## Frequency mixing spectroscopy of spins in diamond

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Frequency mixing processes in spin systems have a variety of applications in meteorology and in quantum data processing. Spin spectroscopy based on frequency mixing offers some advantages, including the ability to eliminate crosstalk between driving and detection. We experimentally explore nonlinear frequency mixing processes with negatively charged nitrogen-vacancy defects in diamond at low temperatures, and near level anti crossing. The experimental setup allows simultaneously applying magnetic driving in the longitudinal and transverse directions. Magnetic resonance detection is demonstrated using both Landau Zener Stückelberg interferometry and two-tone driving spectroscopy. The experimental results are compared with predictions of a theoretical analysis based on the rotating wave approximation.

Introduction - Spins are essentially the most nonlinear systems found in nature. Their magnetic resonance is widely employed for sensing and imaging applications. In some cases, nonlinear response impacts the sensitivity and bandwidth of magnetic resonance imaging systems [1, 2]. Nonlinear response can be exploited for performance enhancement in some cases. One example is enhancement of magnetic resonance detection sensitivity that is achieved using squeezed microwave fields [3]. Another example is driving-induced enhancement of coupling between spins [4]. Moreover, for quantum data processing, nonlinear response is an essential resource enabling the generation of entangled states [5] and topological frequency conversion [6]. In addition, nonlinear response of driven spins can be exploited for the generation of exotic states of matter by means of Floquet engineering [7].

Frequency mixing processes that are enabled by nonlinear response can be employed for a variety of applications. In spectroscopy, sensitivity is commonly limited by crosstalk, when both driving and detection are performed at the same frequency (which is usually the case in cavity-based spectroscopy [2]). This problem can be avoided by employing frequency mixing, which enables resonance driving using only off-resonance driving tones. Other examples for frequency mixing applications are electromagnetically induced transparency [8] and hole burning [9]. Frequency mixing can be used to manipulate transition rates between quantum states, which, in turn, can enable controlling states' population. Cooling induced by frequency mixing has been demonstrated in [10] using a Josephson flux qubit. A similar process of frequency mixing can be implemented to achieve population inversion.

Here we explore frequency mixing processes in negatively charged nitrogen-vacancy (NV<sup>-</sup>) defects in diamond at low temperatures. A NV<sup>-</sup> defect has a spin triplet ground state with magnetic quantum numbers  $m_{\rm S} \in \{-1, 0, 1\}$  [11]. The NV<sup>-</sup> electronic spin state can be polarized and read out with light in the optical band. Dense ensembles of NV<sup>-</sup> defects were demonstrated to be applicable for magnetometry [12], classical [13] and quantum [14–17] information storage, and for a maser implementation [18]. The NV magnetometry sensitivity may reach sub picotesla per  $Hz^{1/2}$  level [19–21]. Dual driving of NV<sup>-</sup> electrons and nuclear spins were investigated by optical detection of magnetic resonance (ODMR) [21– 27], Landau Zener Stückelberg interferometry [28, 29], and electron-spin double resonance [1, 30].

In this work we explore mixing between longitudinal driving in the radio frequency (RF) band and transverse driving in the microwave (MW) band. To account for our experimental results, theoretical predictions derived using the rotating wave approximation (RWA) are presented. We find relatively strong nonlinear response near the NV<sup>-</sup> ground state level anti crossing (GSLAC) [9, 25, 26, 31, 32]. The response enhancement is attributed to a significant state mixing occurring near GSLAC. The same state mixing gives rise to a significant modification in transition selection rules. In particular, the commonly forbidden transition between  $m_{\rm S} = -1$ and  $m_{\rm S} = +1$  states, becomes partially allowed near GSLAC. This transition can be used to double spin detection sensitivity (compared to sensitivity obtained using transitions associated with a change in the magnetic quantum number  $m_{\rm S}$  equals +1 or -1). However, away from GSLAC, detection based on this transition requires double-quantum driving [27].

**Experimental setup** - A diamond sample with nitrogen concentration lower than 200 ppm was electron irradiated at energy and dose of 2.8 MeV and  $8 \times 10^{18}$  cm<sup>-2</sup>, respectively, and annealed at 900°C for 2 hours to create NV<sup>-</sup> defects. The NV<sup>-</sup> concentration is  $3 \times 10^{17}$  cm<sup>-3</sup>. Concentration was estimated by comparing with a reference sample having a known NV concentration (with sufficiently low laser power, for which fluorescence is proportional to NV concentration) [33]. The sample assembly was placed at a cryostat with a base temperature of 3.6 K. To reach the GSLAC region, the diamond [111] axis direction was placed nearly parallel to the externally applied static magnetic field, which was generated by superconducting coils.

Two antennas are employed for spin driving. An RF solenoid, having axis nearly parallel to the applied static magnetic field, allows longitudinal driving, whereas transverse driving is applied using a microwave loop an-



FIG. 1: ODMR in the GSLAC region. The low and high frequency resonances are shown in (a) and (b), respectively. The black solid lines in (a2) and (b2) are based on numerical diagonalization of the triplet Hamiltonian (1).

tenna (LA) having axis nearly orthogonal to the applied static magnetic field. The impedance mismatching coefficient  $\zeta$ , which is given by  $\zeta = \omega L/Z_0$  [34], where  $\omega$  is the driving angular frequency, L is the antenna's inductance, and  $Z_0 = 50 \Omega$  is the impedance of the coaxial cable attached to the antenna, is  $\zeta = 94$  for the RF solenoid at  $\omega/2\pi = 10$  MHz, and  $\zeta = 6.0$  for the MW LA at  $\omega/2\pi = 3$  GHz. An impedance matching capacitor, serially connected to the RF solenoid, is employed for the measurements shown in Fig. 4.

The experimental setup used for performing ODMR measurements is described in Ref. [35]. ODMR signal recorded near the GSLAC region is shown in Fig. 1 as a function of the driving frequency  $\omega$  and magnetic field B.

Triplet ground state - When hyperfine interaction is disregarded the ground state spin triplet Hamiltonian  $\mathcal{H}$  becomes [36, 37]

$$\frac{\mathcal{H}}{\hbar} = \frac{\omega_{\rm D} S_z^2}{\hbar^2} + \frac{\omega_{\rm E} \left(S_+^2 + S_-^2\right)}{2\hbar^2} - \frac{\gamma_{\rm e} \mathbf{B} \cdot \mathbf{S}}{\hbar} , \qquad (1)$$

where  $\omega_{\rm D} = 2\pi \times 2.87 \,\mathrm{GHz}$  is the zero field splitting,  $\omega_{\rm E} \ll \omega_{\rm D}$  is a strain-induced splitting,  $\gamma_{\rm e} = 2\pi \times 28.03 \,\mathrm{GHz} \,\mathrm{T}^{-1}$  is the electron spin gyromagnetic ratio,  $\mathbf{S} = S_x \hat{\mathbf{x}} + S_y \hat{\mathbf{y}} + S_z \hat{\mathbf{z}}$  is the spin S = 1 angular momentum vector operator and  $S_{\pm} = S_x \pm i S_y$ . The applied magnetic field is expressed as  $\mathbf{B} = \gamma_{\rm e}^{-1} \boldsymbol{\omega}$ , where the angular frequency vector  $\boldsymbol{\omega} = (\omega_x, \omega_y, \omega_z) = \boldsymbol{\omega}_{\rm dc} + \boldsymbol{\omega}_{\rm ac}$  is decomposed into a static part  $\boldsymbol{\omega}_{\rm dc} = (\omega_{\rm dc,x}, \omega_{\rm dc,y}, \omega_{\rm dc,z})$  (see Fig. 2) and a time varying part  $\boldsymbol{\omega}_{\rm ac} = (\omega_{\rm ac,x}, \omega_{\rm ac,y}, \omega_{\rm ac,z})$ (having a vanishing averaged value). GSLAC occurs when  $\boldsymbol{\omega} \simeq (0, 0, \omega_{\rm D})$ . The black solid lines in Fig. 1 (a2) and (b2) are derived from a numerical diagonalization of the triplet Hamiltonian (1).

**RWA** - The system's respond to both longitudinal and transverse external driving is estimated below using the



FIG. 2: NV. The NV axis is parallel to the z axis, and the static field  $\omega_{dc}$ , which is nearly parallel to the z axis, lies in the xz plane.

RWA. The  $3 \times 3$  matrix representation  $H = H_{\rm dc} + H_{\rm ac}$  of the Hamiltonian operator  $\mathcal{H}$  is decomposed into a static  $H_{\rm dc}$  and time varying  $H_{\rm ac}$  parts. Two unitary transformations are successively applied to H. The first one is performed with a time independent unitary matrix U, which diagonalizes the static part, i.e. the  $3 \times 3$  matrix  $U^{\dagger}H_{\rm dc}U \equiv H'_{\rm dc}$  is diagonal. The matrix elements of  $\hbar^{-1}H' = \hbar^{-1}U^{-1}HU$  are denoted by  $\omega'_{n',n''}$ , where  $n', n'' \in \{1, 2, 3\}$ . The first transformation is explained below in the next section (titled 'diagonalization') and in appendix A.

The second transformation, which is discussed in this section, and which is applied to H', may help simplifying the evaluation of the steady state response of the driven spin system under study. In general, nonlinear response may give rise to a highly complex steady state. However, in some cases the response is dominated by a single frequency mixing process. For such cases the RWA can be employed to derive an analytical approximation for the system's steady state response.

The second transformation is given by  $H'' = -i\hbar u^{\dagger} (du/dt) + u^{\dagger} H' u$ , where the elements  $u_{n',n''}$  of the diagonal unitary matrix u are given by  $u_{n',n''} = \delta_{n',n''} \exp\left(-i\int^t dt' \,\omega'_{n',n'}(t')\right)$ . The matrix elements  $\omega''_{n',n''}$  of  $\hbar^{-1}H''$  are given by

$$\omega_{n',n''}'' = (1 - \delta_{n',n''}) \,\omega_{n',n''}' e^{-i\int^t \mathrm{d}t' \, \left(\omega_{n'',n''}'(t') - \omega_{n',n'}'(t')\right)} \tag{2}$$

Note that the matrix  $\hbar^{-1}H''$  is hollow, i.e. all its diagonal elements  $\omega''_{n',n'}$  vanish [see Eq. (2)].

Consider the case where for some  $n' \neq n''$  the term  $\omega'_{n'',n''} - \omega'_{n',n'}$  in Eq. (2) can be expressed as  $\omega'_{n'',n''} - \omega'_{n',n'} = \Omega_0 - \Omega_{\rm L1} \cos{(\Omega_{\rm L}t')}$ , and the term  $\omega'_{n',n''}$  as

 $\omega'_{n',n''} = \Omega_{\text{T1}} e^{i\Omega_{\text{T}}t}$ , where  $\Omega_0$ ,  $\Omega_{\text{L1}}$ ,  $\Omega_{\text{L}}$ ,  $\Omega_{\text{T1}}$  and  $\Omega_{\text{T}}$  are all real constants. With the help of the Jacobi-Anger expansion one finds that for this case [see Eq. (2)]

$$\omega_{n',n''}^{\prime\prime} = \sum_{l=-\infty}^{\infty} \frac{\Omega_{1,l}}{2} e^{-i\Omega_{\mathrm{d},l}t} , \qquad (3)$$

where  $\Omega_{1,l} = 2\Omega_{T1}J_l(\Omega_{L1}/\Omega_L)$ ,  $J_l$  denotes the Bessel function of the first kind, and  $\Omega_{d,l}$ , which is given by  $\Omega_{d,l} = \Omega_0 - \Omega_T - l\Omega_L$ , represents the angular frequency detuning corresponding to the *l*'th frequency mixing resonance, occurring when  $\Omega_T + l\Omega_L = \Omega_0$ .

Rapidly oscillating terms are disregarded in the RWA. Consider the case where the matrix elements  $\omega_{n',n''}^{\prime\prime}$  of  $\hbar^{-1}H^{\prime\prime}$  are Fourier expanded as sums of terms having the form  $(\Omega_1/2) e^{-i\Omega_d t}$  [see Eq. (3)], where the driving amplitudes  $\Omega_1$ , and the frequency detunings  $\Omega_d$ , are real constants. The effect of each such a term is strong when the amplitude  $|\Omega_1|$  is large, and the detuning  $|\Omega_d|$ is small. The analysis is greatly simplified when, among all terms  $\omega_{n',n''}^{\prime\prime}$  in the upper diagonal of  $\hbar^{-1}H^{\prime\prime}$  (i.e. n' < n''), a single term having the form  $(\Omega_1/2) e^{-i\Omega_{\rm d}t}$ in a single matrix element  $\omega_{n_1,n_2}''$ , where  $n_1 < n_2$ , dominates (recall that the diagonal elements of H'' vanish). For this case, in the RWA all other terms are disregarded, i.e. it is assumed that the only non-vanishing matrix elements  $\omega_{n',n''}^{"}$  of  $\hbar^{-1}H''$  are  $\omega_{n_1,n_2}^{"} = (\Omega_1/2) e^{-i\Omega_d t}$  and  $\omega_{n_2,n_1}^{\prime\prime} = (\Omega_1/2) e^{i\Omega_{\rm d}t}$ . The effect of driving can be characterized by the coefficient  $\mathcal{P}_{n_1,n_2}$ , which is given by [see Eq. (5) of Ref. [38], and Eq. (D11) of Ref. [39]]

$$\mathcal{P}_{n_1,n_2} = \frac{\frac{\Omega_1^2}{\gamma_1 \gamma_2}}{1 + \frac{\Omega_d^2}{\gamma_2^2} + \frac{\Omega_1^2}{\gamma_1 \gamma_2}}, \qquad (4)$$

where  $\gamma_1$  ( $\gamma_2$ ) is a longitudinal (transverse) spin relaxation rate. Note that for a two-level system  $\mathcal{P} = 1 - P_0/P_s$ , where  $P_0$  ( $P_s$ ) is the population polarization in steady state with (without) driving [see [40], and Eq. (17.265) of Ref. [41]]. The coefficient  $\mathcal{P}_{n_1,n_2}$  can be used to approximately quantify the effect of driving on steady state ODMR signal.

**Diagonalization** - Prior to the above-explained transformation that treats the driving terms using the RWA, a time-independent transformation is applied in order to diagonalize the static part of the Hamiltonian  $H_{dc}$ . The diagonalized static part of the Hamiltonian reveals the transition frequencies, whereas the unitary matrix that diagonalizes  $H_{dc}$  reveals the transition selection rules.

The matrix H is given by [the term  $\omega_{\rm zd}/2$  has been added to the diagonal, see Eq. (1)]

$$\frac{H}{\hbar} = \begin{pmatrix} -\frac{\omega_{zd}}{2} & \frac{\omega_{\delta}}{2} & \omega_{\rm E} \\ \frac{\omega_{\delta}^*}{2} & \frac{\omega_{zd}}{2} & \frac{\omega_{\delta}}{2} \\ \omega_{\rm E} & \frac{\omega_{\delta}}{2} & \omega_{\rm h} \end{pmatrix} , \qquad (5)$$

where  $\omega_{\rm zd} = \omega_z - \omega_{\rm D}$  represents detuning from GSLAC,  $\omega_{\delta} = -\sqrt{2} (\omega_x - i\omega_y)$  represents transverse magnetic fields, and  $\omega_{\rm h} = (\omega_{\rm D} + 3\omega_{\rm zd})/2$ . The static part  $H_{dc}$  of H [see Eq. (5)] can be expressed as (it is assumed that  $\omega_{dc,y} = 0$ )

$$\frac{H_{\rm dc}}{\hbar} = \frac{\omega_{\rm R0}}{2} \begin{pmatrix} -\cos\theta & -\sin\theta & \frac{2\omega_{\rm E}}{\omega_{\rm R0}} \\ -\sin\theta & \cos\theta & -\frac{\omega_{\rm H}}{\omega_{\rm R0}} \\ \frac{2\omega_{\rm E}}{\omega_{\rm R0}} & -\frac{\omega_{\rm H}}{\omega_{\rm R0}} & \frac{2\omega_{\rm dc,h}}{\omega_{\rm R0}} \end{pmatrix} , \qquad (6)$$

where  $\omega_{\rm R0} = \sqrt{(\omega_{\rm dc,z} - \omega_{\rm D})^2 + \omega_{\rm H}^2}$ ,  $\omega_{\rm H} = \sqrt{2}\omega_{\rm dc,x}$ ,  $\omega_{\rm dc,h} = (\omega_{\rm D} + 3\omega_{\rm dc,z})/2$ , and the angle  $\theta$  is given by

$$\cot \theta = \frac{\omega_{\mathrm{dc},z} - \omega_{\mathrm{D}}}{\omega_{\mathrm{H}}} \equiv \eta .$$
 (7)

Note that GSLAC corresponds to the case where  $\omega_{dc,z} = \omega_D$ ,  $\eta = 0$  and  $|\theta| = \pi/2$ , whereas  $|\eta| \to \infty$  and  $\theta \to 0$  far from GSLAC. The GSLAC transition angular frequency is approximately given by  $\omega_{R0} = \omega_H \sqrt{1 + \eta^2}$ .

In the vicinity of the GSLAC, the static part  $H_{\rm dc}$  of H is approximately diagonalized using the transformation  $H'_{\rm dc} = U^{-1}H_{\rm dc}U$ , where the unitary matrix U represents a rotation about the z axis through an angle of  $\theta/2$ . Applying the same transformation U to the time dependent part  $H_{\rm ac}$  yields two terms  $H'_{\rm ac} = U^{-1}H_{\rm ac}U = H'_{\rm acT} + H'_{\rm acL}$ . The first one  $H'_{\rm acT}$ , which is given by (see appendix A)

$$\frac{H_{\text{acT}}'}{\hbar} = \begin{pmatrix} -\omega_{\text{TL}} & \omega_{\text{TT}} & -\frac{\omega_{\text{T}} \sin \frac{\theta}{2}}{\sqrt{2}} \\ \omega_{\text{TT}}^* & \omega_{\text{TL}} & -\frac{\omega_{\text{T}} \cos \frac{\theta}{2}}{\sqrt{2}} \\ -\frac{\omega_{\text{T}} \sin \frac{\theta}{2}}{\sqrt{2}} & -\frac{\omega_{\text{T}} \cos \frac{\theta}{2}}{\sqrt{2}} & 0 \end{pmatrix} , \quad (8)$$

where  $\omega_{\text{TL}} = 2^{-3/2} (\omega_{\text{T}} + \omega_{\text{T}}^*) \sin \theta$ ,  $\omega_{\text{TT}} = 2^{-1/2} (\omega_{\text{T}} \sin^2(\theta/2) - \omega_{\text{T}}^* \cos^2(\theta/2))$ , and where  $\omega_{\text{T}} = \omega_{\text{ac},x} + i\omega_{\text{ac},y}$ , originates from transverse driving, whereas longitudinal driving gives rise to the second term  $H'_{\text{acL}}$ , which is given by (see appendix A)

$$\frac{H_{\rm acL}'}{\hbar} = \frac{\omega_{\rm ac,z}}{2} \begin{pmatrix} -\cos\theta & \sin\theta & 0\\ \sin\theta & \cos\theta & 0\\ 0 & 0 & 3 \end{pmatrix} .$$
(9)

Note that  $\cos \theta = \eta / \sqrt{\eta^2 + 1}$ ,  $\sin \theta = 1 / \sqrt{\eta^2 + 1}$  and  $-\omega_{\text{TL}}^2 - \omega_{\text{TT}} \omega_{\text{TT}}^* = -|\omega_{\text{T}}|^2 / 2$  (determinant of the 2 × 2 top upper left block of  $\hbar^{-1} H_{\text{acT}}^{\prime}$ ). Single antenna drive - The rotation angle  $\theta / 2$  as-

Single antenna drive - The rotation angle  $\theta/2$  associated with the diagonalization of the static part  $H_{\rm dc}$  (6) [see Eq. (A2)] becomes relatively large near GSLAC. Consequently, in this region the contribution of the MW LA to the effective value of longitudinal driving amplitude becomes significant [see Eq. (8)]. As is demonstrated below (see Fig. 3), this effective longitudinal driving gives rise to a sequence of superharmonic resonances occurring near GSLAC.

The plot in Fig. 3(a) exhibits ODMR measurements performed with a monochromatic driving applied only to the MW LA. The driving has a fixed frequency  $\omega_{\rm T}/(2\pi) = 145$  MHz and a varying power  $P_{\rm T}$ . The driving amplitude, which is denoted by  $\omega_{\rm T1}$ , is proportional



FIG. 3: Single antenna drive. (a) ODMR signal as a function of static magnetic filed *B* and driving power  $P_{\rm T}$ . (b) Calculated driving-induced polarization coefficient  $\mathcal{P}$  [see Eq. (4)]. The parameters' assumed values for the calculation of  $\mathcal{P}$  are  $\gamma_1 = 0.5$  MHz and  $\gamma_2 = 2$  MHz. (c) Graphical representation of the frequency mixing resonance condition  $\omega_{\rm R0} = \omega_{\rm H} \sqrt{1 + \eta^2} = l\omega_{\rm T}$ . The driving frequency  $\omega_{\rm T}/(2\pi) = 145$  MHz is represented by the horizontal cyan line.

to  $P_{\rm T}^{1/2}$ . In the transformed basis (in which the static part of the Hamiltonian is diagonalized), the effective longitudinal (transverse) driving amplitude is  $\omega_{\rm TL}$  ( $\omega_{\rm TT}$ ) [see Eq. (8)].

The *l*'th frequency mixing resonance occurs when  $\omega_{\rm R0} = \omega_{\rm H}\sqrt{1+\eta^2} = l\omega_{\rm T}$ , where *l* is a positive integer. This resonance condition is graphically displayed in Fig. 3(c). In the experimental data shown in Fig. 3(a), resonances with  $l \leq 10$  can be resolved. The driving-induced polarization coefficient  $\mathcal{P}$ , which is calculated using Eqs. (3) and (4), is shown in Fig. 3(b). Parameters' assumed values are listed in the caption of Fig. 3. The comparison between the measured ODMR signal (a) and calculated polarization coefficient  $\mathcal{P}$  (b) suggests that the decay of resonance intensity with |l| is theoretically overestimated. The discrepancy is mainly attributed to dipolar coupling, which gives rise to additional driving term, which is disregarded in the calculation of  $\mathcal{P}$  [42–45].

The two resonances labelled in Fig. 3(a) by red cross symbols represent second Larmor lines. The resonance condition for the second Larmor lines, which is given by  $2\omega_{\rm R0} = \omega_{\rm T}$ , is graphically shown in Fig. 3(c) (the term  $2\omega_{\rm R0}$  is represented by the red hyperbola, and  $\omega_{\rm T}/(2\pi) = 145$  MHz by the horizontal cyan line). Note that the nonlinear process responsible for the second Larmor lines is not taken into account in the calculation of the driving-induced polarization coefficient  $\mathcal{P}$  [see Fig. 3(b)].

Two antenna drive - The case of simultaneous driving with both RF solenoid and MW LA is exhibited by the plots shown in Figs. 4 and 5, which demonstrate that frequency mixing between off-resonance monochromatic driving tones can be employed for resonance driving. This frequency mixing process is related to the Landau Zener Stückelberg effect, which describes a coherent interference occurring in a two-level system under simultaneously applied two driving tones [46]. Landau Zener Stückelberg interferometry (see Fig. 4 below) commonly requires intense longitudinal driving. For exploring this interferometry, a capacitor having capacitance of  $36 \,\mu\text{F}$  is serially connected to the RF solenoid. The added capacitor helps enhancing the magnetic field driving amplitude by suppressing the undesirable effect of parasitic capacitance of the RF solenoid. The RF solenoid with the added capacitor is operated near its resonance frequency of 10.5 MHz.

For the ODMR signal shown in the plot in Fig. 4(a), the RF solenoid is driven with a varying amplitude  $\Omega_{L1}$ (proportional to the applied voltage amplitude  $V_{RF}$ ) and a fixed frequency of  $\Omega_{L}/(2\pi) = 10.5$  MHz, and the MW LA is driven with a fixed amplitude of  $\Omega_{T1} = 5.6$  MHz and a fixed frequency of  $\Omega_{T}/(2\pi) = 3.15$  GHz. The plot shown in Fig. 4(b) displays the calculated polarization coefficient  $\mathcal{P}$  [see Eq. (4)] in the same region, as a function of  $V_{RF}$  and the current  $I_{mag}$  applied to the solenoid generating the static magnetic field. The assumed parameters' values used for the calculation of  $\mathcal{P}$ , which is base on Eqs. (3) and (4), are listed in the caption of Fig. 4.

While the measurements presented in Fig. 4 have been performed away from the GSLAC, the case of two antenna driving near GSLAC is demonstrated by the plots shown in Fig. 5. The plot in Fig. 5(a) displays ODMR data as a function of static magnetic filed *B* and RF solenoid driving power  $P_{\rm L}$ . For this measurement, the RF solenoid driving frequency is  $\omega_{\rm L}/(2\pi) = 185$  MHz, MW LA driving frequency is  $\omega_{\rm T}/(2\pi) = 5.87$  GHz and MW LA driving power is 25 dBm. The plot shown in Fig. 5(b) displays the calculated polarization coefficient  $\mathcal{P}$  in the same region [see Eqs. (3) and (4)].

The calculated energy eigenvalues of the triplet Hamiltonian (1) are plotted as a function of *B* in Fig. 5(c). To graphically display the frequency mixing matching condition, the two lowest eigen energies [green solid lines in Fig. 5(c)] are vertically shifted upwards by the MW LA driving frequency  $\omega_{\rm T}/(2\pi) = 5.87$  GHz. The highest eigen energy is represented by the solid blue line. The length of the red vertical arrows in Fig. 5(c) is the RF solenoid driving frequency  $\omega_{\rm L}/(2\pi) = 185$  MHz.

Summary - Nonlinear magnetic resonance sensing of



FIG. 4: Two antenna drive. (a) ODMR signal as a function of RF solenoid applied voltage amplitude  $V_{\rm RF}$ , and current applied to the solenoid generating the static magnetic field  $I_{\rm mag}$ . The fixed RF solenoid driving frequency is  $\Omega_{\rm L}/(2\pi) =$ 10.5 MHz, MW LA driving amplitude is  $\Omega_{\rm T1} = 5.6$  MHz, and MW LA driving frequency is  $\Omega_{\rm T}/(2\pi) = 3.15$  GHz. (b) Calculated driving-induced polarization coefficient  $\mathcal{P}$  [see Eq. (4)]. The parameters' assumed values for the calculation of  $\mathcal{P}$  are  $\gamma_1 = 0.5$  MHz and  $\gamma_2 = 2$  MHz.

NV<sup>-</sup> defects at low temperatures is demonstrated using ODMR. Several frequency mixing configurations are employed, including Landau Zener Stückelberg interferometry and two-tone driving spectroscopy. Magnetic driving is applied in the longitudinal and transverse directions using MW and RF fields. The experimental results were compared with prediction of theoretical analysis based on the RWA.

Frequency mixing offers some advantages for sensing applications, including the ability to eliminate crosstalk between driving and detection. This crosstalk problem is important for cavity-based detection of magnetic resonance [2]. At cryogenic temperatures, the method of frequency mixing can be used for applications such as magnetometery of superconducting materials [47]. In this work, ODMR has been employed for exploring the efficiency of a variety of frequency mixing driving configurations. Future study will explore and optimize protocols for cavity-based spin spectroscopy that are based on frequency mixing.

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## Appendix A: Derivation of Eqs. (8) and (9)

The approximate diagonalization of the static part  $H_{dc}$ (6) of H in the vicinity of the GSLAC is based on the



FIG. 5: Two antenna drive near GSLAC. (a) ODMR signal as a function of static magnetic filed *B* and RF solenoid driving power  $P_{\rm L}$ , with a fixed RF solenoid driving frequency of  $\omega_{\rm L}/(2\pi) = 185$  MHz, MW LA driving frequency of  $\omega_{\rm T}/(2\pi) = 5.87$  GHz and MW LA driving power of 25 dBm. (b) The calculated polarization coefficient  $\mathcal{P}$  [see Eq. (4)]. (c) Graphical representation of the frequency mixing matching condition.

relation

$$U^{-1} \begin{pmatrix} -\cos\theta & -\sin\theta & 0\\ -\sin\theta & \cos\theta & 0\\ 0 & 0 & 1 \end{pmatrix} U = \begin{pmatrix} -1 & 0 & 0\\ 0 & 1 & 0\\ 0 & 0 & 1 \end{pmatrix} , \quad (A1)$$

where the unitary matrix U, which is given by

$$U = \begin{pmatrix} \cos\frac{\theta}{2} & -\sin\frac{\theta}{2} & 0\\ \sin\frac{\theta}{2} & \cos\frac{\theta}{2} & 0\\ 0 & 0 & 1 \end{pmatrix} , \qquad (A2)$$

represents a rotation about the z axis through an angle of  $\theta/2$ . The derivation of Eqs. (8) and (9) is performed using the following matrix relations

$$U^{-1} \begin{pmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix} U = \begin{pmatrix} -\cos\theta & \sin\theta & 0 \\ \sin\theta & \cos\theta & 0 \\ 0 & 0 & 0 \end{pmatrix} , \quad (A3)$$
$$U^{-1} \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} U = \begin{pmatrix} \sin\theta & \cos\theta & 0 \\ \cos\theta & -\sin\theta & 0 \\ 0 & 0 & 0 \end{pmatrix} , \quad (A4)$$

$$U^{-1} \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix} U = \begin{pmatrix} 0 & 0 & \cos\frac{\theta}{2} \\ 0 & 0 & -\sin\frac{\theta}{2} \\ \cos\frac{\theta}{2} & -\sin\frac{\theta}{2} & 0 \end{pmatrix},$$
(A5)
$$U^{-1} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix} U = \begin{pmatrix} 0 & 0 & \sin\frac{\theta}{2} \\ 0 & 0 & \cos\frac{\theta}{2} \\ \sin\frac{\theta}{2} & \cos\frac{\theta}{2} & 0 \end{pmatrix},$$
(A6)

and

$$U^{-1} \begin{pmatrix} 0 & x+iy & 0\\ x-iy & 0 & x+iy\\ 0 & x-iy & 0 \end{pmatrix} U$$
$$= \begin{pmatrix} x\sin\theta & x\cos\theta+iy & (x+iy)\sin\frac{\theta}{2}\\ x\cos\theta-iy & -x\sin\theta & (x+iy)\cos\frac{\theta}{2}\\ (x-iy)\sin\frac{\theta}{2} & (x-iy)\cos\frac{\theta}{2} & 0 \end{pmatrix}.$$
(A7)

- [1] Tatsuma Yamaguchi, Yuichiro Matsuzaki, Shiro Saito, Soya Saijo, Hideyuki Watanabe, Norikazu Mizuochi, and Junko Ishi-Hayase, "Bandwidth analysis of ac magnetic field sensing based on electronic spin double-resonance of nitrogen-vacancy centers in diamond", Japanese Journal of Applied Physics, vol. 58, no. 10, pp. 100901, sep 2019.
- [2] Nir Alfasi, Sergei Masis, Roni Winik, Demitry Farfurnik, Oleg Shtempluck, Nir Bar-Gill, and Eyal Buks, "Exploring the nonlinear regime of light-matter interaction using electronic spins in diamond", *Physical Review A*, vol. 97, no. 6, pp. 063808, 2018.
- [3] A Bienfait, P Campagne-Ibarcq, AH Kiilerich, X Zhou, S Probst, JJ Pla, T Schenkel, D Vion, Daniel Estève, JJL Morton, et al., "Magnetic resonance with squeezed microwaves", *Physical Review X*, vol. 7, no. 4, pp. 041011, 2017.
- [4] Roei Levi, Sergei Masis, and Eyal Buks, "Instability in the hartmann-hahn double resonance", *Phys. Rev. A*, vol. 102, pp. 053516, Nov 2020.
- [5] Daniel Gottesman, Alexei Kitaev, and John Preskill, "Encoding a qubit in an oscillator", *Physical Review A*, vol. 64, no. 1, pp. 012310, 2001.
- [6] Ivar Martin, Gil Refael, and Bertrand Halperin, "Topological frequency conversion in strongly driven quantum systems", *Physical Review X*, vol. 7, no. 4, pp. 041008, 2017.
- [7] André Eckardt and Egidijus Anisimovas, "Highfrequency approximation for periodically driven quantum systems from a floquet-space perspective", *New journal* of physics, vol. 17, no. 9, pp. 093039, 2015.
- [8] NB Manson, LJ Rogers, Elena Alexandrovna Wilson, and Changjiang Wei, "Hole burning?eit studies of the nv centre in diamond", *Journal of luminescence*, vol. 130, no. 9, pp. 1659–1667, 2010.
- [9] Pauli Kehayias, Mariusz Mrózek, Victor M Acosta, A Jarmola, DS Rudnicki, Ron Folman, Wojciech Gawlik, and Dmitry Budker, "Microwave saturation spectroscopy of nitrogen-vacancy ensembles in diamond", *Physical Re-*10, 100 (2019) (201

view B, vol. 89, no. 24, pp. 245202, 2014.

- [10] William D Oliver and Sergio O Valenzuela, "Largeamplitude driving of a superconducting artificial atom: Interferometry, cooling, and amplitude spectroscopy", *Quantum Information Processing*, vol. 8, pp. 261–281, 2009.
- [11] Marcus W Doherty, Neil B Manson, Paul Delaney, Fedor Jelezko, Jörg Wrachtrup, and Lloyd CL Hollenberg, "The nitrogen-vacancy colour centre in diamond", *Physics Reports*, vol. 528, no. 1, pp. 1–45, 2013.
- [12] L Rondin, JP Tetienne, T Hingant, JF Roch, P Maletinsky, and V Jacques, "Magnetometry with nitrogen-vacancy defects in diamond", *Reports on Progress in Physics*, vol. 77, no. 5, pp. 056503, 2014.
- [13] Siddharth Dhomkar, Jacob Henshaw, Harishankar Jayakumar, and Carlos A Meriles, "Long-term data storage in diamond", *Science advances*, vol. 2, no. 10, pp. e1600911, 2016.
- [14] Xiaobo Zhu, Shiro Saito, Alexander Kemp, Kosuke Kakuyanagi, Shin-ichi Karimoto, Hayato Nakano, William J Munro, Yasuhiro Tokura, Mark S Everitt, Kae Nemoto, et al., "Coherent coupling of a superconducting flux qubit to an electron spin ensemble in diamond", *Nature*, vol. 478, no. 7368, pp. 221–224, 2011.
- [15] Yuimaru Kubo, Cecile Grezes, Andreas Dewes, T Umeda, Junichi Isoya, H Sumiya, N Morishita, H Abe, S Onoda, T Ohshima, et al., "Hybrid quantum circuit with a superconducting qubit coupled to a spin ensemble", *Physical review letters*, vol. 107, no. 22, pp. 220501, 2011.
- [16] R. Amsüss, Ch. Koller, T. Nöbauer, S. Putz, S. Rotter, K. Sandner, S. Schneider, M. Schramböck, G. Steinhauser, H. Ritsch, J. Schmiedmayer, and J. Majer, "Cavity qed with magnetically coupled collective spin states", *Phys. Rev. Lett.*, vol. 107, pp. 060502, Aug 2011.
- [17] C Grezes, Brian Julsgaard, Y Kubo, M Stern, T Umeda, J Isoya, H Sumiya, H Abe, S Onoda, T Ohshima, et al., "Multimode storage and retrieval of microwave fields in a spin ensemble", *Physical Review X*, vol. 4, no. 2, pp.

021049, 2014.

- [18] Jonathan D Breeze, Enrico Salvadori, Juna Sathian, Neil McN Alford, and Christopher WM Kay, "Continuous-wave room-temperature diamond maser", *Nature*, vol. 555, no. 7697, pp. 493–496, 2018.
- [19] Thomas Wolf, Philipp Neumann, Kazuo Nakamura, Hitoshi Sumiya, Takeshi Ohshima, Junichi Isoya, and Jörg Wrachtrup, "Subpicotesla diamond magnetometry", *Physical Review X*, vol. 5, no. 4, pp. 041001, 2015.
- [20] Scott T. Alsid, Jennifer M. Schloss, Matthew H. Steinecker, John F. Barry, Andrew C. Maccabe, Guoqing Wang, Paola Cappellaro, and Danielle A. Braje, "Solidstate microwave magnetometer with picotesla-level sensitivity", *Phys. Rev. Appl.*, vol. 19, pp. 054095, May 2023.
- [21] Guoqing Wang, Yi-Xiang Liu, Jennifer M Schloss, Scott T Alsid, Danielle A Braje, Paola Cappellaro, et al., "Sensing of arbitrary-frequency fields using a quantum mixer", *Physical Review X*, vol. 12, no. 2, pp. 021061, 2022.
- [22] AK Dmitriev, HY Chen, GD Fuchs, and AK Vershovskii, "Dual-frequency spin-resonance spectroscopy of diamond nitrogen-vacancy centers in zero magnetic field", *Physical Review A*, vol. 100, no. 1, pp. 011801, 2019.
- [23] Alexander K. Dmitriev and Anton K. Vershovskii, "Highcontrast two-frequency optically detected resonances in diamond nitrogen-vacancy centers for timekeeping schemes", *IEEE Sensors Letters*, vol. 4, no. 1, pp. 1– 4, 2020.
- [24] A. K. Dmitriev and A. K. Vershovskii, "Radio-frequency response of the optically detected level anticrossing signal in nitrogen-vacancy color centers in diamond in zero and weak magnetic fields", *Phys. Rev. A*, vol. 105, pp. 043509, Apr 2022.
- [25] Lilian Childress and Jean McIntyre, "Multifrequency spin resonance in diamond", *Physical Review A*, vol. 82, no. 3, pp. 033839, 2010.
- [26] Hannah Clevenson, Edward H. Chen, Florian Dolde, Carson Teale, Dirk Englund, and Danielle Braje, "Diamondnitrogen-vacancy electronic and nuclear spin-state anticrossings under weak transverse magnetic fields", *Phys. Rev. A*, vol. 94, pp. 021401, Aug 2016.
- [27] HJ Mamin, MH Sherwood, M Kim, CT Rettner, K Ohno, DD Awschalom, and D Rugar, "Multipulse double-quantum magnetometry with near-surface nitrogen-vacancy centers", *Physical review letters*, vol. 113, no. 3, pp. 030803, 2014.
- [28] Jingwei Zhou, Pu Huang, Qi Zhang, Zixiang Wang, Tian Tan, Xiangkun Xu, Fazhan Shi, Xing Rong, S Ashhab, and Jiangfeng Du, "Observation of time-domain rabi oscillations in the landau-zener regime with a single electronic spin", *Physical review letters*, vol. 112, no. 1, pp. 010503, 2014.
- [29] Pu Huang, Jingwei Zhou, Fang Fang, Xi Kong, Xiangkun Xu, Chenyong Ju, and Jiangfeng Du, "Landau-zener-stückelberg interferometry of a single electronic spin in a noisy environment", *Physical Review X*, vol. 1, no. 1, pp. 011003, 2011.
- [30] Takumi Mikawa, Ryusei Okaniwa, Yuichiro Matsuzaki, Norio Tokuda, and Junko Ishi-Hayase, "Electron-spin double resonance of nitrogen-vacancy centers in diamond under a strong driving field", *Physical Review A*, vol. 108,

no. 1, pp. 012610, 2023.

- [31] V Jacques, P Neumann, J Beck, M Markham, D Twitchen, J Meijer, F Kaiser, G Balasubramanian, F Jelezko, and J Wrachtrup, "Dynamic polarization of single nuclear spins by optical pumping of nitrogenvacancy color centers in diamond at room temperature", *Physical review letters*, vol. 102, no. 5, pp. 057403, 2009.
- [32] GD Fuchs, Guido Burkard, PV Klimov, and DD Awschalom, "A quantum memory intrinsic to single nitrogen–vacancy centres in diamond", *Nature Physics*, vol. 7, no. 10, pp. 789–793, 2011.
- [33] D. Farfurnik, N. Alfasi, S. Masis, Y. Kauffmann, E. Farchi, Y. Romach, Y. Hovav, E. Buks, and N. Bar-Gill, "Enhanced concentrations of nitrogen-vacancy centers in diamond through tem irradiation", *Applied Physics Letters*, vol. 111, no. 12, pp. 123101, 2017.
- [34] D. M. Pozar, *Microwave Engineering*, John Wiley and sons, 1998.
- [35] Nir Alfasi, Sergei Masis, Oleg Shtempluck, and Eyal Buks, "Detection of paramagnetic defects in diamond using off-resonance excitation of nv centers", *Phys. Rev. B*, vol. 99, pp. 214111, Jun 2019.
- [36] Preeti Ovartchaiyapong, Kenneth W Lee, Bryan A Myers, and Ania C Bleszynski Jayich, "Coherent strainmediated coupling of a single diamond spin to a mechanical resonator", arXiv:1403.4173, 2014.
- [37] ER MacQuarrie, TA Gosavi, NR Jungwirth, SA Bhave, and GD Fuchs, "Mechanical spin control of nitrogenvacancy centers in diamond", *Physical review letters*, vol. 111, no. 22, pp. 227602, 2013.
- [38] D. M. Berns, W. D. Oliver, S. O. Valenzuela, A. V. Shytov, K. K. Berggren, L. S. Levitov, and T. P. Orlando, "Coherent quasiclassical dynamics of a persistent current qubit", *Physical Review Letters*, vol. 97, no. 15, pp. 150502, 2006.
- [39] Eyal Buks, Chunqing Deng, Jean-Luc F. X. Orgazzi, Martin Otto, and Adrian Lupascu, "Superharmonic resonances in a strongly coupled cavity-atom system", *Phys. Rev. A*, vol. 94, pp. 033807, Sep 2016.
- [40] Charles P Slichter, Principles of magnetic resonance, vol. 1, Springer Science & Business Media, 2013.
- [41] Eyal Buks, Quantum mechanics Lecture Notes, http://buks.net.technion.ac.il/teaching/, 2023.
- [42] AG Anderson, "Nuclear spin absorption spectra in solids", *Physical Review*, vol. 125, no. 5, pp. 1517, 1962.
- [43] Hung Cheng, "Spin absorption of solids", Physical Review, vol. 124, no. 5, pp. 1359, 1961.
- [44] LJF Broer, "On the theory of paramagnetic relaxation", *Physica*, vol. 10, no. 10, pp. 801–816, 1943.
- [45] JT Daycock and G Parry Jones, "Subsidiary resonances in nuclear magnetic resonance", *Journal of Physics C: Solid State Physics*, vol. 2, no. 6, pp. 998, 1969.
- [46] SN Shevchenko, S. Ashhab, and F. Nori, "Landau–zener– stückelberg interferometry", *Physics Reports*, vol. 492, no. 1, pp. 1–30, 2010.
- [47] Nir Alfasi, Sergei Masis, Oleg Shtempeluk, Valleri Kochetok, and Eyal Buks, "Diamond magnetometry of meissner currents in a superconducting film", *AIP Ad*vances, vol. 6, pp. 075311, 2016.