Spin State Dynamics in a Bichromatic Microwave Field: Power Narrowing and Broadening of a Magnetic Resonance

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Abstract

Driving an open spin system by two strong, nearly degenerate fields enables addressing populations of individual spin states, characterization of their interaction with thermal bath, and measurements of their relaxation/decoherence rates. With such addressing we observe nested magnetic resonances having nontrivial dependence on microwave field intensity: while the width of one of the resonances undergoes a strong power broadening, the other one exhibits a peculiar field-induced stabilization. We also observe light-induced narrowing of such composite resonances. The observations are explained by the dynamics of bright and dark superposition states and their interaction with reservoir.
Driving spin systems with specific time-dependent fields results in nontrivial dynamics enabling observation of a plethora of interesting phenomena. Representative examples are spin echo [1], spin locking [2, 3], coherent population trapping (CPT) [4–7], coherent population oscillations (CPO) [8–17]. They permit important applications in quantum-state engineering [18, 19], quantum metrology [20], and bio-medical diagnostics [21].

A fundamental condition for successful engineering of spin ensembles is sound understanding of their relaxation, depolarization, decoherence and ability to control interactions with other fields and/or systems. Significant progress in such control has been achieved by dark-state formation [4], application of spin echo-related techniques enabling dynamical decoupling [22], and hybrid coupling of spin system with cavities or opto-mechanical devices [23–26]. Numerous recent experiments with open systems coupled to cavities [25], well controlled samples [27, 28], and specially tailored spin environment [3, 7] demonstrated significant mitigation of the relaxation processes and feasibility of tailoring effects of specific baths.

Below, we present results of our theoretical and experimental study of bichromatic driving of \( S = 1 \) spins and show that by application of hole burning [29–31] and CPO [13] we can create coherent superposition states in an open inhomogeneously broadened two-level spin ensemble. This enables observation of long-lived states, i.e. *system’s stabilization* and reduction or even elimination of power broadening of one of the dark state superpositions. These resonances can be regarded as Fourier transforms of multiexponential decays of populations and coherences of spin-states which represent decoherence and relaxation. In a more fundamental context, such behavior reflects nonlinear and nonmarkovian dynamics of open quantum systems coupled to a heat bath (reservoir) [32, 33].

This Letter presents alternative approach to the issue of multiexponent decays and reduction of relaxation by lineshape analysis of composite hole-burning resonances, rather than by measurements of time evolution. Our theoretical modelling is verified experimentally with ensemble of nitrogen vacancy (NV) color centres in diamond crystal excited by a green light and perturbed by two microwave (MW) fields of comparable strengths and nearly resonant frequencies \( \omega_1, \omega_2 \). As described in Refs. [13, 29], such situation enables observation of the holes burned in the inhomogeneously broadened optically recorded magnetic resonance (ODMR). Two different situations may be realized: (i) one of the MW fields is tuned to one transition, e.g. \( m_S = 0 \leftrightarrow m_S = -1 \) while the second MW is close to resonance with
FIG. 1. The model system of an open $S = 1$ spin system perturbed by two MW fields nearly resonant with the $|0\rangle - |1\rangle$ transition and coupled to a reservoir $R$ with rates $\gamma_0, \gamma_1$. In appropriately strong magnetic field one of the $|\pm 1\rangle$ states (marked with a broken line) is nonresonant with the MWs and can be ignored in the modelling.

another one, e.g. $m_S = 0 \leftrightarrow m_S = +1$, and (ii) both MWs are close to resonance with one transition between either spin states of the ground state. In the first case, Lorentzian-shaped holes of the natural linewidth $\Gamma = 1/T_2^*$ are burned within the inhomogenously broadened ODMR dips [29]. In the second case, characteristic coherent population oscillations (CPO) take place between the coupled spin states and result in a complex resonance shape (Fig. 2) composed of three resonance contributions associated with populations of two spin states and their coherence [13], herewith defined as composite resonance.

In this Letter, we focus on the second case and analyze peculiar dependence of the composite resonances on light and MW powers. In particular, we show that the relaxation rate associated with the population of one spin state becomes stabilized by an external light and/or MW fields, i.e. the width of one resonance component reaches a constant value, whereas the population of another state undergoes strong power broadening by MW field and narrowing by light. Light-power induced narrowing was observed also in optical pumping experiments [34–36] and with ODMR studies with NV centers [37], but the narrowing reported here addresses a narrow (sub-natural, i.e. below $1/T_2^*$ linewidth) spectral feature and has not been observed previously.
Since in a magnetic field of a few mT the $m_S = 0 \leftrightarrow m_S = \pm 1$ transitions of the NV center are well resolved, we analyse two-state dynamics of states $m_S = 0$, $m_S = -1$, denoted as $|0\rangle$ and $|1\rangle$, respectively, perturbed by two quasi resonant MW fields (frequencies $\omega_1, \omega_2$). We use a density matrix formalism and assume that each state population $\rho_{ii}$ ($i = 0, 1$), and their coherence, $\gamma_{01}$, relax to equilibrium with rates $\gamma_i$ and $\Gamma$ as: $\dot{\rho}_{ii} = -\gamma_i (\rho_{ii} - \rho_{0i}^0)$ and $\dot{\rho}_{01} = -\Gamma \rho_{01}$, where $\rho_{0i}^0$ are the initial equilibrium populations. For a closed system, the individual rates $\gamma_i$ need to be equal for both states, hence their difference $\gamma_0 \neq \gamma_1$ reveals system’s interaction with thermal bath, consisting of, e.g. crystal impurities, phonon modes and, most importantly, other NV states beyond the applied two-level model (Fig. 1). Such situation occurs in measurements of longitudinal relaxation time ($T_1$) of dense samples and causes multiexponential decays [13, 27, 28, 38].

By taking into account the first harmonics of the beat frequency $\Delta \omega = \omega_1 - \omega_2$ we extend the analysis of CPO in Ref. [13] to a nonperturbative range of strong MW fields and calculate the population difference $\Delta n = n_0 - n_1$ between spin states $|0\rangle$, $|1\rangle$. $\Delta n$ is proportional to the change of fluorescence intensity that reproduces the ODMR and hole-burning signals. Making the steady-state approximation with respect to $\omega_1$ and $\omega_2$ but following the time evolution with $\Delta \omega$ [39] we arrive at a convenient analytical expressions:

$$\Delta n = \bar{D} \left[ 1 - \frac{M}{2} \left( \frac{e^{i \Delta \omega t}}{m \Gamma/2 + L} + \text{c.c.} \right) \right],$$

$$D = \frac{\gamma \Gamma/2}{\gamma \Gamma/2 + L - M^2/(4R)} (\rho_{22}^0 - \rho_{11}^0),$$

where:

$$\frac{1}{\gamma} \equiv \frac{1}{\gamma_0} + \frac{1}{\gamma_1}, \quad M \equiv \Omega_1 \Omega_2 [\mathcal{L}(\delta_1) + \mathcal{L}(\delta_2)],$$

$$L \equiv \frac{\Omega_1^2 \mathcal{L}(\delta_1) + \Omega_2^2 \mathcal{L}(\delta_2)}{m \Gamma/2 + L}, \quad \frac{1}{m} \equiv \frac{1}{\gamma_0 + i \Delta \omega} + \frac{1}{\gamma_1 + i \Delta \omega},$$

$$\frac{1}{\Gamma} \equiv \frac{1}{m \Gamma/2 + L} + \frac{\Gamma}{m \Gamma/2 + L}, \quad \mathcal{L}(\delta_i) = \frac{(\Gamma/2)^2}{\delta_i^2 (\Gamma/2)^2},$$

and $\delta_i$ denotes the detuning of $\omega_i$ from the $|0\rangle$ - $|1\rangle$ transition. $\Omega_i$ stands for the Rabi frequency of the $\omega_i$ field related with its power $P_i$ via dimensionless saturation parameter $G_i = P_i/(\gamma \Gamma)$. The right-hand side of Eq.(1a) consists of a constant part $\bar{D}$ and a time-dependent one which represents CPO with frequency $\Delta \omega$. As discussed in Ref.[13], its observation requires careful phase synchronization of the applied MWs, whereas the $\bar{D}$ part in observed already under DC conditions when scanning $\Delta \omega$.

Expression (1) enables calculation of the composite resonance shape for a range of experimental conditions. Figure 2(a) depicts the calculated fluorescence signals and Fig. 2(b) its
FIG. 2. (a) Simulated resonance profile (solid red) in the center of an ODMR signal (dashed black). The inset shows a single-shot (solid black) and a random-phase averaged (red) CPO signals reflecting $\Delta n = \bar{D}$. (b) Experimental data fitted with composite triple-Lorenz curve (widths $w_\Gamma$, $w_1$ and $w_0$) on a Gaussian background.

comparison with the observations presented in Ref. [13] demonstrating very good agreement. Furthermore, Fig. 2(b) depicts the expansion of experimental curve into elementary components associated with three contributions determined by fitting the composite resonance to three Lorentzian profiles having different amplitudes and widths $w_0$, $w_1$, $w_\Gamma$ related with the population relaxation rates: $\gamma_0$ and $\gamma_1$ and relaxation rate $\Gamma$ of coherence $\rho_{01}$, respectively.

The results shown in Fig. 2(b) were obtained with averaging over the MW phases, hence they reflect the time-averaged part of Eq. (1), $\Delta n = \bar{D}$.

By changing MW power, ie. the value of the saturation parameters $G_i$, we observe that widths of the three signal components exhibit qualitatively different dependence on MW powers. These widths are found by analyzing complex poles of Eq. (1). When $\Gamma \gg \gamma_0, \gamma_1$, and both MWs are equally strong ($G_1 = G_2 = G$), $w_\Gamma$ is almost constant and broadening of $w_0$ and $w_1$ appears to depend on whether $\gamma_0$ and $\gamma_1$ are equal or not.

In the general case, when $\gamma_0 \neq \gamma_1$, the analytical expressions for $w_{0,1}(G)$ takes the form

$$w_{0,1}(G) = \sqrt{\frac{b}{2} \pm \frac{1}{2} P},$$

where

$$b = 2\gamma_0^2 \left[ 1 + 2G + \frac{x}{1+2G} - 2x(3-x)\frac{G^2}{1+2G} \right],$$

$$P = \left( \frac{1}{1+2G} \right)^{\frac{1}{2}}.$$
FIG. 3. Power dependence of the widths $w_0(G)$ and $w_1(G)$ versus MW power (in units of G). (a) for $x = 0$ with $\gamma_0 = \gamma_1 = 0.3$ (red) and $x \neq 0$ with $\gamma_0 = 0.1 \gamma_1 = 5$ (blue), (b) for $\gamma_d = \frac{1}{2}(\gamma_0 - \gamma_1)$ gradually changing between 0 and 0.2 ($x$ from 0 to 0.44). The widest component with $w_\Gamma \gg w_0, w_1$, while not displayed, is not power broadened in the considered range of $G$.

For weak driving $G \ll 1$, the system’s coupling with reservoir (R) dominates the interaction and the widths of both curves $w_0(G)$ and $w_1(G)$ exhibit power broadening with the same slope $\gamma_S \sqrt{G}$. When $G$ increases above $G \approx 1$ the two widths behave differently: $w_0(G)$ asymptotically approaches $\gamma_S$, whereas $\omega_0(G)$ broadens monotonously (approximately as $\sqrt{G}$) as illustrated by Fig. 3.

For strong driving, $G \gg 1$, Eqs. (2) may be approximated and cast in the form $w_{0,1}^2(G) \approx \gamma_S^2 + 2\gamma_S^2 G \pm 2\gamma_S^2 G$, with solutions:

\[
\begin{align*}
    w_{0,1} &\approx \begin{cases} 
        \gamma_S \sqrt{1 + 4G} & \text{if } x = 0, \\
        \gamma_S & \text{if } x \neq 0.
    \end{cases}
\end{align*}
\]

Equations (3) reveal two fundamentally different behaviors of the effective widths: $w_1(G)$ strongly broadened and $w_0(G)$ immune to power broadening. If $\gamma_0 = \gamma_1 = \gamma_S$ both components have equal widths at $G = 0$, i.e. $w_0 = w_1 = \gamma_S$. When $G$ increases, only $w_1(G)$ becomes power broadened while $w_0$ remains constant for all $G$.

Figure 3(a) illustrates power dependences of $w_0(G)$ and $w_1(G)$ calculated using Eqs. (2)
for equal and differing initial values of $\gamma_0$ and $\gamma_1$. Transition between the two cases is illustrated in Fig. 3(b) for a constant value of $\gamma_S$ and gradually changing $\gamma_d$ or dimensionless parameter $x$.

We interpret the above results in terms of the superposition states, similarly as in the effects of coherent population trapping (CPT) [4] and collisional decoupling of sensitized fluorescence of excited atoms [40, 41]. The similarity derives from the fact that in the intermediate range of $G$ there is a competition between direct relaxation of each state’s population to the reservoir and their coherent driving by two nearly degenerate MW fields. The MWs mix states $|0\rangle$ and $|1\rangle$ and create superpositions $|\pm\rangle = \frac{1}{\sqrt{2}}(|0\rangle \pm |1\rangle)$. These superpositions are related with the ± signs in Eq. (3a) and represent either constructive $(2\gamma_S^2 G + 2\gamma_S^2 G)$, or destructive $(2\gamma_S^2 G - 2\gamma_S^2 G)$ interference in their coupling to $\mathcal{R}$, which is responsible for the observed nontrivial dynamic of the spin states. Indeed, denoting the interaction of states $|0,1\rangle$ with the reservoir $|R\rangle$ as $V_R$ we have $\langle R| V_R |0\rangle = -i\sqrt{\gamma_0}$ and $\langle R| V_R |1\rangle = -i\sqrt{\gamma_1}$, which for the superpositions created by strong MW driving yields $\langle R| V_R |\pm\rangle = -i\sqrt{\gamma_0 \pm \gamma_1}$, i.e. reflects strong broadening for one resonance component and its reduction for the other one.

The above analysis was verified in a CW ODMR experiment with the laser-excited (532 nm light) NV diamond ensemble ([NV]∼10 ppm) and two MWs tuned to the same $m_S = 0 \leftrightarrow m_S = -1$ transition in the ground state [13, 29] in a magnetic field of 4 mT. The composite resonances were recorded for various powers of the MW fields and fitted to the superposition of three resonance shapes as illustrated in Fig. 2(b). Since there was no phase locking of the MWs, the oscillatory CPO structure shown in inset in Fig. 2 was absent. This is noteworthy that even for $\gamma_0 \approx \gamma_1$ and $G \ll 1$, when the lineshapes of individual contributions to composite resonance differ so little that their reliable addressing seems to be impossible, the measurements with higher $G$ cause strong power broadening of only one of the components and almost no broadening of the other one, which enables their reliable identification and addressing.

Figure 4 presents results of the measurements of the width of all three contributions $w_0$, $w_1$, and $w_1$ as a function of MW power. In agreement with the theoretical predictions we observe very weak dependence of $w_0$ on MW power which we interpret as the light-induced stabilization of the state $|0\rangle$ population.

Our theoretical modelling strongly simplifies the role of optical pumping, reducing it to
FIG. 4. MW-power dependences of the $w_0$, $w_1$, and $w_\Gamma$ widths (blue triangles, red circles and black squares, respectively). (b) fit of the measured $w_0$ and $w_1$ values to theoretical predictions (solid lines based on Eqs. (3)).

the establishment of the initial spin polarization (state populations $\rho_{00}^{(0)}, \rho_{11}^{(0)}$). To get more insight on that role, we studied experimentally the effect of light power $P_{\text{light}}$ on the measured spin dynamics. Figure 5 shows the results of measurements of the widths $w_0$ and $w_1$ as a function of CW light power with constant MW power ($P_{MW} = -15 \text{ dBm}$, corresponding to $G \sim 10$). Similarly to Fig. 4, in the applied range of light intensities we did not observe any significant change of $w_\Gamma$ and $w_0$, which demonstrates stabilization of population $\rho_{00}$ and coherence $\rho_{01}$ against light perturbation [42]. On the other hand, a strong power narrowing of $w_1$ is clearly visible.

Although the power narrowing effect has been already observed with NV diamonds for ODMR resonances [37], Fig. 5 represents the narrowing for much finer spectral structures burnt in the ODMR signal, which looks qualitatively consistent with the earlier observations but occurs in a much narrower intensity range (light power below 20 mW). At higher powers both resonance widths become saturated and do not depend on $P_{\text{light}}$. Similarly as in [37], we ascribe this narrowing to the effect of optical pumping and intersystem crossing which can be described in a five-state energy level system.

In summary, we have described novel effect of interaction of two MW fields with an open system. Reported analysis of the structure of the composite resonances and their power
FIG. 5. Light-narrowing effect the of the resonance width for a fixed MW power. Structures of the composite resonance analyzed here have sub-natural linewidths related to the population dynamics.

broadening offers a sensitive way of studying the relaxation/decoherence mechanism of spin ensembles. Since the Fourier transform of the composite resonances represents multiexponential decay of populations $\rho_{00}$, $\rho_{11}$ and coherence $\rho_{01}$, the developed CPO-based methodology provides a useful alternative to standard time-resolved studies of spin dynamics. This two-field methodology enables addressing of individual spin states and studies of their interaction with environment which is impossible with standard, CW ODMR measurements where the resonances are jointly affected by both relaxation rates. The individually addressed resonance components manifest properties of the strongly driven spin ensemble, but by extrapolation of their widths to $G = 0$, relaxation rates of the unperturbed populations of individual states and their coherence can be determined exactly.

Applying such procedure we have found that the unperturbed population relaxation rates
are different for the two considered spin states in high NV-density sample which evidences their unequal interaction with thermal reservoir. Moreover, we demonstrate that components of the composite resonance are very differently perturbed by strong MW field: while one component is strongly broadened, the other is nearly broadening-free. We regard this behavior as stabilization of spin states by strong driving field and interpret it as analogy to the well-known quantum interference effect with dark and bright state superpositions. We also demonstrated light-power narrowing of these composite resonances.

While the reported theoretical analysis and measurements are focused on spin ensemble in NV diamond, the analysis and discovered phenomena are quite general and may be applied for precision characterization of spin dynamics of various paramagnetic samples and control of their interaction with external fields.

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The observed independence of $w_0$ on light power should not be confused with the effect reported in A. Dréau, M. Lesik, L. Rondin, P. Spinicelli, O. Arcizet, J.-F. Roch, and V.
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Supplemental material.

Spin State Dynamics in a Bichromatic Microwave Field: Power Narrowing and Broadening of a Magnetic Resonance

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We analyze interaction of a two-level open system with two electromagnetic fields of nearly equal frequencies $\omega_1$ and $\omega_2$, close to resonance with the transition frequency $\omega_0$. This general model may be applied to interpret results of a magnetic resonance experiment with two microwave fields acting on a spin system, like our recent study of coherent population oscillations resonances in NV diamonds [S1].

The analysis is performed with the density matrix formalism and is based on the master equation for the simplified two-level system:

\[\dot{\rho}_{00} = -\gamma_0(\rho_{00} - \rho^0_{00}) - i(W_1 + W_2)(\rho_{01} - \rho^*_{01}),\]
\[\dot{\rho}_{11} = -\gamma_0(\rho_{11} - \rho^0_{11}) + i(W_1 + W_2)(\rho_{01} - \rho^*_{01}),\]  \hspace{1cm} (S1)
\[\dot{\rho}_{01} = -(\frac{\Gamma}{2} - i\omega_0)\rho_{01} - i(W_1 + W_2)(\rho_{00} - \rho_{11}),\]

where $\gamma_s$ denote relaxation rates of populations of states $|s\rangle$ ($s = 0, 1$) and $\rho^0_s$ stands for their equilibrium populations, $\Gamma$ is the relaxation rate of coherence $\rho_{01}$; $W_k = \frac{1}{2}\Omega_k(e^{-i\omega_k t} + e^{i\omega_k t})$ represents interaction of each of the MW fields ($k = 1, 2$) with the two-level system, the interaction strengths is characterized by the Rabi frequencies $\Omega_k$.

By substituting $\rho_{01} = \frac{1}{2}[\sigma_1(e^{-i\omega_1 t} + e^{i\omega_1 t}) + \sigma_2(e^{-i\omega_2 t} + e^{i\omega_2 t})]$ and within a rotating-wave approximation (RWA) with respect to frequencies $\omega_k$ we eliminate the fast evolution, follow the slowly-varying coherence envelopes $\sigma_k$ and arrive at coupled equations for coherence amplitudes

\[\dot{\sigma}_1 = -(\frac{\Gamma}{2} + i\delta_1)\sigma_1 - i\Omega_1(\rho_{00} - \rho_{11}),\]  \hspace{1cm} (S2)
\[\dot{\sigma}_2 = -(\frac{\Gamma}{2} + i\delta_2)\sigma_2 - i\Omega_2(\rho_{00} - \rho_{11}),\]

where $\delta_n = \omega_n - \omega_0$ (similar decomposition has been used also in Ref.[S2] ).

Substitution of (S1) to (S2) enables adiabatic elimination of coherence and allows the expansion of the time evolution of population difference:

\[\Delta n = \bar{D} + D^{(+)}e^{i\Delta\omega t} + D^{(-)}e^{-i\Delta\omega t},\]  \hspace{1cm} (S3)

as a sum of stationary population difference $\bar{D}$ and contributions oscillating as $e^{\pm i\Delta\omega t}$ with $\Delta\omega = \omega_1 - \omega_2$; Equation (S3) determines amplitudes of the resonances in ODMR measurements described in the main article and is a common result in many-field interaction.
analysis, in particular, it is at the roots of wave-mixing phenomena in nonlinear optics and
coherence population oscillations [S3–S11].

In principle, higher-order harmonics \((e^{±ni\Delta_\omega t} \text{ terms with } n>1)\) of the frequency difference \(\Delta_\omega\), should be taken into account, but the analysis limited only to the first harmonic already reproduces the salient features of the observed effects. Especially, since the most important measurements reported in this work concern extrapolation of the resonance widths to zero power where the higher harmonics become negligible weak, the applied simplification is fully justified. Solving Eqs. (S1) with the first harmonic to all orders in MW power, we arrive at
handy analytical expressions \(m_i\) (Eq. (1) of the main text):

\[
\Delta n = \bar{D} \left[ 1 - \frac{M}{2} \left( \frac{e^{i\Delta_\omega t}}{m\Gamma/2 + L} + \frac{e^{-i\Delta_\omega t}}{m^*\Gamma/2 + L} \right) \right],
\]

(S4)

\[
\bar{D} = \frac{\gamma\Gamma/2}{\gamma\Gamma/2 + L - M^2/(4R)} D^0,
\]

(S5)

here we have introduced the following notation: \(D^0 = \rho^0_{00} - \rho^0_{11}, \; L = \Omega_1^2\mathcal{L}(\delta_1) + \Omega_2^2\mathcal{L}(\delta_2), \; M = \Omega_1\Omega_2 [\mathcal{L}(\delta_1) + \mathcal{L}(\delta_2)], \; \mathcal{L}(\delta_k) = \frac{(\Gamma/2)^2}{\delta_k^2 + (\Gamma/2)^2}, \; \frac{1}{\gamma} = \frac{1}{\gamma_0} + \frac{1}{\gamma_1}, \; \frac{1}{R} = \frac{1}{m\Gamma/2 + L} + \frac{1}{m^*\Gamma/2 + L}, \; m = \frac{m_{m_1}}{m_0 + m_1}, \; m_0 = \gamma_0 + i\Delta_\omega, \; \text{and} \; m_1 = \gamma_1 + i\Delta_\omega.

Equation (S4) consists of two parts, the time-dependent term in square brackets and the static amplitude \(\bar{D}\). Both parts, however, depend on the interaction with MW fields. The quantity \(L\) reflects saturation exerted by each of the fields independently, whereas quantity \(M\) reflects the \textit{cross-saturation}, i.e. joint action of both MWs which is the essence of the CPO effect where populations \(\rho^0_{00}, \rho^0_{11}\) are modulated by the interference (beating) of both MW fields. As discussed in [S1], observation of the time-dependent part requires careful phase control. Without proper phase synchronization, the time dependent part of Eq. (S4) averages to zero. Hereinafter, we focus on phase-averaged signals \(\Delta n = \bar{D}\).

The initial, equilibrium population difference, \(D^0\), reflects the inhomogenous broadening of our ensemble, which we model as a Gaussian \(D^0(\omega) = \rho^0_{00} - \rho^0_{11} = e^{-\left(\frac{\omega - \omega_0}{w}\right)^2}\), with \(w = 2\sqrt{\ln 2}\Gamma^*, \) where \(\Gamma^*\) represent the inhomogenously broadenend width. Moreover, \(D^0\) is determined by the optical pumping of the green laser beam and, in general, depends on its intensity, as reported in earlier ODMR studies [S12]. In this work we did not account for this dependence but applied constant light intensity and took \(D^0\) as a constant parameter in our theoretical analysis. By measuring the widths of the composite resonances for a range of light intensities we found that they undergo \textit{light narrowing}. This process has been
observed for high laser intensities in a cw single-MW-field ODMR where the resonance width (typically $\sim 1$ MHz) results from the spin dephasing additionally broadened by MW field. In this work we observe a similar effect, however, with much narrower ($\sim$1-100 kHz) structures of composite resonances and for lower laser intensities.

To get more insight into the physics of the observed composite resonances we analyze the spectral properties of the signal when scanning the frequency of one of the MW fields with the assumption that both are equally strong $\Omega_1 = \Omega_2 = \Omega$ and one of them is resonant with the transition frequency $\omega_1 = \omega_0$. In that case formula S5 simplifies to:

$$\bar{D} = \frac{1}{1 + S_L - \frac{1}{2} S_L^2 \text{Re} \left[ \frac{\gamma}{m + \gamma S_L} \right]} D^0,$$

(S6)

where $S_L = G(1 + \mathcal{L})$ and $G = \frac{\Omega^2}{\gamma^2/2}$ is the dimensionless saturation parameter. In the denominator of (S6), the term $S_L$ represents the regular hole-burning, i.e. the $+/-$ case of Ref. [S13] where $\omega_1$ and $\omega_2$ drive distinct transitions (e.g. $m_S = 0 \leftrightarrow m_S = -1$ and $m_S = 0 \leftrightarrow m_S = +1$, where as the term with $S_L^2$ represents the CPO effects responsible for the appearance of composite resonances ($++$ or $-/$ case). The resonance shape is determined by the dependence of $D(G)$. Since in experiments with dense ensembles we have $\Gamma \gg \gamma_0, \gamma_1$ Eq. (S6) can be further simplified by restricting the analysis to the narrow spectral range around $\gamma_0, \gamma_1$ where $S_L = 2G$. Under such conditions, it is the last term of denominator in (S6) that reflects the narrowest structure of $\bar{D}$. It can be analyzed by decomposition of the function $\frac{1}{m + \gamma S_L}$ into simple fractions

$$\frac{1}{m + \gamma S_L} = \frac{m_0 + m_1}{m_0 m_1 + \gamma S_L (m_0 + m_1)} = \frac{\gamma_0 + \gamma_1 + 2i \Delta_{\omega}}{-\Delta_{\omega}^2 + i \Delta_{\omega} (\gamma_0 + \gamma_1 + 2\gamma S_L) + (+i \Delta_{\omega}) + \gamma_0 \gamma_1 + \gamma S_L (\gamma_0 + \gamma_1)}$$

(S7)

which has the following complex roots

$$d_{1,2} = \frac{i}{2} (\gamma_0 + \gamma_1) + i \gamma S_L \pm i \sqrt{(\gamma S_L)^2 + (\frac{\gamma_0 - \gamma_1}{2})^2}.$$

(S8)

With $d_{1,2}$ given by (S8) one can write (S6) as

$$\bar{D} = \left( \frac{D^0}{1 + S_L} \right) \frac{(\Delta_{\omega}^2 + d_1^2)(\Delta_{\omega}^2 + d_2^2)}{\Delta_{\omega}^4 + \Delta_{\omega}^2 [d_1^2 + d_2^2 - \gamma_G (d_1 + d_2 - \gamma_S)] + d_1^2 d_2^2 - \gamma_G d_1 d_2},$$

(S9)
where $\gamma_G = \gamma \frac{G^2}{1+G^2}$ and $\gamma_S = \frac{1}{2}(\gamma_0 + \gamma_1)$ which yields roots of (S8) $\Delta_{1,2}$ as $\sqrt{D_{1,2}}$ in the form

$$D_{1,2} = -\frac{b}{2} \pm \frac{1}{2} \sqrt{b^2 - 4c}, \quad (S10)$$

where $b = d_1^2 + d_2^2 - \gamma_G(d_1 + d_2 - \gamma_S)$, $c = d_1^2 d_2^2 - \gamma \gamma_G d_1 d_2$.

Based on Eq. (S10), one can find $D_{1,2}$ as function of all relevant parameters, in particular the MW power, and then determine effective widths $w_{1,2}$ of the narrow components of composite resonance as $\sqrt{D_{1,2}}$. As we show in the main article, these quantities have different behavior depending on initial, unperturbed values of the population relaxation constants $\gamma_0$ and $\gamma_1$ which result from competition of strong coherent driving by the two MWs with a coupling to the reservoir. In analogy with the well-known properties of coupled and uncoupled superpositions (Refs.[4, 40, 41] of the main text), this competition is responsible for the discovered stabilization effect and the specific dependence of the MW power broadening as well as the light-induced narrowing effect described in the main paper.

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