basis in m_z can be written in terms of the m_x eigen states,

$$\overline{|n, m_z\rangle} = \frac{1}{\sqrt{2}}\overline{|n_+\rangle}|+\rangle_x + i m_z\overline{|n_-\rangle}|-\rangle_x, \tag{7}$$

where

$$\overline{|n_+\rangle} = e^{-\frac{1}{2}\frac{\eta}{\omega}(a^\dagger - a)}|n\rangle, \tag{8}$$

$$\overline{|n_-\rangle} = e^{\frac{1}{2}\frac{\eta}{\omega}(a^\dagger - a)}|n\rangle, \tag{9}$$

and $|n\rangle$ is the basis for the quantum simple harmonic oscillator Hamiltonian. Furthermore, from Refs. [1,2], we have

$$\overline{\langle n'_+|n_+\rangle} = \delta_{n'n}, \tag{10}$$

$$\overline{\langle n'_-|n_-\rangle} = \delta_{n'n}, \tag{11}$$

$$\begin{aligned}\overline{\langle n'_+|n_-\rangle} &= \langle n'|e^{-\frac{1}{2}\frac{\eta}{\omega}(a - a^\dagger)}e^{\frac{1}{2}\frac{\eta}{\omega}(a^\dagger - a)}|n\rangle\\ &= \langle n'|e^{\frac{\eta}{\omega}(a^\dagger - a)}|n\rangle \approx J_{n'-n}\left(\frac{\gamma B_1}{\omega}\right),\end{aligned} \tag{12}$$

where $J_{n'-n}$ is the Bessel function of the first kind with order $n' - n$. In Refs. [1,2] they find the energy,

$$\frac{E_n}{\hbar} = n\omega + \frac{\omega'_0 m}{2} - \frac{\eta^2}{4\omega}, \tag{13}$$

where here, and for the rest of the document, $m = m_z$ with $m = \pm 1$ and η is given by

$$\eta = \frac{\gamma B_1}{2\lambda^{1/2}}. \tag{14}$$

It is then shown in Refs. [1,2] that the Larmor precession (ω_0) has been effectively scaled ($\omega_0 \to \omega'_0$) and in the limit of $\frac{\omega_0}{\omega} \ll 1$ the scaling factor reduces to

$$\omega'_0 = J_0\left(\frac{\gamma B_1}{\omega}\right)\omega_0, \tag{15}$$

which we refer to as the J_0 approximation. Note that this approximation breaks down when $\frac{\omega_0}{\omega} \ll 1$ is not valid. This is discussed in Refs. [16] and [17] and will be addressed below.

The wave function of the system is written as a sum of the basis states weighted by Glauber coefficients,

$$\psi(t) = \sum_{n,m=\pm 1} e^{in\omega t + i\frac{1}{2}m\omega'_0 t}\frac{a_n}{\sqrt{2}}\overline{|n, m\rangle}. \tag{16}$$

Notice that the last term of the energy in Eq. (13) is a constant energy shift and thus does not contribute to the dynamics; this is the reason for its absence in the phase of the wave function in Eq. (16). Furthermore, the changes in η are suppressed by a factor ω and $\lambda^{\frac{1}{2}}$, this allows us to add an energy shift as a perturbation in the Hamiltonian while introducing negligible error. We now present a brief summary of critical dressing and show the results of optimization within a simulation of critical spin dressing for a system of neutrons and ^{3}He.

A. Critical dressing

In the presence of two spin species with different gyromagnetic ratios, e.g., γ_j, γ_k the dressing parameters (B_1, ω) can be tuned such that the effective Larmor precession ($\omega'_0 = \gamma' B_0$) is the same for the two species, i.e., $\gamma'_j = \gamma'_k$. In the J_0 approximation this can be achieved if we define $\alpha = \gamma_j/\gamma_k$; then, in terms of the dressing parameter $x = \gamma B_1/\omega$, critical dressing occurs when $x = x_c$ where x_c is found by solving the following equation,

$$J_0(x_c) = \alpha J_0(\alpha x_c). \tag{17}$$

However, if the J_0 approximation is not valid then the effective gyromagnetic ratio can be calculated numerically to high accuracy by writing the matrix elements of the Hamiltonian over a large range of n and n' and then diagonalizing the resulting matrix. The resulting eigen values of the diagonalized matrix will determine the Zeeman splitting where the nth entry is the order of the perturbation. This method is further discussed in Refs. [16] and [17]. Thus, the effective frequency can be determined from

$$\omega'_0 = \frac{\Delta E}{\hbar}. \tag{18}$$

Where ΔE is the Zeeman splitting determined numerically from nth order perturbation theory.

TABLE I. Critical dressing parameters for a range of ω values where B_{J_0} is the solution to critical dressing given by Ref. [5], B_{pt} the solution found numerically from higher-order perturbation theory, and B_{sim} is the critical dressing field found using simulations. The value of B_{sim} was optimized such that $\gamma_n' = \gamma_{\text{He}}'$ to within 1 part per 10^{10}. This corresponds to θ changing by less than 5×10^{-5} rad over 1000 s. All the above were optimized using a $B_0 = 3\ \mu$T, where, in this case, $\omega_0' \sim 380$ rad/s.

ω (rad s^{-1})	$\frac{\omega}{\omega_0}$	B_{J_0} (μT)	B_{pt} (μT)	B_{sim} (μT)	$\frac{B_{\text{sim}}-B_{J_0}}{B_{J_0}}\times 100\%$	$\frac{B_{\text{sim}}-B_{\text{pt}}}{B_{\text{pt}}}\times 100\%$
100000	164	648.8541	648.8433379	648.8345280	−0.003	-1.3578×10^{-3}
30000	49.1	194.6562	194.6202953	194.6176552	−0.020	-1.3565×10^{-3}
18000	29.4	116.7937	116.7338304	116.7322440	−0o.053	-1.3590×10^{-3}
10000	16.4	64.8854	64.7775066	64.7766232	−0.168	-1.3637×10^{-3}
6000	9.81	38.9313	38.7511230	38.7505920	−0.466	-1.3704×10^{-3}
4200	6.87	27.2519	26.9938929	26.9935194	−0.948	-1.3836×10^{-3}
3000	4.91	19.4656	19.1026874	19.1024180	−1.866	-1.4100×10^{-3}
2400	3.93	15.5725	15.1162445	15.1160267	−2.931	-1.4410×10^{-3}
2100	3.43	13.6259	13.1019402	13.1017478	−3.847	-1.4682×10^{-3}
1800	2.94	11.6794	11.0633252	11.0631580	−5.276	-1.5115×10^{-3}

First observations of critical spin dressing are discussed in Ref. [18], which is a thorough reference for critical spin dressing. Here we present results from Monte Carlo simulations optimized for the critical dressing parameters of neutrons and ^{3}He over a wide range of ω and where, in this case, $\alpha = \gamma_3/\gamma_n \approx 1.11$ and γ_3, γ_n are the ^{3}He and neutron gyromagnetic ratios. The optimization was performed until the effective gyromagnetic ratios differ by less than 1 part in 10^{10}. It was observed that the J_0 approximation is overall very good for $\omega \gg \omega_0$; however, as the dressing frequency approaches the holding field frequency, differences between the simulation and the J_0 approximation can grow large. Monte Carlo results are compared to numerically calculated higher order perturbation theory, and the J_0 approximation in Table I.

While for estimation purposes the J_0 approximation is fast and fairly accurate, for precision measurements it may not be accurate enough, for example, in the search for permanent electric dipole moments.

B. Critical dressing applied to the neutron electric dipole moment

A specific case where critical dressing is of particular interest is in a system of neutrons and ^{3}He for the search for the neutron electric dipole moment, due to a spin-dependent interaction of neutrons and ^{3}He. Making use of the spin-dependent interaction via critical spin dressing is a sensitive probe of the relative phase of the ^{3}He and neutrons. This is further discussed in Refs. [5,19].

In the absence of spin dressing the frequency of precession in a uniform magnetic field (B_0) and electric field (E), given the existence of a permanent neutron electric dipole moment (d_n), with magnetic dipole moment μ_n is given as

$$\omega_0 = \frac{2\mu_n B_0}{\hbar} + \frac{2d_n E}{\hbar}. \tag{19}$$

However, in the case of critical spin dressing the precession frequency will be modified according to Eq. (18). The experimental signature of a nonzero d_n is a difference in precession frequency for electric and magnetic fields parallel versus antiparallel,

$$\Delta\omega = \omega_{\uparrow\uparrow}' - \omega_{\uparrow\downarrow}', \tag{20}$$

where $\omega_{\uparrow\uparrow}'$ is the frequency when the magnetic and electric field are parallel and $\omega_{\uparrow\downarrow}'$ is the frequency when the magnetic field and electric field are antiparallel. In the limit where the J_0 approximation is valid the frequency shift from the electric dipole moment, d_n, can be written in terms of the Bessel function J_0,

$$\Delta\omega = J_0(x_c)\frac{4d_n E}{\hbar}. \tag{21}$$

Beyond the J_0 approximation, ω_0' can be determined numerically as discussed above or from the Monte Carlo simulation. We illustrate the onset of the breakdown of the J_0 approximation in Fig. 1, where it is shown that the precession frequency shift determined from the simulation is best predicted by the numerical diagonalization of a higher-order perturbation theory where the shift due to the electric field is included in the Hamiltonian used to determine the perturbed Zeeman splitting. From this result we might assume that this numerical method can be used to predict the relaxation and frequency shifts that are introduced with inhomogeneities in the holding fields. However this technique will not provide an accurate prediction, because the frequency shift is an extrinsic effect, it is determined from the strength of the spectrum for the correlation function at the Larmor frequency. Since the frequency has been changed by the spin dressing, we may also expect the contribution from the correlation function to change as well. Therefore a more detailed investigation of the relaxation and frequency shifts due to inhomogeneities of the field is desired; in the following section we present a model for this purpose.

III. RELAXATION AND FREQUENCY SHIFTS FROM FIELD INHOMOGENEITIES IN A SPIN-DRESSED SYSTEM

We assume here that Eq. (16) is the solution to the Hamiltonian; for $\omega_0 \ll \omega$ the J_0 approximation is valid. When that condition is broken the exact expansion must be used, or alternately, computed to high accuracy numerically, this is discussed in Sec. II A. We use this solution to add additional

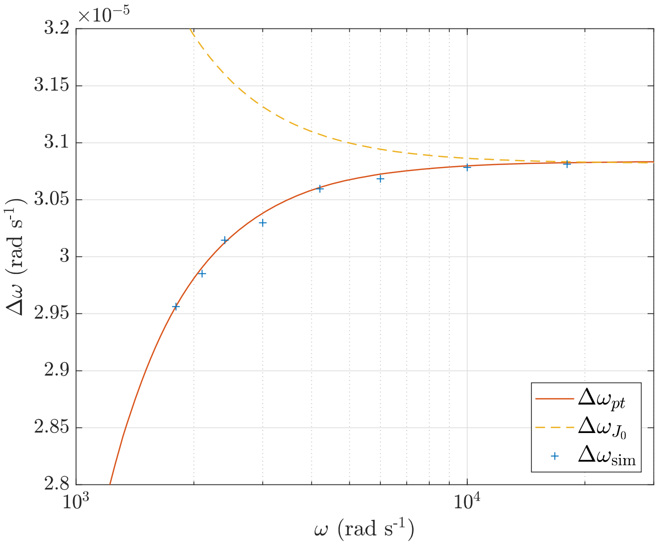

FIG. 1. Frequency shift due to a neutron EDM of $d_n = 1 \times 10^{-25}$ e cm and 75 kV/cm electric field in a critically dressed system of ^{3}He and neutrons. Excellent agreement is observed between the shift predicted from the simulation ($\Delta\omega_{\rm sim}$) and the higher order perturbation theory ($\Delta\omega_{\rm pt}$). The deviation from the simulation and the shift predicted from the J_0 approximation ($\Delta\omega_{J_0}$) is apparent at lower frequencies. The deviation from a constant for the J_0 approximation is due to deviation of x_c from that given by Eq. (17). For these calculations $B_0 = 3\ \mu$T giving $\omega_0' \sim 380$ rad/s

perturbations in the Hamiltonian, these perturbations take the form of an electric field $\mathbf{E} = E\hat{z}$, and inhomogeneous field functions for the spin-dressing B_1 oscillating at a frequency ω, and DC holding field B_0. We will discuss solutions in the particle rest frame such that spatial B-field inhomogeneities couple with the particle motion to become time-dependent variations. The perturbation is thus time varying $\langle n\rangle = \lambda(t)$. However, this will be represented by a change in the field $B_1 \to \langle B_1\rangle + \delta B_1(t)$, where for now $\delta B_1(t)$ is some arbitrary function of time, but is small compared to $\langle B_1\rangle$. A similar term for the Hamiltonian occurs in Ref. [20], where they also model the gradient of a dressing field. Recall that this change is a negligible shift in the total energy, shifting all levels by the same amount at any instant, thus the overall dynamics can be well described if we include the term $\frac{\gamma\delta B_1(t)}{\langle\lambda\rangle^{\frac{1}{2}}}\frac{\sigma_x}{4}(a^\dagger + a)$, this will result in a time-dependent energy but does not introduce significant error as long as $\gamma\delta B_1$ is small compared to ω and γB_1. Interestingly, $\gamma\delta B_1$ need not be small compared to γB_0. Furthermore, we require $\omega \gg \omega_0$ if we wish to use the approximation in Eq. (15). For simplicity we use the notation

$$\omega_{1i}(t) = \gamma\delta B_{1i}(t), \tag{22}$$

$$\omega_{0i}(t) = \gamma\delta B_{0i}(t) + \gamma v_j E_z/c, \tag{23}$$

where the i index represents the fluctuations due to the field in that direction, the number index determines the source, the 0 index indicates it is generated from inhomogeneities of the holding field, while terms with the 1 index are generated from the dressing field which oscillates at frequency ω. With this simplification, the perturbing Hamiltonian is written as

$$H_{\rm pert} = \frac{1}{4\lambda^{1/2}}[\omega_{1x}(t)\sigma_x + \omega_{1y}(t)\sigma_y + \omega_{1z}(t)\sigma_z](a + a^\dagger) + \omega_{0x}(t)\frac{\sigma_x}{2} + \omega_{0y}(t)\frac{\sigma_y}{2} + \omega_{0z}(t)\frac{\sigma_z}{2}. \tag{24}$$

Thus, the total Hamiltonian is written as

$$\frac{H}{\hbar} = \omega a^\dagger a + \frac{\omega_1}{4\lambda^{1/2}}\sigma_x(a + a^\dagger) + H_{\rm pert}. \tag{25}$$

We continue to evaluate this Hamiltonian to second order in perturbation theory for all off-diagonal matrix elements and diagonal elements oscillating at ω, assuming a constant energy for the time evolution. This is equivalent to neglecting the $\omega_{0z}(t)\sigma_z$ term in Eq. (24). We include the DC field diagonal matrix elements that arise from the $\omega_{0z}(t)\sigma_z$ term in Sec. III B, where time evolution in the energy is required, this is discussed in detail in that section. We will find expressions for frequency shifts, and transverse relaxation, T_2. We do no explicitly consider T_1, because it does not seem to hold much interest given that we are considering the spin-dressed system of a gas, which has typically undergone a $\pi/2$ pulse. We will identify the leading terms that contribute to T_2, and show that this model is qualitatively in agreement with previous formulations found in reference [12,21–23] considering the effects of spin dressing.

A. Relaxation and frequency shifts in second-order perturbation theory

We will write the relaxation and frequency shifts in terms of the wave function determined from perturbation theory. To do this we will find the expectation value of the spins aligned in the plane perpendicular to the DC holding field,

$$\langle\sigma_x + i\sigma_y\rangle = \langle\sigma_+\rangle. \tag{26}$$

From the real part we find T_2 relaxation and the imaginary part the accumulated phase due to the frequency shift. Despite deriving the matrix elements in the representation where the diagonal Pauli Matrix is along x, we write them in terms of a basis diagonal in z. We start with the spins in the plane of Larmor precession, for example, after a $\frac{\pi}{2}$ pulse. Since we are now in the z basis, we have

$$\psi^{(0)} = \frac{1}{\sqrt{2}}\begin{pmatrix}1\\1\end{pmatrix} = \frac{1}{\sqrt{2}}(|+\rangle + |-\rangle), \tag{27}$$

where to first order the elements of the wave function are, after having summed over the Glauber states,

$$\psi^{(1)} = -i\int_0^t dt'|f\rangle\langle f|H|i\rangle dt', \tag{28}$$

the contribution to second order is

$$\psi^{(2)} = (-i)^2\int_0^t dt'\int_0^{t'} dt''|f\rangle\langle f|H|k\rangle\langle k|H|i\rangle dt'dt'', \tag{29}$$

so that our total perturbed wave function to second order is

$$\psi = \psi^{(0)} + \psi^{(1)} + \psi^{(2)}. \tag{30}$$

Our task is the evaluation of the matrix elements of the perturbing Hamiltonian. However, we can be more restrictive of the terms required in our final calculation if we consider a general perturbed wave function,

$$\psi = \begin{pmatrix} a \\ b \end{pmatrix}, \tag{31}$$

from this we find the expectation value of $\langle\sigma_+\rangle$,

$$\langle\sigma_+\rangle = 2a^*b. \tag{32}$$

Now, $\langle\sigma_+\rangle$ can be written in terms of the matrix elements used to build ψ. For this notation we write $\langle +|H|+\rangle = H_{++}$, and $\langle -|H|+\rangle = H_{-+}$, etc. The evaluation and discussion of $\langle\sigma_+\rangle$ is completed in the Appendix, where we find

$$\langle\sigma_+\rangle = 1 - 2t\int_0^t H_{-+}(0)H_{+-}(\tau)d\tau - 4t\mathrm{Re}\left(\int_0^t H_{--}(0)H_{--}(\tau)d\tau\right). \tag{33}$$

From this we find the frequency shift,

$$\delta\omega = \mathrm{Im}\left(\frac{d\langle\sigma_+\rangle}{dt}\right) = \mathrm{Im}\left(2\int_0^t H_{-+}(0)H_{+-}(\tau)d\tau\right), \tag{34}$$

$$\delta\omega = 2\mathrm{Im}\left(\int_0^\infty H_{-+}(0)H_{+-}(\tau)d\tau\right), \tag{35}$$

where we assumed the observation time is much longer than the correlation time to change the limit from t to ∞. Reference [11] asserts that the equations are valid for intermediate times, as we find here; however, we will continue with the Fourier transform due to their simplicity. For the transverse relaxation, we have

$$\frac{1}{T_2} = 2\mathrm{Re}\left[\int_0^\infty H_{-+}(0)H_{+-}(\tau)d\tau\right) + 4\mathrm{Re}\left[\int_0^t H_{--}(0)H_{--}(\tau]d\tau\right]. \tag{36}$$

The matrix elements for all unique terms in the Hamiltonian are evaluated in the Appendix. The results are presented here:

$$\langle m'|\sigma_x|m\rangle = \frac{1}{2}(1-mm')e^{i\frac{1}{2}(m-m')\omega_0't}, \tag{37}$$

$$\langle m'|\sigma_x(a+a^\dagger)|m\rangle = \lambda^{\frac{1}{2}}e^{i\frac{1}{2}(m-m')\omega_0't}\cos(\omega t)(1-mm'), \tag{38}$$

$$\langle m'|\sigma_y|m\rangle = \frac{i}{2}e^{i\frac{1}{2}(m-m')\omega_0't}(m-m')J_0\left(\frac{\gamma B_1}{\omega}\right) + (m+m')\sin(\omega t)J_1\left(\frac{\gamma B_1}{\omega}\right), \tag{39}$$

$$\langle m'|\sigma_y(a+a^\dagger)|m\rangle = i\lambda^{\frac{1}{2}}J_0\left(\frac{\gamma B_1}{\omega}\right)e^{i\frac{1}{2}(m-m')\omega_0't}\cos(\omega t)(m-m'), \tag{40}$$

$$\langle m'|\sigma_z|m\rangle = \frac{1}{2}(m+m')e^{i\frac{1}{2}(m-m')\omega_0't} - i(m-m')e^{i\frac{1}{2}(m-m')\omega_0't} \times J_1\left(\frac{\gamma B_1}{\omega}\right)\sin(\omega t), \tag{41}$$

$$\langle m'|\sigma_z(a+a^\dagger)|m\rangle = e^{i\frac{1}{2}(m-m')\omega_0't}\lambda^{\frac{1}{2}}J_0\left(\frac{\gamma B_1}{\omega}\right)\cos(\omega t)(m+m'). \tag{42}$$

The matrix elements shown above are all in terms of the J_0 approximation, if more accuracy is required the dressing factor $[J_0(\gamma B_1/\omega)]$ should be replaced by a dressing factor found numerically using Eq. (18). For simplicity we write the result as a double sum of possible terms. The cross terms do not vanish, if there is a field shape that depends on the same variable, these can be written in terms of the correlation of a function of the same variable [e.g., $B'_{1x} \propto f(x)$, $B'_{1y} \propto g(x)$], then the fields will be correlated. These terms only vanish in special cases, e.g., for $B'_{1x} \propto x$, with $B'_{1y} \propto x^2$. We ignore cross terms between the DC and AC fields as these terms always contain some rapidly oscillating phase and can be neglected, this is shown in the Appendix. The relaxation can be written in terms of modified variables $\omega_{1j} \rightarrow \omega'_{1j}$, specifically,

$$\omega'_{1x}(t) = \gamma\delta B_{1x}(t), \tag{43}$$

$$\omega'_{1y}(t) = i\gamma J_0(\langle x\rangle)\delta B_{1y}(t), \tag{44}$$

$$\omega'_{1z}(t) = \gamma J_0(\langle x\rangle)\delta B_{1z}(t), \tag{45}$$

$$\omega'_{0x}(t) = \gamma\left[\delta B_{0x}(t) + \frac{v_y(t)}{c^2}E\right], \tag{46}$$

$$\omega'_{0y}(t) = i\gamma J_0(\langle x\rangle)\left[\delta B_{0y}(t) + \frac{v_x(t)}{c^2}E\right], \tag{47}$$

$$\omega'_{0z}(t) = \gamma J_0(\langle x\rangle)\delta B_{0z}(t), \tag{48}$$

$$\omega'_{0zq}(t) = \gamma J_1(\langle x\rangle)\delta B_{0z}(t), \tag{49}$$

where ω'_{0zq} corresponds to the first harmonic of the oscillating terms in the matrix elements, which arise due to the dressing field; this is discussed in the Appendix. Notice that $\omega'_{1x}(t) = \omega_{1x}(t)$, because x corresponds to the direction of the applied dressing field, it does not obtain a factor $J_0(x)$. With these definitions we can write our phase shift as

$$\delta\omega = 2\mathrm{Im}\left[\frac{1}{8}\sum_{k=x,y}\sum_{j=x,y}\int_0^\infty \omega'_{1j}(0)\omega'_{1k}(\tau)\cos(\omega\tau)e^{-i\omega_0'\tau}d\tau\right] \tag{50}$$

$$+ 2\mathrm{Im}\left[\frac{1}{4}\sum_{k=x,y}\sum_{j=x,y}\int_0^\infty \omega'_{0j}(0)\omega'_{0k}(\tau)e^{-i\omega_0'\tau}d\tau\right]. \tag{51}$$

Notice that there is no cross correlation between the AC and DC terms, ω_{1i} and ω_{0k}, in the frequency shift. This is because a quickly oscillating phase appears in all cross correlation terms in the expansion, this is shown in the Appendix. Thus, there is no linear in E frequency shift generated by gradients in the dressing field. Typically, the first term(s) can be ignored, and the second term(s) dominates the frequency shift. We turn our attention to the transverse relaxation where we hold off on writing the zero frequency term for the DC field inhomogeneity. This is investigated in Sec. III B, where we show that for the zero frequency part of the DC relaxation we must include the time dependent energy in the Hamiltonian. The current method does not take into account the time variation of the energy. This fails for the diagonal DC field component (ω'_{0z}), because $\omega'_{0z}(t)$ is a relatively large contribution to the overall phase. In fact, given that this is a static contribution to the diagonal matrix element, it is the whole contribution of the phase for that term, and while it is small compared to $\langle\omega_0\rangle$, $\langle\omega_1\rangle$, and ω, it cannot be ignored when it is the sole contribution to the phase. For the off-diagonal terms, or diagonal terms oscillating at ω, the energy dependence of the time evolution operator can be ignored due to the negligible size of the phase shift compared to $\langle\omega'_0\rangle$, and ω. Starting from the evaluation of $\langle\sigma_+\rangle$ in terms of the wave function constructed from second-order perturbation theory, we find (after omitting ω'_{0z})

$$\frac{1}{T_2} = 2\mathrm{Re}\left[\frac{1}{4}\sum_{k=x,y}\sum_{j=x,y}\int_0^\infty \omega'_{1j}(0)\omega'_{1k}(\tau)\cos(\omega\tau)e^{-i\omega'_0\tau}d\tau\right] \tag{52}$$

$$+2\mathrm{Re}\left[\frac{1}{4}\sum_{k=x,y}\sum_{j=x,y}\int_0^\infty \omega'_{0j}(0)\omega'_{0k}(\tau)e^{-i\omega'_0\tau}d\tau\right] \tag{53}$$

$$+4\mathrm{Re}\left[\frac{1}{2}\int_0^\infty \omega'_{1z}(0)\omega'_{1z}(\tau)\cos(\omega\tau)d\tau\right] \tag{54}$$

$$+\mathrm{Re}\left[i\int_0^\infty e^{-i\omega'_0\tau}\sin(\omega\tau)\omega'_{1x}(0)\omega'_{0zq}(\tau)d\tau\right] \tag{55}$$

$$+\mathrm{Re}\left[i\int e^{-i\omega'_0\tau}\sin(\omega\tau)\omega'_{0zq}(0)\omega'_{0zq}(\tau)d\tau\right]. \tag{56}$$

In general, there is no reason a field in one direction will not be correlated with another over the same position coordinate. For example, Maxwell's equation does not preclude the relation $\frac{dB_x}{dy}\propto\frac{dB_y}{dy}$, in this case these two field inhomogeneities would be correlated due to a similar dependence on the y-position variable, and thus the cross terms must be included for an accurate prediction of the relaxation. Furthermore, we will not consider the terms found in Eqs. (55) and (56) any further. These terms contribute as the difference in the real part of the spectrum at $\omega \pm \omega'_0$ and are highly suppressed for typical spin-dressing parameters, when $\omega \gg \omega'_0$, but we included them here to maintain generality.

The transverse relaxation that we find is nearly in agreement with the formulation found in Ref. [21], with the exception that our contribution from the y and z field power spectrum contain an extra factor $J_0^2(\langle x\rangle)$ due to dressing. We now finish the derivation of T_2 by examining terms in the diagonal component of the Hamiltonian containing ω_{0z} without assuming a dressed energy that is constant in time. We will find that diagonal terms typically dominate the relaxation, this is because typically the magnitude of the gradient for the on-axis field component is similar to the off-axis component, so that all other terms can be ignored. However, we should be aware that there are solutions to Maxwell's equation where this assumption is not valid.

B. T_2 due to spatial inhomogeneities in a spin-dressing and holding field

Now we create a model to determine transverse relaxation (T_2) that incorporates diagonal heterogeneities in the spin-dressing field. This is not considered in the previous section or in Ref. [21], where the AC gradients that cause relaxation spatially average to zero. The model requires that we incorporate our time dependence into the energy of the state determined from the Hamiltonian, given by

$$H = \omega_z(t)\frac{\sigma_z}{2}. \tag{57}$$

Here, $\omega_z = \gamma J_0[x(t)][\langle B_0\rangle + \delta B_0(t)]$ and the time evolution operator is given as

$$U(\delta t, 0) = \exp\left\{-i\gamma J_0(x(t))[\langle B_0\rangle + \delta B_0(t)]\frac{\sigma_z}{2}\delta t\right\}. \tag{58}$$

Alternatively, one could proceed with the propagator given in Ref. [8], where they account for nonharmonic waveforms, or in Ref. [10], where they account for a distribution of frequencies. However, we do not consider these because typically for systems investigated here the precision and stability of the frequency and generated waveform is far greater than the field uniformity. The general solution is

$$|\alpha\rangle = c_+|+\rangle_z + c_-|-\rangle_z. \tag{59}$$

We start with the spins along the $+x$ direction, equivalent to a system directly after a $\frac{\pi}{2}$ pulse, our solution becomes

$$|\alpha\rangle = \frac{1}{\sqrt{2}}\left(\exp\left\{-i\gamma\int_0^t J_0[x(t')]B_0(t')dt'\right\}|+\rangle + \exp\left\{i\gamma\int_0^t J_0[x(t')]\delta B_0(t')dt'\right\}|-\rangle\right). \tag{60}$$

To find an expression for T_2, we evaluate $\langle\sigma_x + i\sigma_y\rangle = \langle\sigma_+\rangle$ and find its rate of decay,

$$\langle\alpha, t|\sigma_+|\alpha, t=0\rangle = \frac{1}{2}\left(\langle-|\exp\left\{-i\gamma\int_0^t J_0[x(t'')][B_0(t'')]dt''\right\} + \langle+|\exp\left\{i\gamma\int_0^t J_0[x(t'')][\delta B_0(t'')]dt''\right\}\right)$$
$$\times |+\rangle\langle-|\left(\exp\left\{-i\gamma\int_0^t J_0[x(t')][B_0(t')]dt'\right\}|+\rangle + \exp\left\{i\gamma\int_0^t J_0[x(t')][\delta B_0(t')]dt'\right\}\right)|-\rangle. \tag{61}$$

We concentrate on the phase in the exponential. To simplify the time dependence in the Bessel function, we write

$$\int d\omega_\phi = \gamma \int d\left\{J_0\left(\frac{\gamma\langle B_1\rangle + \omega_{1x}}{\omega}\right)\left[B_0 + \delta B_{0z}(t)\right]\right\}, \tag{62}$$

$$\omega_\phi(t) = \gamma \int J_1\left(\frac{\gamma\langle B_1\rangle}{\omega}\right)\frac{d\omega_{1x}}{\omega}[B_0 + \delta B_{0z}(t)] + \gamma \int J_0\left(\frac{\omega_1}{\omega}\right) d\delta B_{0z}(t), \tag{63}$$

$$\omega_\phi(t) \simeq \frac{\gamma}{\omega}\int J_1\left(\frac{\gamma\langle B_1\rangle}{\omega}\right)[B_0 d\omega_{1x} + \delta B_{0z}(t) d\omega_1' x] + \gamma \int J_0\left(\frac{\gamma\langle B_1\rangle}{\omega}\right) d\delta B_{0z}(t), \tag{64}$$

$$\omega_\phi(t) \simeq \frac{\gamma}{\omega}\int J_1\left(\frac{\gamma\langle B_1\rangle}{\omega}\right) B_0 d\omega_{1x} + \gamma \int J_0\left(\frac{\gamma\langle B_1\rangle}{\omega}\right) d\delta B_{0z}(t), \tag{65}$$

$$\omega_\phi(t) \simeq \frac{\gamma}{\omega} J_1\left(\frac{\gamma\langle B_1\rangle}{\omega}\right) B_0\omega_{1x} + \gamma J_0\left(\frac{\gamma\langle B_1\rangle}{\omega}\right)\delta B_{0z}(t). \tag{66}$$

We substitute this back into Eq. (61),

$$\langle\sigma_+\rangle = \exp\left\{i\int_0^t \left[\frac{\gamma}{\omega} J_1\left(\frac{\gamma\langle B_1\rangle}{\omega}\right) B_0\omega_{1x}(t') + \gamma J_0\left(\frac{\gamma\langle B_1\rangle}{\omega}\right)\delta B_0(t')\right] dt'\right\}. \tag{67}$$

This is the general form of the zero-frequency decay component of T_2. Theoretically, if we know $B_0(t)$, then the solution is known. However, perfect knowledge of $B_0(t)$ for every particle is challenging. Thus, we describe thermal motion by a conditional probability distribution function, a model that is appropriate for this derivation is described by Refs. [13,24]. The evaluation of $\langle\sigma_+\rangle$ is completed in the Appendix, and diagonal DC field contribution to T_2 is

$$\frac{1}{T_{2\ \mathrm{DC}}} = \mathrm{Re}\left[S_{\omega'_{0z}\omega'_{0z}}(0) \right. \tag{68}$$

$$+ J_1(x)^2\frac{\omega_0^2}{\omega^2} S_{\omega_{1x}\omega_{1x}}(0) \tag{69}$$

$$\left. - 2J_1(x)\frac{\omega_0}{\omega} S_{\omega_{1x}\omega'_{0z}}(0)\right]. \tag{70}$$

Again, we use $\omega_0' = \gamma J_0(x) B_0$, where $S_{\omega_0'\omega_0'}(0)$ is the zero frequency of the spectrum of the autocorrelation function of $\omega_0'(t)$. In general, we define the spectrum to be

$$S_{\omega_{ij}\omega_{kl}}(\omega) = \int_0^\infty \langle\omega_{ij}(0)\omega_{kl}(\tau)\rangle e^{-i\omega\tau} d\tau. \tag{71}$$

Defining the spectrum from $t = 0 \to \infty$ allows us to keep the imaginary parts, which are necessary for the prediction of the frequency shift.

The DC diagonal rate is summed with the off-diagonal and AC diagonal rates for the full prediction of T_2,

$$\frac{1}{T_2} = \mathrm{Re}\left\{\frac{1}{4}\sum_{k=x,y}\sum_{j=x,y}\left[S_{\omega'_{1j}\omega'_{1k}}(\omega+\omega_0') + S_{\omega'_{1j}\omega'_{1k}}(\omega-\omega_0')\right] + \frac{1}{2}\sum_{k=x,y}\sum_{j=x,y} S_{\omega'_{0j}\omega'_{0k}}(\omega_0') + S_{\omega'_{1z}\omega'_{1z}}(\omega) + S_{\omega'_0\omega'_0}(0) + J_1(x)^2\frac{\omega_0^2}{\omega^2} S_{\omega_{1x}\omega_{1x}}(0) - 2J_1(x)\frac{\omega_0}{\omega} S_{\omega_{1x}\omega'_{0z}}(0)\right\}. \tag{72}$$

In the case of the cross correlation $S_{\omega_{1x}\omega'_{0z}}(0)$ we will find that if the variation in ω_{1x} and ω'_{0z} is due to spatial inhomogeneities then only terms that are a function of the same variable will be correlated. For example, if there is a spin-dressing field gradient along x, and the static field gradient along x then the cross correlation will not vanish, $[S_{\omega_1'\omega_0'}(0) \neq 0]$. This formulation of the relaxation offers the curious ability to make the cross term the opposite sign as the squared terms, implying that one can slow down and essentially cancel the DC relaxation as long as the gradients in each field are of the right proportion. The spin-dressing gradient that allows this cancellation, i.e., $\frac{1}{T_2} \approx 0$, corresponds to

$$G_{1xj} = G_{0zj}\frac{\omega}{\omega_0'}\frac{J_0(x)}{J_1(x)}, \quad \text{for} \quad \frac{1}{T_2} \approx 0, \tag{73}$$

where $G_{1xj} = \frac{dB_1x}{dj}$ and j can be x, y, or z, and for the DC field gradient, $G_{0zj} = \frac{dB_0z}{dj}$. Physically, the gradient in the spin-dressing field changes the effective gyromagnetic ratio in the correct proportion to the gradient in the holding field allowing the spins to have the same effective frequency throughout the whole volume. When the spins have the same effective frequency across the spatial volume the relaxation from these terms vanishes. Matching the effective gyromagnetic precession across the volume to extend coherence times has recently been reported in Ref. [25], where they independently predict and experimentally verify that a spin-dressed inhomogeneity can counteract an applied DC inhomogeneity to recover the original coherence times, even in the presence of large gradients. Therefore this result is not limited to the Redfield regime as presented here, and is valid where the signal decay becomes dominated by the reversible process of gradient dephasing. We expand on this and claim that from Eq. (72) it is clear that the attenuation of the polarization due to the motion of the spins is also suppressed, making the technique technically superior when compared to refocusing the spins with spin echo. However, some relaxation is unavoidable, because the AC relaxation terms, specifically from the first line in Eq. (72), are dominant when the cancellation is sufficient.

C. Comparison to simulations

Monte Carlo simulations coupled with a fifth-order Runge-Kutta integrator are compared to the theoretical predictions for the relaxation and frequency shifts of the spin-dressed system. The simulation package is further described in Refs. [14,15]. The simulation is for a system of ^{3}He in a superfluid ^{4}He bath

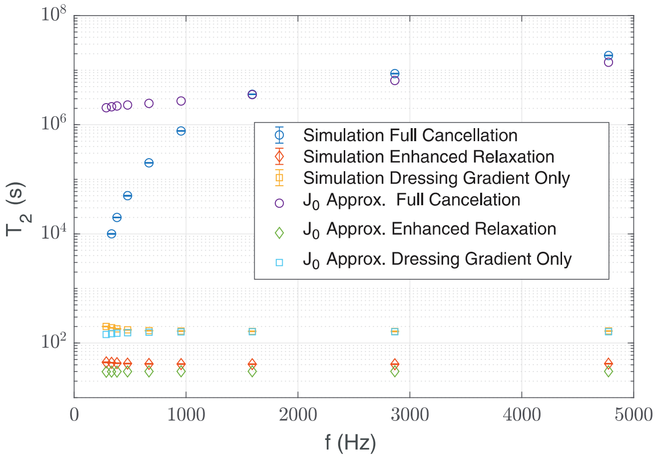

FIG. 2. T_2 in the J_0 approximation due to a spin-dressing gradient, showing full theoretical cancellation. Expectedly, full theoretical cancellation does not result in an infinite T_2 as the AC terms in the relaxation become the dominant source of relaxation. Otherwise, results agree within the spin-dressing $J_0(\omega_1/\omega)$ approximation, the deviation observed is due to this approximation breaking down. The gradient in the uniform field is 3×10^{-5} B_0/cm in the z direction, and the AC gradient is determined from Eq. (73).

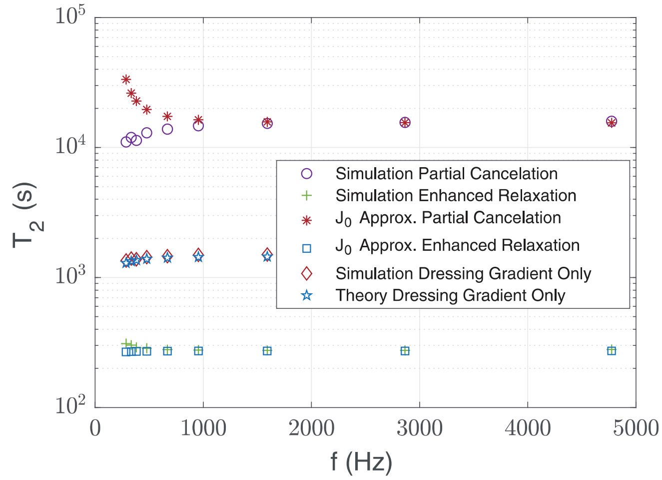

FIG. 3. T_2 in the J_0 approximation due to a spin-dressing gradient, showing partial cancellation, 3% effective remnant gradient according to Eq. (73). The theory points are circles and the simulation data are stars, results agree until the Bessel function approximation breaks down due to relatively slow AC frequency compared to the Larmor frequency. The gradient in the uniform field is 1×10^{-5} B_0/cm in the z direction and $B_0 = 3\ \mu$T

below 450 mK, as this is the system used to search for the neutron electric dipole moment (discussed in Sec. II B and Refs. [5,19]). This superfluid solution is chosen because of the ability to change the diffusion coefficient of the ^{3}He (D_3), via scattering off of phonons, by small changes in temperature (T). In Refs. [26,27] we find $D_3 \propto T^{-7}$. The ability to change the diffusion coefficient and thus, the mean free path, allows the ability to study the effect of the particle's trajectory on the relaxation and frequency shifts. With the intention of mitigating unwanted relaxation or frequency shifts. The volume in the simulation is confined to a rectangular cell of dimensions, (x, y, z), $40 \times 10.2 \times 7.6$ cm.

The vanishing relaxation effect, simulated with gradients in both the holding and dressing fields simultaneously, is shown in Figs. 2 and 3. Full cancellation of the holding field gradient ($G_{zz} = 3 \times 10^{-5}$ B_0/cm) by the dressing field gradient is shown in in Fig. 2. In Fig. 3, the spin-dressing gradient is tuned to partially cancel the holding field gradient ($G_{zz} = 10^{-5}$ B_0/cm), with 3% residual gradient remaining according to Eq. (73). Although the gradient cancellation is not exact in Fig. 3, it still shows a sizable gain in coherence time. For this example, in either of the holding field gradients, the applied gradient in the AC field need only be stable to $\approx 5\%$ of its target value, found from Eq. (73), for a substantial gain in the dressed coherence time. As expected, total cancellation is not observed in Fig. 2 as the AC field terms become dominant. The cross term that extends the relaxation time can also enhance the relaxation rate so that relaxation is faster than the sum of the individual rates. The enhanced relaxation is shown in Figs. 3 and 2, where the gradient in Eq. (73) has the opposite sign. Deviation from the simulations is observed in both Figs. 3 and 2 as the dressing frequency decreases. This is due to the approximation in Eq. (15) becoming invalid. Presumably this discrepancy is largely removed by using the full expansion found in Ref. [2], or by finding it numerically as described in Sec. II A. The slope of the J_0 approximation around the critical dressing parameter x_c is also required. Nonetheless, a substantial gain in T_2 is observed. This effect allows the ability to change the relaxation rate by either tuning the dressing field gradient or the holding field gradient. Typically for dressed systems we have $\frac{\omega}{\omega_0} > \frac{J_0(x)}{J_1(x)}$. Thus, a change in the dressing field gradient will change the relaxation at a slower rate compared to the holding field gradient. Therefore the resolution for changing the relaxation is smaller for the dressing field gradient, making it technically easier to manipulate the relaxation through manipulation of that gradient. However, it requires tuning an AC gradient to be in phase with the original dressing field. With modern timing resolution this problem is certainly solvable, but achieving the stability required may be challenging. Furthermore, we can determine the size of the gradient in the spin-dressing or holding field by keeping the gradient in one constant and varying the gradient of the other.

Typically, the zero frequency components of the field will dominate the relaxation, however there are solutions to Maxwell's equation where this is not the case. Thus, simulations for gradients in ω_{1x} and ω_{1y}, were performed and compared to the theory. This is shown in Fig. 4.

For the simulations of the phase shift we include an electric field $\mathbf{E} = E\,\hat{z}$, and include a magnetic gradient in the z direction with the negative of the x direction, $G_{0zz} = -G_{0xx}$. This example scenario is simple but realistic since it satisfies Maxwell's equations, a condition that can broken in a simulation. This gradient is efficient in examining the model as it will give us the minimum number of similarly behaving terms, and thus the most direct in comparison to simulations. For this case, we have the following terms in the Hamiltonian that must be considered:

$$\omega'_{1_z}(t) = \gamma J_0(\langle x \rangle) G_{1zz} z(t), \tag{74}$$

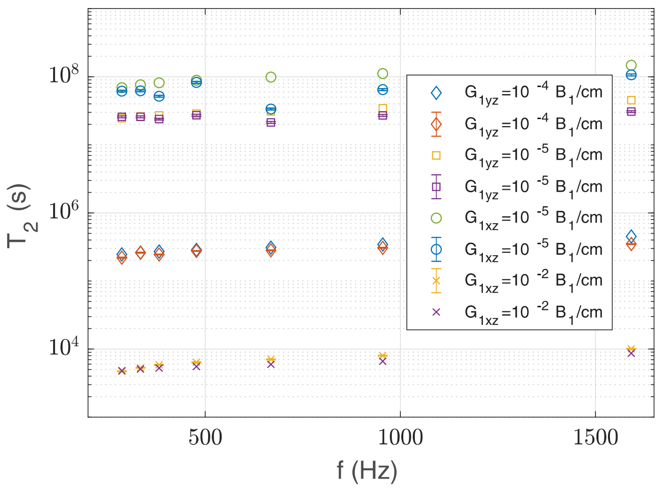

FIG. 4. Comparison of T_2 due to gradients in the off-axis components of the spin-dressing field for the numerically solved dressing factor versus the simulation. Each gradient was simulated across a range of dressing frequencies. The circles indicate theory prediction and the error bars are from the simulation. There is no deviation observed at the lower frequencies because the dressing factor was computed numerically for the simulation and theory, rather than using the J_0 approximation.

$$\omega'_{1x}(t) = \gamma G_{1xx} x(t), \tag{75}$$

$$\omega'_{0z} = \gamma J_0(\langle x\rangle) G_{0zz} z(t), \tag{76}$$

$$\omega'_{0x} = \gamma \left[G_{0xx} x(t) + \frac{v_y(t)}{c^2} E \right], \tag{77}$$

$$\omega'_{0y}(t) = i\gamma J_0(\langle x\rangle) \frac{v_x(t)}{c^2} E. \tag{78}$$

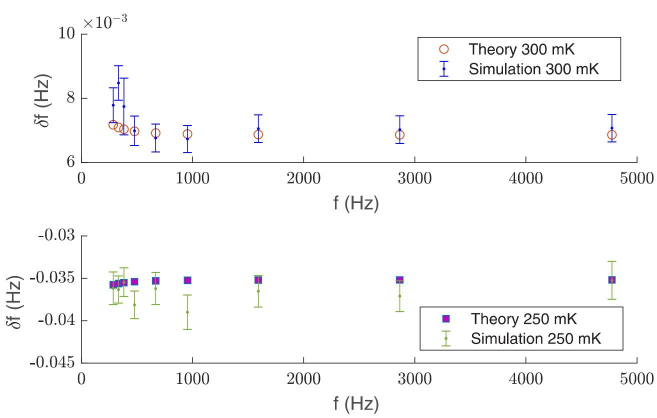

FIG. 5. Linear in E frequency shift in a rectangular cell of $0.4 \times 0.102 \times 0.076$ m (x, y, z) dimensions, respectively. The spins contained in the cell are under the influence of a dressed field. The upper plot is modeling ^{3}He in a superfluid helium-II bath at 250 mK in a 2 μT holding field along the z direction. The lower plot is for the same system 300 mK in a 3 μT holding field. The E field is 750 kV/cm along the z direction and the DC field gradient is 2 μT/m along the x direction, this gradient is large compared to gradients achieved in laboratory fields to increase the effect above the resolution of the simulation. In this simulation the EDM is set to zero.

For this example, we ignore contributions of $\frac{E^2}{c^4}$ as typically these are negligible, although we should be cautious in simulations not to make E large enough to become the dominant term when searching for other effects. Typically, by making E unphysically large we magnify the desired linear in E effect (for simulations purposes), but it can be obscured if the relaxation and frequency shifts due to E^2 become dominant. If we assume that E^2 terms can be neglected, and keeping only terms linear in E, we find

$$\delta\omega = \frac{\gamma^2}{c^2} E G_{0xx} J_0(\langle x\rangle) \mathrm{Re}\left[\int_0^\infty x(0) v_x(\tau) e^{-i\omega'_0 \tau} \right], \tag{79}$$

$$\delta\omega = -\frac{\gamma^2}{c^2} E G_{0xx} J_0(\langle x\rangle) \times \left\{ \omega'_0 \mathrm{Im}\left[\int_0^\infty x(0) x(\tau) e^{-i\omega'_0 \tau} \right] + \frac{L_x^2}{12} \right\}, \tag{80}$$

$$\delta\omega = -\frac{\gamma^2}{c^2} E G_{0xx} J_0(\langle x\rangle) \left\{ \omega'_0 \mathrm{Im}[(S_{xx}(\omega)] + \frac{L_x^2}{12} \right\}. \tag{81}$$

This was simulated for the range of critically dressed frequencies found in Table I. In Fig. 5 we find good agreement that is consistent with the combined error of the simulation and extraction of the frequency shift.

We now examine the effect of modulation of the spin-dressing parameters on the phase shifts and relaxation.

D. Modulated critical dressing

Modulated critical spin dressing is the modulation of parameters of the critically dressed spin system around the critically dressed value. It is a technique that can be utilized to optimize statistical precision and mitigate systematic drifts. Experimental observation of the systematic improvement is presented in Ref. [18]. In Ref. [28] they find pulsed modulation to be the optimum modulation technique for statistical sensitivity. In pulse modulated dressing the gyromagnetic frequencies of the two spin species are momentarily allowed to shift to a large difference so that a known angle θ_m is accumulated between the spins. When θ_m is accumulated an observation period of critical dressing is continued for a given time. After this time has been completed the frequency change is reversed and the angle θ_m is undone. It is shown in Refs. [19,28] that a statistically optimum value for the angle θ_c between the spins exists. The modulated dressing is used to swing neutron and ^{3}He spins to $\pm\theta_c$. For example, this is achieved at the start of the critical dressing when the initial phase between the spin species is equal to $-\theta_c$, after the critical dressing observation period the modulation pulse is applied where $\theta_m = 2\theta_c$ bringing the phase to θ_c, after another observation period a pulse $\theta_m = -2\theta_c$ is applied. The pulse train repeats for the duration of the measurement. For more details refer to Refs. [5,19].

The modulation technique was optimized across a number of different modulation strategies and pulses. It is clear from modern timing resolution that frequency modulation is most precise experimentally. Initially square pulses in the frequency ω of the pulse were pursued as the modulation technique, however this was repeatedly shown to have poor coherence times. Despite great effort, a set of parameters that approached

TABLE II. Optimized parameters for range of ω_c values for the modulation function given in Eqs. (83) and (84). For all frequencies $n = 51$ was used and $B_{\rm rf}$ values are the same as the $B_{\rm sim}$ values found in Table I. θ_0 is the time average value of θ between modulation pulses and $\sqrt{\langle\Delta\theta^2\rangle}$ is the rms in θ between modulation pulses. These parameters were tuned such that θ_0 changes by less than $5{\times}10^{-5}$ rad between up and down pulses after 1000 s.

ω (rad s^{-1})	$\omega_{\rm amp}$ (rad s^{-1})	a	θ_0 (rad)	$\sqrt{\langle\Delta\theta^2\rangle}$ (rad)
30 000	12711.5053	0.7256	0.7998	0.012
18 000	7624.44225	0.7257	0.7998	0.012
10 000	4231.97665	0.7260	0.7998	0.012
6000	2542.33540	0.7260	0.8022	0.013
4200	1766.76950	0.7280	0.7999	0.014
3000	1255.05106	0.7300	0.8012	0.015
2400	994.364100	0.7330	0.8006	0.018
2100	862.250665	0.7357	0.7996	0.019
1800	730.012950	0.7395	0.7995	0.020

acceptable behavior was never determined for the square pulses, despite an analytic solution in Ref. [5]. It was found that if the square pulses where smoothed the modulation of frequency would remain coherent, and ultimately no relaxation or distortion was observable after 10^3 modulation cycles. Of the functions studied the parametric function that demonstrates the best performance is

$$\omega_{\rm var}(r) = \omega + \omega_{\rm amp} f(r), \tag{82}$$

where

$$f(r) = \frac{1}{a} e^{-n[r(t)-\frac{\pi}{2}]^2} - a e^{-n[r(t)-\frac{3\pi}{2}]^2} \tag{83}$$

and

$$r(t) = \mathrm{mod}_{2\pi}(\omega_{\rm fm} t + \phi_m), \tag{84}$$

where $\omega_{\rm amp}$ is the amplitude and $\omega_{\rm fm}$ is the frequency of the modulation (about 1 Hz), ϕ_m is a modulation phase, the parameter a controls the relative heights of the positive and negative modulation pulses, and n controls the sharpness of the peaks. The integral of this pulse can be written in closed form with the use of the error function, which is a well tabulated function. Using this modulation pulse the required parameters of the pulse for the desired effect on the spin solution can be calculated accurately, and quickly, leaving the possibility of feedback timing corrections during the measurement. A table for the modulation parameters for a range of ω is shown in Table II. A time sequence plot of the pulse train for a particular frequency ($\omega = 10\,000$) and the response of the spins are shown in Fig. 6.

Pulsed modulation corrects for slow electronic drifts in the dressing parameters, specifically drifts slower than the frequency of modulation $f_{\rm fm} \approx 1$ Hz. This is discussed further in the Appendix. However, modulation does not decrease sensitivity to phase shifts compared to a dressed system that is not modulated. This includes the frequency shift due to an electric dipole moment and any geometric frequency shifts, nor will it increase T_2. Therefore, we do not expect the analysis of the theory to change other than by the amount prescribed by the theory due to the changing dressing pulse that enables

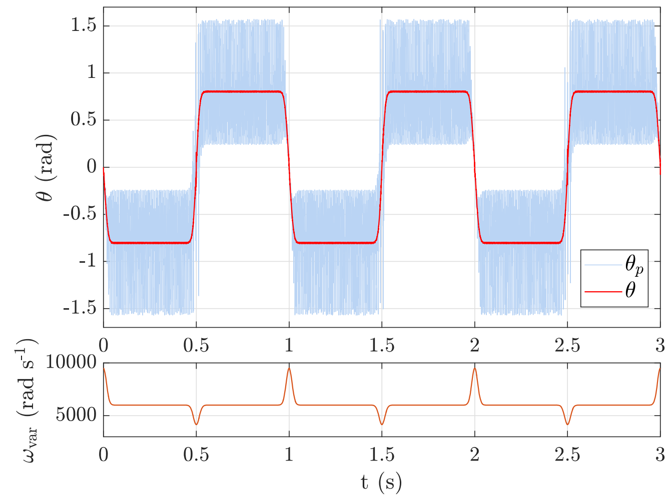

FIG. 6. Plot of θ, θ_p and $\omega_{\rm var}$ against time for three complete modulation cycles where parameters are given by those in Table II for $\omega_c = 10\,000$ rad s^{-1} and $\phi_{\rm mod} = \pi/2$. θ is the total angle between the two species' spins and θ_p is the angle between the projection of the two species' spins on the plane perpendicular to B_0.

the modulation. The geometric phase was simulated under the modulation parameters shown in Table II and compared to the theory; the results are shown in Fig. 7.

IV. CONCLUSION

An analysis of critical dressing was completed in simulations, values that optimize the critical dressing are proposed, and are found to be in very close agreement with theory. Furthermore, an example of modulation was proposed for a

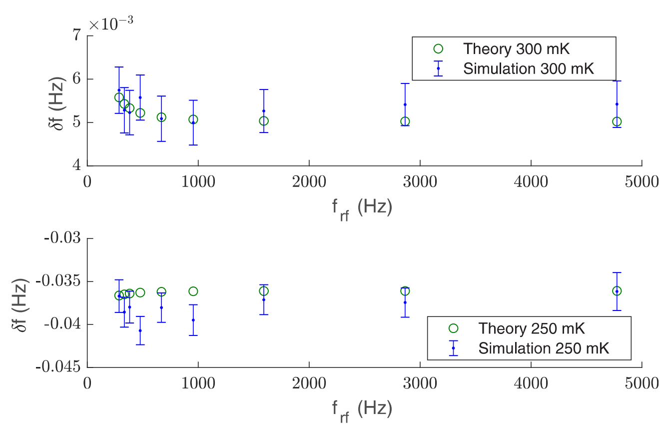

FIG. 7. Linear in E frequency shift for modulated spin dressing in a rectangular cell of 0.4 × 0.102 × 0.076 m (x, y, z) dimensions, respectively. The spins contained in the cell are under the influence of a modulated dressed field. The upper plot is modeling ^{3}He in a superfluid helium-II bath at 250 mK in a 2 μT holding field along the z direction. The lower plot is for the same system 300 mK in a 3 μT holding field. The E field is 750 kV/cm along the z direction and the magnetic field gradient is 2 μT/m along the x direction; this gradient is large compared to gradients achieved in laboratory fields to increase the effect above the resolution of the simulation. In this simulation the EDM is set to zero.

specific pulse shape over various critical dressing frequencies and shown to be stable for periods long compared to relaxation times in laboratories. The effect of field inhomogeneities of a dressed system was investigated, simulations are compared to a new analytical model.

A model that incorporates Redfield-like gradient relaxation and frequency shifts into a spin-dressed system has been proposed and compared to simulations. Good agreement between the analytical model and the simulations is observed, allowing confidence in fast estimations without the use of a spin-dressing simulation. This provides a means of predicting observables, in situ, that is to say, in a time frame that is much faster than the transverse relaxation of a typical run of an experiment. Full and accurate simulations of dressed spin systems typically take several days to complete due to the computational complexity, this complexity is further discussed in Ref. [14].

We highlight the prediction to cancel DC gradient relaxation by an AC gradient relaxation, a rare scenario where two wrongs make a right. It also gives rise to the unfortunate ability to create a relaxation rate that is faster than the sum of the two individual rates. Nonetheless, this technique holds promise of achieving better than previously expected coherence times given an ambient field inhomogeneity.

Finally, the model elucidates the correct approach for calculating systematic frequency shifts; where we find the correlation function is evaluated at the dressed energy splitting, and not the intrinsic Zeeman splitting that arises from the holding field alone. Furthermore, it is shown that there is no linear in E frequency shift generated by gradients in the dressing field.

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APPENDIX

Here we evaluate the matrix elements given the Hamiltonian in Eq. (25). The perturbative terms can be classified by those proportional to $\sigma_x, \sigma_y,\ \sigma_z,\ \sigma_x(a+a^\dagger)$, etc. For simplicity we consider the contributions from the operators separately, first we consider the matrix elements of $\langle m'|\sigma_y|m\rangle$,

$$\overline{\langle n', m'|}\sigma_y\overline{|n, m\rangle} = \frac{1}{2}e^{\cdots}a^*_{n'}a_n(\overline{\langle n'_+|}\langle +|_x - im'\overline{\langle n'_-|}\langle -|_x)(\sigma_y)(\overline{|n_-\rangle}|+\rangle_x + im\overline{|n_-\rangle}_+|-\rangle_x), \tag{A1}$$

$$\overline{\langle n', m'|}\sigma_y\overline{|n, m\rangle} = \frac{1}{2}e^{\cdots}a^*_{n'}a_n(\overline{\langle n'_+|}\langle +|_x - im'\overline{\langle n'_-|}\langle -|_x)(|+\rangle\langle -| + |-\rangle\langle +|)(\overline{|n\rangle}_+|+\rangle_x + im\overline{|n\rangle}_-|-\rangle_x),$$

$$\overline{\langle n', m'|}\sigma_y\overline{|n, m\rangle} = \frac{1}{2}e^{\cdots}a^*_{n'}a_n(\overline{\langle n'_+|}\langle +|_x|+\rangle\langle -|im\overline{|n_-\rangle}|-\rangle_x - im'\overline{\langle n'_-|}\langle -|_x|-\rangle\langle +|\overline{|n_+\rangle}|+\rangle_x), \tag{A2}$$

$$\overline{\langle n', m'|}\sigma_y\overline{|n, m\rangle} = \frac{1}{2}e^{\cdots}ia^*_{n'}a_n(m\overline{\langle n'_+|n_-\rangle} - m'\overline{\langle n'_-|n_+\rangle}), \tag{A3}$$

$$\overline{\langle n', m'|}\sigma_y\overline{|n, m\rangle} = \frac{1}{2}e^{i(n-n')\omega t + i\frac{1}{2}(m-m')\omega'_0 t}ia^*_{n'}a_n\left[mJ_{n'-n}\left(\frac{\gamma B_1}{\omega}\right) - m'J_{n-n'}\left(\frac{\gamma B_1}{\omega}\right)\right]. \tag{A4}$$

Now we set $n' = n + q$, and sum over n, giving us $\langle m'|\sigma_y|m\rangle$,

$$\langle m'|\sigma_y|m\rangle = \sum_n\sum_q \frac{1}{2}e^{i(n-n')\omega t + i\frac{1}{2}(m-m')\omega'_0 t}ia^*_{n+q}a_n\left[mJ_q\left(\frac{\gamma B_1}{\omega}\right) - m'J_{-q}\left(\frac{\gamma B_1}{\omega}\right)\right]. \tag{A5}$$

We keep all the terms, for now, but note here large values of q do not contribute due to the behavior of the a_n coefficients, and q determines the harmonic of the term while large harmonics will not contribute at our desired sensitivity. We continue to simplify the expression,

$$\langle m'|\sigma_y|m\rangle = \sum_{q=-\infty}^{\infty} \frac{1}{2}e^{iq\omega t + i\frac{1}{2}(m-m')\omega'_0 t}i\left[mJ_q\left(\frac{\gamma B_1}{\omega}\right) - m'J_{-q}\left(\frac{\gamma B_1}{\omega}\right)\right]. \tag{A6}$$

We note here that large values of q do not contribute due to the behavior of the a_n coefficients, and q determines the harmonic of the term. We find that it is enough to only consider the first harmonic where $q = \pm 1$,

$$\langle m'|\sigma_y|m\rangle = \frac{i}{2}e^{i\frac{1}{2}(m-m')\omega'_0 t}(m-m')J_0\left(\frac{\gamma B_1}{\omega}\right) + (m+m')\sin(\omega t)J_1\left(\frac{\gamma B_1}{\omega}\right). \tag{A7}$$

Now we turn to σ_x,

$$\begin{aligned}
&\overline{\langle n',m'|}\sigma_x\overline{|n,m\rangle}, && \text{(A8)}\\
&= \frac{1}{2}e^{i(n-n')\omega t+i\frac{1}{2}(m-m')\omega_0' t}a_{n'}^* a_n(\overline{\langle n'|}_+\langle +|_x - im\overline{\langle n'|}_-\langle -|_x)(\sigma_x)(\overline{|n\rangle}_+|+\rangle_x + im\overline{|n\rangle}_-|-\rangle_x),\\
&= \frac{1}{2}e^{\cdots}a_{n'}^* a_n(\overline{\langle n'|_+\langle +|_x} - im\overline{\langle n'|}_-\overline{\langle -|_x})(|+\rangle\langle +| - |-\rangle\langle -|)(\overline{|n\rangle}_+\overline{|+\rangle}_x + im\overline{|n\rangle}_-\overline{|-\rangle}_x), && \text{(A9)}\\
&= \frac{1}{2}e^{i(n-n')\omega t+i\frac{1}{2}(m-m')\omega_0' t}a_{n'}^* a_n(\overline{\langle n'|_+}\langle +|_x|+\rangle\langle +|\overline{|n\rangle}_+\overline{|+\rangle}_x + i^2 m'm\overline{\langle n'|}_-\overline{\langle -|_x|n\rangle}_+\overline{|-\rangle}_x), && \text{(A10)}\\
&= \frac{1}{2}(1-mm')e^{i\frac{1}{2}(m-m')\omega_0' t}a_{n'}^* a_n\delta_{n'n}, && \text{(A11)}
\end{aligned}$$

summing over n and n' we have

$$\langle m'|\sigma_x|m\rangle = \sum_n \frac{1}{2}(1-mm')e^{i\frac{1}{2}(m-m')\omega_0' t}a_{n'}^* a_n\delta_{n'n}, \tag{A12}$$

$$\langle m'|\sigma_x|m\rangle = \frac{1}{2}(1-mm')e^{i\frac{1}{2}(m-m')\omega_0' t}. \tag{A13}$$

Now that we have a simple expression of the matrix elements required for $\langle\sigma_x\rangle$ we consider $\sigma_x(a+a^\dagger)$, where we must commute $a+a^\dagger$ with $e^{\mp\frac{1}{2}\frac{\eta}{\omega}(a^\dagger-a)}$.

$$\begin{aligned}
&\sum_{m=\pm1}\sum_{m'=\pm1}\overline{\langle n',m'|}\sigma_x(a+a^\dagger)\overline{|n,m\rangle}, && \text{(A14)}\\
&= \sum_{m=\pm1}\sum_{m'=\pm1}\frac{1}{2}e^{i\frac{1}{2}(m-m')\omega_0' t}(\overline{\langle n'_+|}\langle +|_x - im'\overline{\langle n'_-|}\langle -|_x)(\sigma_x)(a+a^\dagger)(\overline{|n_+\rangle}|+\rangle_x + im\overline{|n_-\rangle}|-\rangle_x), && \text{(A15)}\\
&= \sum_{m=\pm1}\sum_{m'=\pm1}\frac{1}{2}e^{\cdots}(\overline{\langle n'_+|}\langle +|_x - im'\overline{\langle n'_-|}\langle -|_x)(|+\rangle\langle +| - |-\rangle\langle -|)(a+a^\dagger)(\overline{|n_+\rangle}|+\rangle_x + im\overline{|n_-\rangle}|-\rangle_x),\\
&= \sum_{m=\pm1}\sum_{m'=\pm1}\frac{1}{2}e^{i\frac{1}{2}(m-m')\omega_0' t}(\overline{\langle n'_+|}\langle +|_x|+\rangle\langle +|(a+a^\dagger)\overline{|n_+\rangle}|+\rangle_x + i^2 m'm\overline{\langle n'_-|}\langle -|_x(a+a^\dagger)\overline{|n_-\rangle}|-\rangle_x),\\
&= \sum_{m=\pm1}\sum_{m'=\pm1}e^{i\frac{1}{2}(m-m')\omega_0' t}\frac{1}{2}(\overline{\langle n'_+|}(a+a^\dagger)\overline{|n_+\rangle} - m'm\overline{\langle n'_-|}(a+a^\dagger)\overline{|n_-\rangle}), && \text{(A16)}
\end{aligned}$$

Concentrating on the evaluation of $(a+a^\dagger)\overline{|n_\pm\rangle}$, we start by writing $\overline{|n_\pm\rangle}$ in terms of $|n\rangle$,

$$(a+a^\dagger)\overline{|n_\pm\rangle} = (a+a^\dagger)e^{\mp\frac{1}{2}\frac{\eta}{\omega}(a^\dagger-a)}|n\rangle. \tag{A17}$$

For simplicity, set $w=\frac{1}{2}\frac{\eta}{\omega}$, $c=a+a^\dagger$, and $b=a^\dagger-a$. Notice the similarities of b and c with the position and momentum operators of a simple harmonic oscillator,

$$x = \sqrt{\frac{\hbar}{2m\omega}}(a+a^\dagger) = \sqrt{\frac{\hbar}{2m\omega}}c \tag{A18}$$

and

$$p = i\sqrt{\frac{m\omega\hbar}{2}}(a^\dagger-a) = i\sqrt{\frac{m\omega\hbar}{2}}b. \tag{A19}$$

We know that $[x,p]=i\hbar$, thus,

$$[c,b]=2, \tag{A20}$$

with the commutation relation,

$$[c,e^{\mp wb}] = \mp 2we^{\mp wb}, \tag{A21}$$

the matrix elements are

$$\begin{aligned}
&\overline{\langle n'_\pm|}(a+a^\dagger)\overline{|n_\pm\rangle} = \langle n'|\mp 2w + c|n\rangle,\\
&\overline{\langle n'_\pm|}(a+a^\dagger)\overline{|n_\pm\rangle} = \mp\frac{\eta}{\omega}\delta_{n'n} + \sqrt{n}\langle n'|n-1\rangle + \sqrt{n+1}\langle n'|n+1\rangle. && \text{(A22)}
\end{aligned}$$

Evaluating $\langle m'|\sigma_x(a+a^\dagger)|m\rangle$, we have

$$\langle m'|\sigma_x(a+a^\dagger)|m\rangle = \frac{1}{2}\sum_n\sum_{n'} e^{i(n-n')\omega t+i\frac{1}{2}(m-m')\omega_0' t} a_{n'}a_n(\overline{\langle n'_+|}(a+a^\dagger)\overline{|n_+\rangle} - m'm\overline{\langle n'_-|}(a+a^\dagger)\overline{|n_-\rangle}), \tag{A23}$$

$$\langle m'|\sigma_x(a+a^\dagger)|m\rangle = \frac{1}{2}\sum_n\sum_{n'} e^{\cdots}a_{n'}a_n\Big[\sqrt{n}\delta_{n'n-1}+\sqrt{n+1}\delta_{n'n+1}-\frac{\eta}{\omega}\delta_{n'n}-m'm\Big(\sqrt{n}\delta_{n'n-1}+\sqrt{n+1}\delta_{n'n+1}+\frac{\eta}{\omega}\delta_{n'n}\Big)\Big], \tag{A24}$$

$$\langle m'|\sigma_x(a+a^\dagger)|m\rangle = \frac{1}{2}\sum_n\sum_{n'} e^{\cdots}a_{n'}a_n\Big[\sqrt{n}\delta_{n'n-1}+\sqrt{n+1}\delta_{n'n+1}-\frac{\eta}{\omega}\delta_{n'n}-m'm\Big(\sqrt{n}\delta_{n'n-1}+\sqrt{n+1}\delta_{n'n+1}+\frac{\eta}{\omega}\delta_{n'n}\Big)\Big]. \tag{A25}$$

We proceed by summing over n,

$$\langle m'|\sigma_x(a+a^\dagger)|m\rangle = \frac{1}{2}\sum_n\sum_{n'} e^{i(n-n')\omega t}a_{n'}a_n e^{i\frac{1}{2}(m-m')\omega_0' t} \tag{A26}$$

$$\times\Big[\Big(\sqrt{n+1}\delta_{n'n+1}+\sqrt{n}\delta_{n'n-1}-\frac{\eta}{\omega}\delta_{n'n}|m-m'|\Big)(1-m'm)\Big], \tag{A27}$$

$$\langle m'|\sigma_x(a+a^\dagger)|m\rangle = \frac{1}{2}e^{i\frac{1}{2}(m-m')\omega_0' t}\left[\left(\sum_n e^{i\omega t}a_{n-1}a_n\sqrt{n}+\sum_n e^{-i\omega t}a_{n+1}a_n\sqrt{n+1}\right)(1-m'm)-\sum_n|a_n|^2\frac{\eta}{\omega}\delta_{nn}|m-m'|\right], \tag{A28}$$

$$\langle m'|\sigma_x(a+a^\dagger)|m\rangle = \frac{1}{2}e^{i\frac{1}{2}(m-m')\omega_0' t}\Big[(e^{i\omega t}\sqrt{\lambda}+e^{-i\omega t}\sqrt{\lambda})(1-m'm)-\frac{\eta}{\omega}\delta_{nn}|m-m'|\Big], \tag{A29}$$

$$\langle m'|\sigma_x(a+a^\dagger)|m\rangle = \lambda^{\frac{1}{2}}e^{i\frac{1}{2}(m-m')\omega_0' t}\left[\cos(\omega t)(1-m'm)-\frac{\eta}{\lambda^{\frac{1}{2}}\omega}|m-m'|\right]. \tag{A30}$$

Where in the second-to-last step with large λ the Poisson distribution behaves like a Dirac δ function in n, numerically this is found to be an extremely good approximation. We continue by noting that the term containing η carries a $1/\lambda$ which is the number of photons in the field, which is very large, and so the term with η can be neglected. We have

$$\langle m'|\sigma_x(a+a^\dagger)|m\rangle = \lambda^{\frac{1}{2}}e^{i\frac{1}{2}(m-m')\omega_0' t}\cos(\omega t)(1-mm'). \tag{A31}$$

Now we find

$$\overline{\langle n',m'|}\sigma_y(a+a^\dagger)\overline{|n,m\rangle} \tag{A32}$$

$$= \frac{1}{2}e^{i(n-n')\omega t}e^{i\frac{1}{2}(m-m')\omega_0' t}(\overline{\langle n'_+|}\langle+|-im'\overline{\langle n'_-|}\langle-|)(\sigma_x)(a+a^\dagger)(\overline{|n_+\rangle}|+\rangle+im\overline{|n_-\rangle}|-\rangle), \tag{A33}$$

$$= \frac{1}{2}e^{\cdots}(\overline{\langle n'_+|}\langle+|-im'\overline{\langle n'_-|}\langle-|)(|+\rangle\langle-|+|-\rangle\langle+|)(a+a^\dagger)(\overline{|n_+\rangle}|+\rangle+im\overline{|n_-\rangle}|-\rangle),$$

$$= \frac{1}{2}e^{\cdots}(\overline{\langle n'_+|}\langle+||+\rangle\langle-|-im'\overline{\langle n'_-|}\langle-||-\rangle\langle+|)(a+a^\dagger)(\overline{|n_+\rangle}|+\rangle+im\overline{|n_-\rangle}|-\rangle),$$

$$= \frac{1}{2}e^{\cdots}(\overline{\langle n'_+|}\langle-|-im'\overline{\langle n'_-|}\langle+|)(a+a^\dagger)(\overline{|n_+\rangle}|+\rangle+im\overline{|n_-\rangle}|-\rangle), \tag{A34}$$

$$= e^{i(n-n')\omega t}e^{i\frac{1}{2}(m-m')\omega_0' t}\frac{i}{2}(m\overline{\langle n'_+|}(a+a^\dagger)\overline{|n_-\rangle}-m'\overline{\langle n'_-|}(a+a^\dagger)\overline{|n_+\rangle}). \tag{A35}$$

Therefore, we must find

$$\overline{\langle n'_\mp|}(a+a^\dagger)\overline{|n_\pm\rangle} = \overline{\langle n'_\mp|}c\overline{|n_\pm\rangle} = \overline{\langle n'_\mp|}ce^{\mp\frac{1}{2}\frac{\eta}{\omega}b}|n_\pm\rangle. \tag{A36}$$

With the commutation relation

$$[c, e^{\mp wb}] = \mp 2we^{\mp wb}, \tag{A37}$$

we have

$$\overline{\langle n'_{\mp}|}c\overline{|n_{\pm}\rangle} = \overline{\langle n'_{\mp}|}e^{\mp\frac{1}{2}\frac{\eta}{\omega}b}c|n\rangle \mp \frac{\eta}{\omega}J_0(x), \tag{A38}$$

$$\overline{\langle n'_{\mp}|}c\overline{|n_{\pm}\rangle} = \sqrt{n}\overline{\langle n'_{\mp}|}e^{\mp\frac{1}{2}\frac{\eta}{\omega}b}|n-1\rangle + \sqrt{n+1}\overline{\langle n'_{\mp}|}e^{\mp\frac{1}{2}\frac{\eta}{\omega}b}|n+1\rangle \mp \frac{\eta}{\omega}J_0(x) \tag{A39}$$

$$= \sqrt{n}J_{n'-n+1}\left(\frac{\gamma B_1}{\omega}\right)\delta_{n'n-1} + \sqrt{n+1}J_{n'-n-1}\left(\frac{\gamma B_1}{\omega}\right)\delta_{n'n-1} \mp \frac{\eta}{\omega}J_0(x)\delta_{n'n}. \tag{A40}$$

We see that the term containing η is negligible compared to the other two terms. Substituting this back in and summing over n and n', we have

$$\langle m'|\sigma_y(a+a^\dagger)|m\rangle = \frac{i}{2}\sum_n\sum_{n'} e^{i\frac{1}{2}(m-m')\omega_0' t}, \tag{A41}$$

$$\times a_{n'}a_n\left\{me^{i(n-n')\omega t}\left[\sqrt{n}J_{n'-n+1}\left(\frac{\gamma B_1}{\omega}\right)\delta_{n'n-1} + \sqrt{n+1}J_{n'-n-1}\left(\frac{\gamma B_1}{\omega}\right)\delta_{n'n-1}\right]\right. \tag{A42}$$

$$\left.-m'e^{i(n-n')\omega t}\left[\sqrt{n}J_{n'-n+1}\left(\frac{\gamma B_1}{\omega}\right)\delta_{n'n-1} + \sqrt{n+1}J_{n'-n-1}\left(\frac{\gamma B_1}{\omega}\right)\delta_{n'n-1}\right]\right\}, \tag{A43}$$

$$\langle m'|\sigma_y(a+a^\dagger)|m\rangle = \frac{i}{2}e^{i\frac{1}{2}(m-m')\omega_0' t}\left\{m\left[e^{i\omega t}\sqrt{\lambda}J_0\left(\frac{\gamma B_1}{\omega}\right) + e^{-i\omega t}\sqrt{\lambda}J_0\left(\frac{\gamma B_1}{\omega}\right)\right]\right.$$

$$\left.- m'\left[e^{i\omega t}\sqrt{\lambda}J_0\left(\frac{\gamma B_1}{\omega}\right) + e^{-i\omega t}\sqrt{\lambda}J_0\left(\frac{\gamma B_1}{\omega}\right)\right]\right\}, \tag{A44}$$

$$\langle m'|\sigma_y(a+a^\dagger)|m\rangle = ie^{i\frac{1}{2}(m-m')\omega_0' t}\lambda^{\frac{1}{2}}J_0\left(\frac{\gamma B_1}{\omega}\right)\cos(\omega t)(m-m'). \tag{A45}$$

This is expected if we consider the comparison of the $q=0$ elements with $\langle m'|\sigma_x|m\rangle$ to $\langle m'|\sigma_y|m\rangle$. We now consider

$$\langle m'|\sigma_z(a+a^\dagger)|m\rangle = \sum_n\sum_{n'}\overline{\langle n',m'|}\sigma_z(a+a^\dagger)\overline{|n,m\rangle}. \tag{A46}$$

Concentrating on individual terms of sum, we have

$$\overline{\langle n',m'|}\sigma_z(a+a^\dagger)\overline{|n,m\rangle} \tag{A47}$$

$$= \frac{1}{2}e^{i(n-n')\omega t}e^{i\frac{1}{2}(m-m')\omega_0' t}(\overline{\langle n'_+|}\langle +| - im'\overline{\langle n'_-|}\langle -|)(\sigma_x)(a+a^\dagger)(\overline{|n_+\rangle}|+\rangle + im\overline{|n_-\rangle}|-\rangle), \tag{A48}$$

$$= \frac{i}{2}e^{\cdots}(\overline{\langle n'_+|}\langle +| - im'\overline{\langle n'_-|}\langle -|)(-|+\rangle\langle -| + |-\rangle\langle +|)(a+a^\dagger)(\overline{|n_+\rangle}|+\rangle + im\overline{|n_-\rangle}|-\rangle),$$

$$= \frac{i}{2}e^{\cdots}(-\overline{\langle n'_+|}\langle +||+\rangle\langle -| - im'\overline{\langle n'_-|}\langle -||-\rangle\langle +|)(a+a^\dagger)(\overline{|n_+\rangle}|+\rangle + im\overline{|n_-\rangle}|-\rangle),$$

$$= \frac{i}{2}e^{\cdots}(-\overline{\langle n'_+|}\langle -| - im'\overline{\langle n'_-|}\langle +|)(a+a^\dagger)(\overline{|n_+\rangle}|+\rangle + im\overline{|n_-\rangle}|-\rangle), \tag{A49}$$

$$= -e^{i(n-n')\omega t}e^{i\frac{1}{2}(m-m')\omega_0' t}\frac{i^2}{2}(m\overline{\langle n'_+|}(a+a^\dagger)\overline{|n_-\rangle} + m'\overline{\langle n'_-|}(a+a^\dagger)\overline{|n_+\rangle}). \tag{A50}$$

Thus, we find

$$\langle m'|\sigma_z(a+a^\dagger)|m\rangle = e^{i\frac{1}{2}(m-m')\omega_0' t}\lambda^{\frac{1}{2}}J_0\left(\frac{\gamma B_1}{\omega}\right)\cos(\omega t)(m+m'). \tag{A51}$$

Furthermore, we have

$$\langle m'|\sigma_z|m\rangle = \frac{1}{2}(m+m')J_0\left(\frac{\gamma B_1}{\omega}\right)e^{i\frac{1}{2}(m-m')\omega_0' t} - i(m-m')e^{i\frac{1}{2}(m-m')\omega_0' t}J_1\left(\frac{\gamma B_1}{\omega}\right)\sin(\omega t). \tag{A52}$$

1. Evaluation of the perturbed expectation of σ_+

Evaluation of the perturbed expectation of σ_+ generates terms to fourth order. We only include terms to second order, first order terms average to zero (which is not precisely true for ultracold neutrons), and terms that include $H_{ii}H_{ik}$ oscillate fast compared to other terms, and will be ignored. The only matrix elements that are found to contribute are

$$\langle\sigma_+\rangle = 1 - \int_0^t\int_0^{t'} H_{-+}H_{+-}dt''dt' - \int_0^t\int_0^{t'} H^*_{+-}H^*_{-+}dt''dt' + \int_0^t\int_0^t H^*_{++}H_{--}dt'dt'' \tag{A53}$$

$$- \int_0^t\int_0^{t'} H^*_{++}H^*_{++}dt''dt' - \int_0^t\int_0^{t'} H_{--}H_{--}dt''dt', \tag{A54}$$

because the functions in H_{jk} are ultimately functions of the trajectories of stationary ensembles is valid except for the phases; however, nonstationary phases oscillate fast and will vanish. Notice that the third integral on the right-hand side has a range $[0, t]$ for both t' and t''. This term arises from the first-order part of the wave function. We can use the symmetry of the stationary trajectory correlation function to express this term in the common form,

$$\int_0^t\int_0^t H^*_{++}H_{--}dt'dt'' = -2\mathrm{Re}\left(\int_0^t\int_0^{t'} H_{--}H_{--}dt''dt'\right). \tag{A55}$$

We substitute this into the equation for $\langle\sigma_+\rangle$ and continue,

$$\langle\sigma_+\rangle = 1 - \int_0^t\int_0^{t'} H_{-+}H_{+-}dt''dt' - \int_0^t\int_0^{t'} H^*_{+-}H^*_{-+}dt''dt' \tag{A56}$$

$$- 2\mathrm{Re}\left(\int_0^t\int_0^{t'} H_{--}H_{--}dt''dt'\right) - \int_0^t\int_0^{t'} H^*_{++}H^*_{++}dt''dt' - \int_0^t\int_0^{t'} H_{--}H_{--}dt''dt', \tag{A57}$$

$$\langle\sigma_+\rangle = 1 - 2\int_0^t\int_0^{t'} H_{-+}(t')H_{+-}(t'')dt''dt' - 4\mathrm{Re}\left(\int_0^t\int_0^{t'} H_{--}H_{--}dt''dt'\right), \tag{A58}$$

$$\langle\sigma_+\rangle = 1 - 2t\int_0^t H_{-+}(0)H_{+-}(\tau)d\tau - 4t\mathrm{Re}\left(\int_0^t H_{--}(0)H_{--}(\tau)d\tau\right). \tag{A59}$$

In the last step we used the fact that the functions are stationary and replaced $\tau = t'' - t'$. In the above equation terms that contain an oscillating phase that is fast compared to the scale of the measurement, for example, it may contain $e^{i\omega_0(t'+t'')}$, are considered negligible. Interestingly, this oscillating phase ensures that the terms that contribute are indeed stationary. Furthermore, from the derivation of the matrix elements found in the next section of the Sec. 2 of the Appendix, we find that the complex conjugate is the equivalent of changing the order of the quantum index, thus, $H^*_{+-} = H_{-+}$.

2. Evaluation of the second-order Matrix Elements

Now we concentrate on the $\omega_{1j}(t')\omega_{1k}(t'')$ terms,

$$[H_{+-}H_{-+}]_{\omega_{1j}\omega_{1k}} = \int_0^t\int_0^{t'} \frac{1}{4}\omega_{1j}(t')\omega_{1k}(t'')e^{i\omega_0't'}\cos(\omega t')e^{-i\omega_0't''}\cos(\omega t'')dt''dt'. \tag{A60}$$

The first term can be separated into harmonics in t'' and t', for which we have

$$[H_{+-}H_{-+}]_{\omega_{1j}\omega_{1k}} = \frac{1}{4}\iint dt'dt''\omega_{1j}(t')\omega_{1k}(t'')e^{i\omega_0't'}\cos(\omega t')e^{-i\omega_0't''}\cos(\omega t''), \tag{A61}$$

$$[H_{+-}H_{-+}]_{\omega_{1j}\omega_{1k}} = \frac{1}{16}\int dt'\int dt''\omega_{1j}(t')\omega_{1k}(t'')(e^{i\omega_0't'+i\omega t'} + e^{i\omega_0't'-i\omega t'})(e^{-i\omega_0't''+i\omega t''} + e^{-i\omega_0't''-i\omega t''}),$$

$$[H_{+-}H_{-+}]_{\omega_{1j}\omega_{1k}} = \frac{1}{16}\int dt'\int dt''\omega_{1j}(t')\omega_{1k}(t'')(e^{i(\omega_0'+\omega)t'-i(\omega_0'-\omega)t''} + e^{i(\omega_0'-\omega)t'-i(\omega_0'+\omega)t''} + e^{i(\omega_0'+\omega)(t'-t'')} + e^{i(\omega_0'-\omega)(t'-t'')}), \tag{A62}$$

$$[H_{+-}H_{-+}]_{\omega_{1j}\omega_{1k}} = \frac{1}{16}\int dt'\int dt''\omega_{1j}(t')\omega_{1k}(t'')(e^{i\omega_0'(t'-t'')+i\omega(t'+t'')} + e^{i\omega_0'(t'-t'')-i\omega(t'+t'')} + e^{i(\omega_0'+\omega)(t'-t'')} + e^{i(\omega_0'-\omega)(t'-t'')}). \tag{A63}$$

Any exponential argument containing $\omega(t'+t'')$ will oscillate much faster than ones with $\omega(t'-t'')$ and can be ignored. Furthermore, we will substitute $t''-t'=\tau$,

$$[H_{+-}H_{-+}]_{\omega_{1j}\omega_{1k}} = \frac{1}{16}\int dt'\int dt''\omega_{1j}(t')\omega_{1k}(t'')[e^{i(\omega_0'+\omega)(t'-t'')} + e^{i(\omega_0'-\omega)(t'-t'')}], \tag{A64}$$

$$[H_{+-}H_{-+}]_{\omega_{1j}\omega_{1k}} = \frac{\gamma^2}{8}G_x^2 t\int \omega_{1j}(0)\omega_{1k}(\tau)[e^{-i\omega_0'\tau}\cos(\omega\tau)]d\tau. \tag{A65}$$

More generally, we can write

$$[H_{+-}H_{-+}]_{\omega_{1j}\omega_{1k}} = \frac{\gamma^2}{8}t\int d\tau\omega_{1j}(0)\omega_{1k}(\tau)[e^{-i\omega_0'\tau}\cos(\omega\tau)]. \tag{A66}$$

Here we show that the cross terms between ω_0 and ω_1 for the off-diagonal elements can be neglected.

$$[H_{-+}H_{+-}]_{\omega_1'\omega_0'} = \frac{1}{4}\iint \omega_{1x}'(t'')\omega_{0x}'(t')\cos(\omega t'')e^{i\omega_0'(t''-t')}dt'dt'' \tag{A67}$$

$$+\frac{1}{4}\iint \omega_{1x}'(t')\omega_{0x}'(t'')\cos(\omega t')e^{i\omega_0'(t''-t')}dt'dt'', \tag{A68}$$

$$[H_{-+}H_{+-}]_{\omega_1'\omega_0'} = \frac{1}{8}\iint \omega_{1x}'(t'')\omega_{0x}'(t')(e^{i\omega t''}+e^{-i\omega t''})e^{i\omega_0'(t''-t')}dt'dt'' \tag{A69}$$

$$+\frac{1}{8}\iint \omega_{1x}'(t')\omega_{0x}'(t'')(e^{i\omega t''}+e^{-i\omega t''})e^{i\omega_0'(t''-t')}dt'dt''. \tag{A70}$$

We set $\tau = t''-t'$,

$$[H_{-+}H_{+-}]_{\omega_1'\omega_0'} = \frac{1}{8}\iint \omega_{1x}'(\tau+t')\omega_{0x}'(t')(e^{i\omega(\tau+t')}+e^{-i\omega(\tau+t')})e^{i\omega_0'\tau}dt'dt'' \tag{A71}$$

$$+\frac{1}{8}\iint \omega_{1x}'(t')\omega_{0x}'(\tau+t')(e^{i\omega t'}+e^{-i\omega t'})e^{i\omega\tau}dt'dt'', \tag{A72}$$

$$[H_{-+}H_{+-}]_{\omega_1'\omega_0'} = \frac{1}{8}\iint \omega_{1x}'(\tau+t')\omega_{0x}'(t')(e^{i(\omega+\omega_0')(\tau+t')}+e^{i\omega(-\omega+\omega_0')(\tau+t')})dt'dt'' \tag{A73}$$

$$+\frac{1}{8}\iint \omega_{1x}'(t')\omega_{0x}'(\tau+t')(e^{i\omega t'}+e^{-i\omega t'})e^{i\omega\tau}dt'dt''. \tag{A74}$$

We see that all terms contain oscillating phases that allow the result to be neglected,

$$[H_{-+}H_{+-}]_{\omega_1'\omega_0'} \approx 0. \tag{A75}$$

Here we examine the cross frequency terms of the diagonal elements (σ_z), and show they can be neglected,

$$[H_{--}H_{--}]_{\omega_1'\omega_0'} = \frac{1}{4}\iint \omega_{1z}'(t'')\omega_{0z}'(t')\cos(\omega t'')dt'dt'' + \frac{1}{4}\iint \omega_{1z}'(t')\omega_{0z}'(t'')\cos(\omega t')dt'dt'', \tag{A76}$$

substituting $\tau = t''-t'$,

$$[H_{--}H_{--}]_{\omega_1'\omega_0'} = \frac{1}{4}\iint \omega_{1z}'(\tau+t')\omega_{0z}'(t')(e^{i\omega(\tau+t')}+e^{-i\omega(\tau+t')})dt'dt'' \tag{A77}$$

$$+\frac{1}{4}\iint \omega_{1z}'(t')\omega_{0z}'(t'')(e^{i\omega t'}+e^{-i\omega t'})dt'dt''. \tag{A78}$$

Again, all terms contain a rapidly oscillating phase, so we have

$$[H_{--}H_{--}]_{\omega_1'\omega_0'} \approx 0. \tag{A79}$$

The evaluation of the matrix elements for $\omega_0'\omega_0'$ terms are not included here. However, identical derivations are found in Refs. [6,11,12] with the exception of the factor $J_0(x)^2$. Now we show evaluation of the matrix elements that contribute from the terms where $q = \pm 1$. Several of these terms cancel due to fast oscillating factors, or by symmetry, here we consider terms that contribute in principle,

$$[H_{-+}H_{+-}]_{\omega'_{1x}\omega'_{0zq}} \tag{A80}$$

$$= \iint \frac{i}{2} e^{i\omega'_0(t-t')}[\sin(\omega t')\cos(\omega t)\omega_{1x}(t)\omega_{0qz}(t') - \sin(\omega t)\cos(\omega t')\omega_{1x}(t')\omega_{0qz}(t)]dtdt', \tag{A81}$$

$$= \iint \frac{i}{2} e^{i\omega'_0(t-t')}\omega_{1x}(t')\omega_{0qz}(t)\frac{i}{4}[(e^{i\omega t'} - e^{-i\omega t'})(e^{i\omega t} + e^{-i\omega t}) - (e^{i\omega t} - e^{-i\omega t})(e^{i\omega t'} + e^{-i\omega t'})]dtdt', \tag{A82}$$

$$= \iint \frac{i}{2} e^{i\omega'_0(t-t')}\frac{i}{4}\omega_{1x}(t')\omega_{0qz}(t)[(e^{i\omega t'-i\omega t} - e^{i\omega t-i\omega t'}) - (e^{i\omega t-i\omega t'} - e^{i\omega t'-i\omega t})]dtdt', \tag{A83}$$

$$= -t\int \frac{i}{2} e^{-i\omega'_0\tau}\sin(\omega\tau)\omega_{1x}(0)\omega_{0zq}(\tau)d\tau.[4pt] \tag{A84}$$

We point out that this is a phase- and frequency-shifted sine transform. This translates into the real part of the difference in the spectrum shifted by $\omega \pm \omega'_0$ and when $\omega \gg \omega'_0$ these terms are negligible. A similar derivation is completed for the squared terms of the off-diagonal components,

$$[H_{-+}H_{+-}]_{\omega'_{0zq}\omega'_{0zq}} \tag{A85}$$

$$= \iint e^{i\omega'_0(t-t')}[\sin(\omega t')\sin(\omega t)\omega_{0zq}(t)\omega_{0zq}(t')(t)]dtdt' \tag{A86}$$

$$= -\iint e^{i\omega'_0(t-t')}\left[\frac{1}{4}(e^{i\omega t'-i\omega t} - e^{i\omega t-i\omega t'})(e^{i\omega t} - e^{-i\omega t})\right]dtdt' \tag{A87}$$

$$= -\iint e^{i\omega'_0(t-t')}[(e^{i\omega t'-i\omega t} - e^{i\omega t-i\omega t'})]dtdt' \tag{A88}$$

$$= -\frac{it}{2}\int e^{-i\omega'_0\tau}\sin(\omega\tau)\omega_{0zq}(0)\omega_{0zq}(\tau)d\tau. \tag{A89}$$

Again, we point out that this will be highly suppressed as it goes with the difference in the spectrum at $\omega \pm \omega'_0$. Which is negligible when $\omega \gg \omega'_0$.

The last term that contributes, in principle, is the diagonal components for $q = \pm 1$. The only nonvanishing term is the squared term from σ_y. It can be shown to be

$$[H_{--}H_{--}]_{\omega'_{0yq}\omega'_{0yq}} = -\frac{it}{2}\int \sin(\omega\tau)\omega_{0yq}(0)\omega_{0yq}(\tau)d\tau. \tag{A90}$$

From symmetry, this term is zero when we take the real part to find the relaxation.

3. Simplification of the DC field diagonal contribution to T_2

Starting from Eq. (67), we continue by expanding the exponential in a series and taking the leading-order terms,

$$\langle\sigma_+\rangle = e^{i\gamma\int_0^{t'}[\frac{1}{\omega}J_1(\frac{\gamma\langle B_1\rangle}{\omega})B_0\omega_{1x}(t')+1J_0(\frac{\gamma\langle B_1\rangle}{\omega})\delta B_0(t')]d}, \tag{A91}$$

$$\langle\sigma_+\rangle \approx 1 - i\gamma\int_0^{t'}\left[\frac{\gamma}{\omega}J_1\left(\frac{\gamma\langle B_1\rangle}{\omega}\right)B_0\omega_{1x}(t') + \gamma J_0\left(\frac{\gamma\langle B_1\rangle}{\omega}\right)\delta B_0(t)\right]dt', \tag{A92}$$

$$-\frac{\gamma^2}{2}\int_0^{t'}\left[\frac{\gamma}{\omega}J_1\left(\frac{\gamma\langle B_1\rangle}{\omega}\right)B_0\omega_{1x}(t') + \gamma J_0\left(\frac{\gamma\langle B_1\rangle}{\omega}\right)B'_0(t')\right]dt' \times \dots \tag{A93}$$

$$\int_0^{t'}\left[\frac{\gamma}{\omega}J_1\left(\frac{\gamma\langle B_1\rangle}{\omega}\right)B_0\omega_{1x}(t') + \gamma J_0\left(\frac{\gamma\langle B_1\rangle}{\omega}\right)B'_0(t')\right]dt''. \tag{A94}$$

Now we write $x = \frac{\gamma\langle B_1\rangle}{\omega}$, to signify that there is no more time dependence within the Bessel functions. From the definition of $\delta B_0(t)$ we see that the first term averages to zero, for the last term on the right-hand side we find

$$\langle\sigma_+\rangle \approx 1 - \frac{\gamma^2}{2}J_0(x)^2\int^t \delta B_0(t')dt'\int^{t'}\delta B_0(t'')dt'', \quad \text{(A95)}$$

$$-\frac{J_1(x)^2\omega_0^2}{2\omega^2}\int_0^t \omega_{1x}(t')dt'\int_0^{t'}\omega_{1x}(t'')dt'', \quad \text{(A96)}$$

$$+\frac{\gamma^2 J_0(x)J_1(x)B_0}{\omega}\int^t \omega_{1x}(t')dt'\int^{t'}\delta B_0(t'')dt''. \quad \text{(A97)}$$

We recognize that all functions are mechanically stationary and proceed to write them in terms of the correlation functions of the variable $\tau = t'' - t'$,

$$\langle\sigma_+\rangle \approx 1 - \frac{\gamma^2}{2}J_0(x)^2(t-|\tau|)\int_{-t}^t B_0'(0)B_0'(\tau)d\tau, \quad \text{(A98)}$$

$$-\frac{J_1(x)^2\omega_0^2}{2\omega^2}(t-|\tau|)\int_{-t}^t \omega_1'(0)\omega_1'(\tau)d\tau, \quad \text{(A99)}$$

$$+\frac{\gamma J_0(x)J_1(x)B_0}{\omega}(t-|\tau|)\int_{-t}^t \omega_1'(0)B_0'(\tau)d\tau. \quad \text{(A100)}$$

Now we take the limit that τ_c is much smaller than t, but at time t the decay is small, allowing us to write the results in the usual form of a Transform. From this simplification, we can write

$$\langle\sigma_+\rangle \approx 1 - \gamma^2 J_0(x)^2 t\int_0^\infty B_0'(0)B_0'(\tau)d\tau \quad \text{(A101)}$$

$$-\frac{J_1(x)^2\omega_0^2}{\omega^2}t\int_0^\infty \omega_1'(0)\omega_1'(\tau)d\tau \quad \text{(A102)}$$

$$+2\frac{\gamma^2 J_0(x)J_1(x)B_0}{\omega}t\int_0^\infty \omega_1'(0)B_0'(\tau)d\tau. \quad \text{(A103)}$$

This result is used to derive Eq. (70).

4. Modulation alternative

The pulsed modulation is optimized to maximize sensitivity while correcting for linear drifts below about $f_{\mathrm{fm}} = 1$ Hz. However, noise, either external noise or noise generated by field inhomogeneities, can be further reduced by cosine modulation in conjunction with a sharp band pass around f_{fm} to "lock-in" to the signal. It is expected that in general the ultimate sensitivity will be larger for pulsed frequency modulation. This is due to the strict requirement on the inhomogeneities of the dressing field from the requirement of transverse relaxation time, simultaneously ensuring minimal noise from spin-dressing inhomogeneities.

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