Optimal demonstration of Autler–Townes splitting

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The atom-light interaction in a three-level system has shown significant physical phenomena, such as electromagnetically induced transparency and Autler–Townes splitting (ATS), for broad applications in classical and quantum information techniques. Here, we optimally demonstrated the ATS with a quantum state manipulation method. The ATS in the dephasing-dominated diamond NV center system was successfully recovered by coherent microwave control, which cannot be observed with traditional method. The dynamical process of ATS was investigated in detail, revealing a non-trivial quantum interference with geometric phase modulations. Based on the quantum interference, the signal of the optimal ATS is twice as intense as those with traditional observation method.

I. INTRODUCTION

Atom-light interaction is a fundamental topic in quantum optics and atomic physics. In this realm, the optical response of the quantum multilevel system can be dramatically modified by quantum interferences among different transition pathways, or by the strong Stark effect [1]. As a typical representative of the former, electromagnetically induced transparency (EIT) [2–4] can create an ultra-narrow transparency window and delicately control the absorption and dispersion of the medium, and thus many remarkable applications are being explored [2–4]. The Stark effect also creates a transparency window because of the doublet splitting structure in the absorption spectrum, which is called Autler–Townes splitting (ATS). ATS has been employed to measure transition dipole moments [5], to control the spin-orbit interaction in quantum system [6], to suppress quantum decoherence [7–10], to dynamically control resonance fluorescence spectra [11] and to create disorder for time crystals [12]. In the last two decades, many systems have been used to investigate EIT and ATS, such as atoms [6, 13], superconducting systems [14–16], quantum dots [17–21], defects in diamond [9, 19–22], and nano-photonic systems [23–27].

Until now, almost all studies of EIT and ATS have been performed on atomic-like system based on the traditional spectral-domain observational method with long-duration driving (coupling and probe) pulses [5–11, 15, 16, 18, 27], wherein quantum decoherence is dominated by the longitudinal relaxation process [2–4]. However, with recent developments in materials science [28–31], rapid pure dephasing processes dominate the quantum decoherence, such as in solid spin systems and superconducting systems [29]. In these systems, when the driving pulses is much longer than the dephasing time, the quantum coherence is lost and the EIT and ATS phenomena disappear. Hence, the traditional observation method imposes a serious restriction on the investigation and application of both phenomena. In this Letter, we optimally demonstrated the ATS by applying the quantum state manipulation [32, 33] method. The ATS was successfully recovered in diamond nitrogen vacancy (NV) center system where the quantum decoherence is dominated by the dephasing process [28–31].

The diamond NV center has been one of promising candidates for quantum information processes. Many studies have successfully demonstrated one- and multi-qubit coherent operations [34–37] at room-temperature. Using the electron spin triplet state of the NV center, the V-type quantum three-level system can be directly obtained. With the quantum state manipulation method, ATS is observed in such a dephasing dominated system and the dynamic process of ATS is investigated in detail by controlling the pulse sequence. A nontrivial oscillation driven by the probe and coupling field was experimentally revealed, including both geometric phase and quantum interference in this three-level coupling. This dynamic behavior is notably different from that of a two-level system with Rabi oscillation. Moreover, with delicately control of the interference and geometric phase, the signal intensity of ATS is twice that the traditional method due to quantum interference, which is the optimal demonstration of ATS.

II. RECOVERY OF ATS

The NV center consists of a substitutional nitrogen atom adjacent to a carbon vacancy, and the ground state exhibits zero-field splitting between the $m_s = 0$ and degenerate $m_s = ±1$ sub-levels of $D ≈ 2.87$ GHz [38, 39]. The spin-dependent photon luminescence (PL) enables the implementation of optically detected magnetic resonance (ODMR) techniques [39] to detect the spin state with normalized $PL_{m_s=0}=1$ and $PL_{m_s=±1} ≈ 0.78$ in the current experiment. With secular approximation, the effective Hamiltonian of the ground state triplet of the
NV center \([38]\),

\[
H = D S_z^2 - \gamma_e B_z S_z,
\]

is defined by the Zeeman splitting with the external magnetic field \(B_z\) along the electron spin \(S_z\), and \(\gamma_e\) is the electron gyromagnetic ratio. As a result, the \(V\)-type three-level system is formed with the states \(|0\rangle \equiv |m_s = 0\rangle, |1\rangle \equiv |m_s = -1\rangle\) and \(|2\rangle \equiv |m_s = +1\rangle\), where \(\omega_{0,1}\) is the transition frequency between \(|0\rangle\) and \(|1\rangle\) \((|2\rangle\)), as shown in Fig. 1(a).

When microwave (MW) coupling \((\omega_c)\) and probe \((\omega_p)\) fields are applied to drive the NV center, the Hamiltonian of ATS with the rotating-wave approximation is

\[
H_{ATS} = \begin{bmatrix}
0 & \Omega_c & \Omega_p \\
\Omega_c & 0 & \Delta_c \\
\Omega_p & \Delta_c & 0
\end{bmatrix},
\]

where \(\Delta_c = \omega_c - \omega_{0,1}\) \((\Delta_p = \omega_p - \omega_{0,2})\) is the frequency detuning between the coupling (probe) field and the transition between \(|0\rangle\) and \(|1\rangle\) \((|2\rangle\)). Correspondingly, \(\Omega_c\) and \(\Omega_p\) are Rabi oscillation frequencies for the coupling and probe fields, respectively. When the coupling field is resonant with its corresponding transition \((\Delta_c = 0)\) and much stronger than the probe field, the eigen-energy levels are split by \(\pm \Omega_c/2\) with eigenstates \(|+\rangle = (|0\rangle + |1\rangle)/\sqrt{2}\) and \(|-\rangle = (|0\rangle - |1\rangle)/\sqrt{2}\), as shown in Fig. 1(b). For an atomic-like system, the quantum decoherence is dominated by the longitudinal relaxation process such as spontaneous radiation. The system will be in eigenstates with long-duration driving pulses. When the frequency of the probe field is scanned, two absorption peaks can be observed after the probe field is resonant with the eigenenergy levels \([10, 11, 40]\), thus presenting ATS. This is the traditional method based on the spectral measurement. However, for single NV center in bulk diamond with the natural \(^{13}\)C isotope, the dephasing process \((\sim 10\ \mu s)\), which is caused by interaction with a nuclear spin bath \([38, 41]\), is much faster than the longitudinal depolarization process \((\sim 2\ \text{ms})\) \([42]\). Thus when the driving pulse is much longer than the dephasing time, the dephasing dominated system would be in the maximally mixed state \((|0\rangle\ (|0\rangle + |1\rangle) (|1\rangle + |2\rangle) (|2\rangle)/3\). The splitting would not be observed by scanning the probe field (see the Appendix for the theoretical calculation and simulation.).

In this experiment, to fully study the ATS with NV center, we employ the quantum state manipulation method, as shown in Fig. 2. The electronic spin qubit was initialized into the \(m_s = 0\) state by a 3 \(\mu s\) 532 nm optical pulse. At the proper magnetic field strengths \((51\ \text{mT})\), optical pumping also polarizes the \(^{14}\)N nuclear spin of the NV center into \(m_I = +1\), because resonant polarization exchanges with the electron spin in the excited state. Then we simultaneously applied coupling and probe fields with an identical duration time. Finally, the electron spin state was read with another 532 nm optical pulse. When the duration time was set to be \(t = 52.2\ \mu s\), which was much longer than the dephasing time, no splitting was observed as the probe field was scanned. As shown in Fig. 2(b), the PL was maintained at \((PL_{|0\rangle} + PL_{|1\rangle} + PL_{|2\rangle})/3 \approx 0.85\), which corresponded to the maximally mixed state. This result

\[
\begin{align*}
\omega_p & = 0.8 \\
\omega_p & = 0.9 \\
\omega_p & = 1.0
\end{align*}
\]
demonstrates that quantum dephasing has a disastrous effect on the investigation of ATS compare with the quantum longitudinal relaxation process. However, if we control the duration time of the driving fields to \( t = 1.8 \mu s = \pi / \Omega_p = 2 \pi / \Omega_c \), the doublet, which was spaced by the coupling Rabi frequency \( \Omega_c \), was observed to recover the ATS, as shown in Fig.2(b). To explain this result, we present the ATS dynamics based on Eq.(2) under the coupling field resonance with a single NV center, which is expressed as \([10, 11]\):

\[
H_d = \begin{pmatrix}
\frac{\Omega_c}{2} & 0 & \sqrt{\frac{\Omega_c}{2}} \\
0 & -\frac{\Omega_p}{2} & \sqrt{\frac{\Omega_p}{2}} \\
\sqrt{\frac{\Omega_c}{4}} & \sqrt{\frac{\Omega_p}{4}} & \Delta_p
\end{pmatrix},
\]

where \(|+\rangle, |-\rangle\) and \(|2\rangle = |2\rangle\) form the new basis vectors. When \( \Delta_p = 0 \), the effect of the probe field can be neglected due to the large detuning between \(|\pm\rangle\) and \(|2\rangle\). The spin state is in \(|0\rangle\) with the maximal PL. However, when \( \Delta_p \approx \pm \Omega_c / 2 \), we can eliminate the transition matrix element between \(|\mp\rangle\) and \(|2\rangle\) using the second order perturbation theory. Hence, the probe field will drive the system to oscillate between \(|\pm\rangle\) and \(|2\rangle\) with lower PL, which demonstrates the ATS. Theoretically, the splitting frequency from Eq.(3) is \( \Delta_{AT} = \Omega_c + \Omega_p^2 / (4 \Omega_c) \). In Fig.2(c), we independently measured the ATS as a function of the intensity of the coupling field (denoted by Rabi frequency \( \Omega_c \)). Here, the observed ATS is almost equal to the Rabi frequency of the coupling field when \( \Omega_c \gg \Omega_p \), which demonstrates the ATS characteristic. When the coupling field is not resonant, the doublet split dips are not symmetric, which can be attributed to the unbalanced superposition of \(|0\rangle\) and \(|1\rangle\), and the eigenenergies of the driven system are \( E_{\pm} = \omega_{0,2} + \Delta_p / 2 \pm \Omega_{eff} / 2 \), where \( \Omega_{eff} = \sqrt{\Delta_c^2 + \Omega_c^2} \). As shown in Fig.2(d), when \( \Delta_c \ll \Omega_c \), the positions of ATS approximately have a linear relationship with the detuning of the coupling field when the effect of the Rabi frequency \( \Omega_{eff} \) remains unchanged.

III. DYNAMICAL PROCESS OF ATS

In addition to demonstrating splitting, we can study the dynamical process of ATS in detail. In the multi-level system, besides multiple separated transitions between different two levels, the quantum interference between those transitions shows a primary difference with the two-level system. Such quantum interference has been well demonstrated in EIT and usually believed to occur only in EIT. With the quantum state manipulation method, the quantum interference was also revealed with geometric phase modulations in ATS. In the experiment, the spin is first initialized into \(|0\rangle\), which is the superposition of \(|+\rangle\) and \(|-\rangle\), i.e. \(|0\rangle = (+) + (-) / \sqrt{2}\). When the probe field is resonant with the \(|+\rangle \leftrightarrow |2\rangle\) transition by setting \( \Delta_p \approx \Omega_c / 2 \), it will flip the population between \(|+\rangle\) and \(|2\rangle\) at a frequency \( \Omega_{+,2} = \sqrt{2} \Omega_p / 2 \). When the state \(|+\rangle\) is driven by a \( 2 \pi \) probe pulse, it acquires a geometric phase \( e^{-i\pi} \) \([35, 43-45]\). Simultaneously, the state \(|-\rangle\) acquired a dynamical phase of \( e^{i\Omega_c t} \) with the coupling field. If the pulse duration also satisfies \( \Omega_c t = 2n\pi \) \((n = 1, 2, 3, \ldots)\) for \( e^{i\Omega_c t} = 1\), the spin state is at \((-|+\rangle + |-) / \sqrt{2} = -|1\rangle\). In this case, another \( 2 \pi \) probe
pulse would be required to reconver to $|0\rangle$, as shown in Fig.3(c) with the pulses sequences in Fig.3(a). Hence, the geometrical phase doubles the driving duration time and halves the transition frequency. For comparison, Fig.3(d) shows the Rabi oscillation between $|+\rangle \leftrightarrow |2\rangle$ without the coupling field. In experiment, when the electron spin state is detected by the operator $|0\rangle \langle 0|$ with the ODMR method, Fig.3(c) and (d) also show the PL of the two dynamic processes. The Rabi frequency oscillation is $\Omega_p$, whereas the ATS frequency is $\Omega_+/2 = \sqrt{2}\Omega_p/4$. This result can also be obtained by solving Eq.(3), where the population of $|0\rangle \langle 0|$ can be expressed as follows,

$$P_{|0\rangle\langle 0|} = \frac{1}{4} \left| \cos(\sqrt{2}\Omega_p t/4) + e^{i\Omega_c t} \right|^2 .$$  \hspace{1cm} (4)

This expression clearly demonstrates the quantum interference between $|+\rangle \leftrightarrow |2\rangle$ and $|-\rangle$. In the experiment to study the quantum interference with geometric phase modulations in ATS, we set $t = 2n\pi/\Omega_c$ with $\Omega_c/\Omega_p = 14$ and detected the spin-dependent PL. The experimental result is illustrated in Fig.3(e). Because of the dephasing processes, the data can be fitted by a theoretical curve $S(t) = ae^{-(t/T)k} \cos^2[w(t - t_c)] + b$, where $a = 0.211(9)$, $T = 17.5(1) \mu$s, $k = 1.5(3)$, $t_c = 7.855(5)\mu$s, $b = 0.790(3)$ and the frequency $w = 2\pi \times 0.123(2)$MHz. Correspondingly, the results of Rabi oscillation between $|+\rangle \leftrightarrow |2\rangle$ is shown in Fig.3(f) with the Rabi frequency of $\Omega_p = 2\pi \times 0.338(1)$ MHz. We can find $\Omega_p/\omega = 2.74(4) \approx 2\sqrt{2}$, which is consistent with the above theory.

IV. OPTIMAL DEMONSTRATION OF ATS.

The quantum state manipulation method is an optimal way to present the ATS with maximal signal intensity. Because of the quantum interference between $|+\rangle \leftrightarrow |2\rangle$ and $|-\rangle$, the ATS signal can be optimized when the duration time satisfies

$$\Omega_p t = 2\sqrt{2}(2k - 1)\pi, \Omega_c t = 2n\pi,$$  \hspace{1cm} (5)

or

$$\Omega_p t = 2\sqrt{2}(2k)\pi, \Omega_c t = (2n - 1)\pi,$$  \hspace{1cm} (6)

with $n, k = 1, 2, 3, \cdots$. In this case, the system remains in $|1\rangle$, the NV centre provides minimal photon luminescence, and the ATS exhibits the maximal dips. In the experiment, we set $t = 2\times 20\pi/\Omega_c \approx 2\sqrt{2}\pi/\Omega_p$, with $\Omega_c/\Omega_p = 14$. By scanning the probe field, we can obtain the result in Fig.4. The depth of the ATS is almost same as original signal in Fig.2(b), because of the quantum interference of the three-energy-level systems. In contrast, for the traditional method [5–11, 15, 16, 18, 27], the signal intensity of ATS is only half of the original signal for lacking quantum interference with the third energy level. Therefore, such an enhancement in the signal with full control of quantum state in ATS may contribute to the precision measurement of the spectrum of a quantum system.

V. DISCUSSION

In conclusion, we have presented an optimal observation method based on quantum state manipulation to study and demonstrate ATS. The ATS was recovered in a dephasing-dominated quantum system, which can not be observed with traditional observation methods. With the quantum state manipulation methods, the dynamical process of ATS was investigated in detail with a nontrivial behavior from the quantum interference with geometric phase modulations. Consequently, the ATS was optimally demonstrated, and its signal intensity was twice those of other systems observed with the traditional observation method. The study presents a feasible method to optimally observe the atom-light interaction in a multi-level system, which can be applied to investigate quantum optics and atomic physics for a broad applications in high-dimensional quantum control and quantum error correction beyond the dynamically decoupling, decoherence-free subspace.

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Appendix A: Experimental setup and work point of the quantum system

1. Experimental setup

As shown in Fig.5, the NV center was located and detected with a home-built confocal microscopy with a dry objective lens (N.A. = 0.95) at room temperature. The power of 532 nm continuous laser was set at 0.6 mW. The NV center fluorescence was separated from the excitation laser with a 647 nm long pass filter and then detected by single photon counting modules. We constructed two synchronized microwaves that drove the NV center system with two different frequencies. The microwave was coupled to the sample by a coplanar waveguide.

Single photon emission from a single NV center was verified by measuring the photon correlation function $g^2(\tau)$ as shown in Fig.6(b). And $g^2(\tau) < 0.5$ indicates a single NV center. To form a simple V-type three-level system, a magnetic field of 51 mT was applied along the NV axis using a permanent magnet. Under this condition, the flip-flop process between electron-spin and nuclear-spin during optical pumping [39] leads to polarizing the nitrogen nuclear spin of NV center after 3 $\mu$s.

FIG. 6. (a) Confocal image of the NV center used in the experiment with a signal-to-noise ratio of 140:1. A coplanar waveguide antenna was deposited to deliver microwave pulses to the NV center. (b) Fluorescence correlation function. (c)-(d) ODMR spectra for the single NV center. The transition frequencies for $|0\rangle \rightarrow |1\rangle$ and $|0\rangle \rightarrow |2\rangle$ are $\omega_{0,1} = 1.43398(1)$ GHz and $\omega_{0,1} = 4.30738(1)$ GHz, respectively.

2. Decoherence time

For single NV center, the electron spin states dephasing [28, 32, 38, 42] is the main part of quantum decoherence and has an impact on the ATS. The dephasing time of NV center was measured by the Ramsey interferometer [38, 42]. By fitting experimental data as shown in Fig.S3(a), we got $T_{2,0+1} = 8.2(3)$ $\mu$s and $T_{2,0+2} = 8.7(4)$ $\mu$s.

After transforming to a frame co-rotating with the two driving fields with $U_0 = e^{iH_0t}$ with $H_0 = \omega_0 |1\rangle \langle 1| + \omega_2 |2\rangle \langle 2|$, we get

$$ H = \begin{pmatrix} 0 & \Omega_{0} e^{-i\varphi_c} & \Omega_{2} e^{-i\varphi_p} \\ \Omega_{0} e^{i\varphi_c} & 0 & \Delta_c \\ \Omega_{2} e^{i\varphi_p} & \Delta_c & 0 \end{pmatrix}. $$

Appendix B: Theoretical model of the ATS with quantum state manipulation

The Hamiltonian for NV center under driving fields in experiment reads

$$ H = \omega_{0,1} |1\rangle \langle 1| + \omega_{0,1} |2\rangle \langle 2| + \Omega_c \cos(\omega_c t + \varphi_c) |1\rangle \langle 0| + |0\rangle \langle 1| + \Omega_p \cos(\omega_p t + \varphi_p) |2\rangle \langle 0| + |0\rangle \langle 2|. $$

where $\varphi_c$ and $\varphi_p$ are the initial phases of the coupling and probe fields, respectively.
FIG. 7. (a) Result of the Ramsey experiment for the electron spin of NV center. Black squares and red circles correspond to $|0\rangle \leftrightarrow |1\rangle$ and $|0\rangle \leftrightarrow |2\rangle$, respectively. Experimental data are fitted by $y(t) = a\exp\left(-\left(t/T_{2\text{d}}\right)^2\right)\cos(2\pi\omega t) + b$ and denoted with solid blue curves. The dephasing time of NV center was measured to be $T_{2\text{d}} = 8.2(3)$ $\mu$s and $T_{2\text{d}} = 8.7(4)$ $\mu$s. (b)-(c) Rabi oscillation of the electron spin between ground state sublevels of NV center. The experimental data (red dots) was fitted by a damped sine function (blue curves) written as $y(t) = a\exp\left(-\left(t/T_{1}\right)^2\right)\cos(\pi\frac{t}{T_{1}}/\omega) + b$. We can get $T_{1,\rho} = 25.0(8)$ $\mu$s and $T_{1} = 1.7(2)$ ms. The optimal demonstration of ATS in main text Fig.3a-d and $\Omega_{c}/\Omega_{p} = 14$.

FIG. 8. (a)-(b) Rabi oscillation of the coupling and probe field for investigating dynamical process of ATS in main text omitting the initial phases.

which means that the arbitrary initial phases of driving fields do not have any effect and can be neglected in the experiment.

2. The optimal demonstration of ATS

Now we chose the coupling field resonant with the single NV center. And in the dressed-state picture, Eq.(B2) (omitting the initial phases) can be expressed as

$$H_d = \begin{pmatrix} \Omega_c/2 & 0 & \sqrt{2}T_{1p}/4 \\ 0 & -\Omega_c/2 & \sqrt{2}T_{1p}/4 \\ \sqrt{2}T_{1p}/4 & \sqrt{2}T_{1p}/4 & -\Delta_p \end{pmatrix},$$

(B6)

where $|+\rangle = \frac{|0\rangle + |1\rangle}{\sqrt{2}}$, $|-\rangle = \frac{|0\rangle - |1\rangle}{\sqrt{2}}$ and $|2\rangle = |2\rangle$ form new basis vectors. The frequency of the probe field was scanned to make sure that the Rabi frequency of probe field is 1/14 of that of the coupling field, as shown in Fig.8(a)-(b).

Case I: $\Delta_p = 0$, due to the large detuning between energy level of the NV center, the effect of the probe field can be neglected. The spin state is in bright state $|0\rangle$ when $\Omega_c t = 2n\pi, n = 1, 2, 3\cdots$, as shown in Fig.2(b) in the main text.

Case II: $\Delta_p \approx \Omega_c/2$, the middle energy level effect can be eliminated by the second order perturbation theory with

$$H_d = \begin{pmatrix} \Omega_c/2 & 0 & \sqrt{2}T_{1p}/4 \\ 0 & -\Omega_c/2 & \sqrt{2}T_{1p}/4 \\ \sqrt{2}T_{1p}/4 & \sqrt{2}T_{1p}/4 & -\Delta_p + \Omega_c^2/8\Delta_p \end{pmatrix},$$

(B7)

By substituting Eq.(B2) and Eq.(B4) to Eq.(B3), we get

$$P_{|0\rangle\langle 0|} = \left|\langle 0|e^{-i\bar{H}t}|0\rangle\right|^2.$$  

(B5)
and once \( \Delta_p = \frac{\Omega_p}{2} - \frac{\Omega_p^2}{4\Omega_c}, \) we have

\[
P_{\{\Delta\} \{\phi\}} = \left| \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 & 0 \end{pmatrix} e^{-iH_{\text{det}} t} \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ 1 \\ 0 \end{pmatrix} \right|^2
\]

\[
= \left| \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 & 0 \end{pmatrix} \right|^2 \left( \frac{\cos \frac{\sqrt{2}\Omega_p t}{e^{\alpha t}}}{\sqrt{2}} - i \sin \frac{\sqrt{2}\Omega_p t}{e^{\alpha t}} \right)^2
\]

\[
= \frac{1}{4} \cos \frac{\sqrt{2}\Omega_p t}{4} + e^{it} \left| \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 & 0 \end{pmatrix} \right|^2.
\]

To get the highest contrast for the ATS, the conditions are:

\[
\Omega_{p}\bar{c} = 2\sqrt{2}(2k - 1)\pi, \quad \Omega_{c}\bar{t} = 2n\pi,
\]

or

\[
\Omega_{p}\bar{c} = 2\sqrt{2}(2k + 1)\pi, \quad \Omega_{c}\bar{t} = (2n - 1)\pi,
\]

where \( n, k = 1, 2, 3 \ldots \)

Case III: \( \Delta_p \approx -\frac{\Omega_p}{2} \), the situation is similar to Case II. So the conditions for the observation of the highest contrast is same and the resonance frequency of the probe field is \( \Delta_p = -\frac{\Omega_p}{2} + \frac{\Omega_p^2}{4\Omega_c}. \)

3. The ATS with non-resonant driving fields

For \( \Omega_c \gg \Omega_p, \) the ATS can be expressed as

\[
\Delta_{\text{ATS}} \approx \Omega_c - \frac{\Omega_p^2}{4\Omega_c} \approx \Omega_c.
\]  \hspace{1cm} (B11)

If the coupling field is not resonant, we have

\[
\hat{H} = \frac{\Delta_c + \Omega_{c\ell}}{2} \hat{\rho} + \frac{\Omega_{c\ell}}{2\sqrt{2} \rho_{c\ell} + 2\Delta_c \rho_{c\ell}} |+\rangle \langle +| + \frac{\Omega_{c\ell}}{2\sqrt{2} \rho_{c\ell} - 2\Delta_c \rho_{c\ell}} \hat{\rho} |\rangle \langle \rangle + \frac{\Omega_{c\ell} \rho_{c\ell}}{2} |\rangle \langle \rangle + \frac{\Omega_{c\ell} \rho_{c\ell}}{2} |\rangle \langle \rangle
\]

in the bases of \(|+\rangle = \frac{\Omega_{c\ell}}{\sqrt{2\Omega_{c\ell} + 2\Delta_c + \sqrt{2\Omega_{c\ell} + 2\Delta_c}} |0\rangle + \frac{\Delta_c + \sqrt{2\Omega_{c\ell} + 2\Delta_c}}{\sqrt{2\Omega_{c\ell} + 2\Delta_c - \sqrt{2\Omega_{c\ell} + 2\Delta_c}} |1\rangle \) and \(|-\rangle = \frac{\Omega_{c\ell}}{\sqrt{2\Omega_{c\ell} + 2\Delta_c + \sqrt{2\Omega_{c\ell} + 2\Delta_c}} |0\rangle + \frac{\Delta_c - \sqrt{2\Omega_{c\ell} + 2\Delta_c}}{\sqrt{2\Omega_{c\ell} + 2\Delta_c - \sqrt{2\Omega_{c\ell} + 2\Delta_c}} |1\rangle. \)

\[ \Delta_c = \frac{\Omega_{c\ell}}{2\Omega_{c\ell} + 2\Delta_c - \sqrt{2\Omega_{c\ell} + 2\Delta_c}} \]

Hence, when the two resonant transitions have non-zero detuning, the splitting exhibits asymmetric ATS (unequal transmission dips) and \( \Delta_{\text{ATS}} \approx \Omega_{c\ell} = \sqrt{\Omega_c^2 + \Delta_c^2}. \)

4. The failure in the observation of ATS in a dephasing dominated system with traditional method

The traditional method to observe the ATS is based on the distribution of the population of static state under long-pulse driving fields [5–11, 15, 16, 18, 27], which can not be applied to demonstrate the ATS in the dephasing dominated system. Here, we employ Lindblad equation for the steady-state solution with

\[
\rho^l = -i[H_I, \rho^l] + \sum_j D(A_j^l)\rho^l,
\]

where \( D(A_j^l)\rho^l = A\rho^l A^\dagger - \{ A^\dagger A, \rho^l \} /2 \) and \( H_I = \hat{H}. \)

The longitudinal relaxation from \( i \) to \( j \) can be written as \( A_{\text{diss}} = \sqrt{\Gamma_{ij}} |i\rangle \langle j| \) and the dephasing process for state \( i \) is \( A_{de} = \sqrt{2\gamma_i} |a\rangle \langle a|. \)

For the dephasing channels,

\[
D(A_{de})\rho = \begin{bmatrix} 0 & -\gamma_{10} & -\gamma_{20} \\ -\gamma_{10} & 0 & -\gamma_{30} \\ -\gamma_{20} & -\gamma_{30} & 0 \end{bmatrix}.
\]

Just letting \( \rho_{ij} \rightarrow \rho^l_{ij} \) for the expression of the dephasing process, we can transform the lab frame to the rotation frame.

In the NV center system, the dephasing process is much faster than the longitudinal relaxation(\( \gamma \gg \Gamma \)). So we can omit the longitudinal relaxation and get equation for the steady state after long-pulse driving,

\[
0 = -i[H_I, \rho^l] + D(A_{de})\rho^l.
\]

Hence, \( \rho^l_{12} = \rho^l_{01} = \rho^l_{02} = 0 \). At last,

\[
\rho^l_{00} = \rho^l_{11} = \rho^l_{22} = \frac{1}{3},
\]

which means that the ATS or EIT cannot be observed with the traditional observation method. And it also holds for cascade and A EIT cannot be observed based on the traditional observation method in the dephasing dominated quantum decoherence system.

5. The dynamical process of ATS and simulation

The dynamical process of ATS in NV center can also be numerically simulated with the Lindblad equation. The
independent equations of Eq. (B13) are

\[
\begin{align*}
\dot{\rho}_{00}^l &= -i \left[ \frac{\Omega_c}{2} (\rho_{10}^l - \rho_{01}^l) + \frac{\Omega_p}{2} (\rho_{20}^l - \rho_{02}^l) \right], \\
\dot{\rho}_{01}^l &= -i \left[ \frac{\Omega_c}{2} (\rho_{11}^l - \rho_{00}^l) + \frac{\Omega_p}{2} \rho_{21}^l \right] - \gamma_1 \rho_{01}^l, \\
\dot{\rho}_{02}^l &= -i \left[ \frac{\Omega_c}{2} \rho_{12}^l + \frac{\Omega_p}{2} (\rho_{22}^l - \rho_{00}^l) - \Delta_p \rho_{02}^l \right] - \gamma_2 \rho_{02}^l, \\
\dot{\rho}_{11}^l &= -i \left[ \frac{\Omega_c}{2} (\rho_{01}^l - \rho_{10}^l) \right], \\
\dot{\rho}_{12}^l &= -i \left[ \frac{\Omega_c}{2} \rho_{02}^l - \frac{\Omega_p}{2} \rho_{10}^l - \Delta_p \rho_{12}^l \right] - \gamma_3 \rho_{12}^l, \\
1 &= \rho_{00}^l + \rho_{11}^l + \rho_{22}^l. \\
\end{align*}
\]

(B17)

The last equation is the additional constraint of completeness. Just letting

\[
\begin{align*}
\rho_{00}^l &= y_1, \\
\rho_{11}^l &= y_2, \\
\rho_{01}^l &= y_3 + iy_4, \\
\rho_{02}^l &= y_5 + iy_6, \\
\rho_{12}^l &= y_7 + iy_8, \\
\end{align*}
\]

(B18)

we can convert the physical equation to the linear ordinary differential equations and solve them with the Runge-Kutta method. The fluorescence intensity of the NV center is given by \(I = 1 - C + Cy_t\) with \(C = PL_{m_z=0} - PL_{m_z=\pm1} = 0.22\) is fluorescence contrast for different spin states [39]. Fig. 9(a) shows the result of the time dependence of the probabilities for a particular set of conditions beginning with \(|0\rangle\). When the duration time of both driving fields is larger than the dephasing time, the system will become a maximally mixed state.

If the duration time of the driving fields satisfies \(\Omega_c t = 2n\pi\ (n = 1, 2, 3, \ldots)\), the envelope line will be obtained as shown in Fig. 9(b). There is a little discrepancy between the theory and experimental result as shown in Fig. 3(c) in the main text. The most important factors causing the deviation would be the environment treatment of NV center. For the present sample, the decoherence of NV center is dominated by the hyperfine interaction with the \(^{13}\text{C}\) nuclear spins, which form a nuclear spin bath. The bath spins involved in the decoherence of NV center is much more complicated than those in quantum dots and shallow donors [46].

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