1. ESTIMATION OF THE ELECTRON SPIN $T_2^*$ FROM THE 2-BEAM MEASUREMENTS

Figure S1 shows the probe absorption spectra at a fast scan rate with various pump intensities. These spectra are clearly different from the standard dark state spectrum because of the nuclear field dynamics. By the self-locking DNP effect, the nuclear magnetic field changes to adjust the trion resonance as the probe scans so that the probe absorption is maximized. The instantaneous nuclear field is a chirped function of the probe scan frequency (see, for example, Fig. 4(e) in the main text). Because of the frequency chirp, the trion transition linewidth is broadened as compared to the intrinsic linewidth. While the broadening is larger at higher pump intensities, the maximum absorption signal shows no reduction with the increase of the linewidth. This distinguishes the DNP induced broadening from both homogeneous and inhomogeneous broadenings, since the latter two are always accompanied by a reduction in the strength of the optical response. In addition, the frequency chirping also makes the dark state dip appear to be much narrower than expected for a standard dark state dip at a given pump intensity. The narrowing of the dip becomes extremely pronounced in the slow scan spectrum where DNP becomes most efficient (see Fig. 1 and Fig. 4 in the main text).

To get information on the spin $T_2^*$ from these complex spectra, we first take a look at the ordinary dark state spectrum by solving the standard two-beam optical Bloch equations for a three level Lambda system, in the absence of any nuclear field dynamics. When the pump Rabi is much larger than the probe, we get a simplified formula of the probe absorption coefficient by taking all orders of the pump and the first order of the probe

$$\alpha = \alpha_o \frac{X^2 \gamma_s + \gamma_t (\gamma_t^2 + (\delta_1 - \delta_2)^2)}{X^4 + (\gamma_t^2 + \delta_1^2)(\gamma_s^2 + (\delta_1 - \delta_2)^2) + 2X^2(\gamma_t \gamma_s + \delta_1(\delta_2 - \delta_1))} \quad (1)$$

where $\alpha_o$ is a coefficient, $X = \Omega_{pump}/2$, $\gamma_s$ is the electron spin decoherence rate, $\delta_1$ ($\delta_2$) is the probe (pump) detuning to the trion transitions. The trion decoherence rate $\gamma_t$ is determined predominantly by radiation broadening and is about 400 MHz, with less than 10% fluctuations due to the spectral diffusion in this dot. In this formula, we ignore the contribution of electron spin relaxation rate since it is much smaller than $\gamma_s$.

In the presence of a finite nuclear field, $\delta_2 = \omega_{pump} - \omega_{t,+} + \Delta/2$ and $\delta_1 = \omega_{probe} - \omega_{t,-} - \Delta/2$, where $\Delta/2$ is the Overhauser shift of the electron spin states, $\omega_{t,\pm}$ are the two transition resonances in absence of nuclear field, and $\omega_{pump}$ and $\omega_{probe}$ are the two laser frequencies. Assuming $\omega_{t,+} = \omega_{pump}$, we have $\delta_1 - \delta_2 = \delta \omega_p - \Delta$ where $\delta \omega_p \equiv \omega_{probe} - \omega_{t,-}$ is the probe detuning. By the self-
locking DNP effect, accompanying the probe frequency scan, the nuclear field becomes a function of the probe detuning $\Delta(\delta\omega_p)$ (see, for example, Fig. 4(e) in the main text).

Taking into account the nuclear field dynamics, the absorption spectrum is

$$
\alpha = \alpha_o \frac{X^2\gamma_s + \gamma_t(\gamma_s^2 + (\delta\omega_p - \Delta)^2)}{X^4 + 2X^2\gamma_t\gamma_s + (\gamma_s^2 + (\delta\omega_p - \Delta/2)^2)(\gamma_s^2 + (\delta\omega_p - \Delta)^2) - 2X^2(\delta\omega_p - \Delta/2)(\delta\omega_p - \Delta)}
$$

(2)

Since the linewidth broadening of the probe spectrum has been accounted by the nuclear field chirp function $\Delta(\delta\omega_p)$, the trion decoherence rate $\gamma_t$ in the above equation shall always be taken as the intrinsic value of 400 MHz throughout all the data analysis below.

A. Estimation of the electron spin $T_2^*$ from dip-to-peak absorption ratio

From Eq.(2), the minimum absorption at the dark state dip (see Fig. S1) is given by evaluation at $\delta\omega_p = \Delta$,

$$
\alpha(\text{dip}) = \alpha_o \frac{X^2\gamma_s + \gamma_t(\gamma_s^2)}{X^4 + 2X^2\gamma_t\gamma_s + \gamma_s^2 \gamma_t^2} 
$$

(3)

while the maximum absorption at the trion peaks on both sides of the dip (see Fig. S1) is given by evaluation at $\delta\omega_p \approx \Delta \pm X$,

$$
\alpha(\text{peak}) = \alpha_o \frac{X^2\gamma_s + \gamma_t(\gamma_s^2 + X^2)}{2X^2\gamma_t\gamma_s + \gamma_t^2\gamma_s^2 + (\gamma_t^2 + \gamma_s^2)X^2} 
$$

(4)

We see clearly from Eq. (3) and (4) that the dip-to-peak absorption ratio only depends on the three parameters $X$, $\gamma_t$, and $\gamma_s$. When $X >> \gamma_t >> \gamma_s$, Eq. (3) and (4) can be simplified to $\sim \alpha_o \frac{\gamma_s}{X^2}$ and $\sim \alpha_o \frac{1}{\gamma_t}$, respectively. Thus, the dip-to-peak absorption ratio can provide a reliable estimation on the value of $\gamma_s$, as shown by the black dots in Fig. 3a in the main text. The corresponding error bar comes from the uncertainties in determining $\alpha_{\text{peak}}$ and $\alpha_{\text{dip}}$ because of the data fluctuations, which is the measurement noise measured independently under the same experimental conditions.

We would like to remark that this dip-to-peak ratio provides only a lower bound estimation of spin $T_2^*$ in several aspects. First, we note that $\alpha(\text{dip})$, only reflects the $\gamma_s$ at the dark state, while $\alpha(\text{peak})$ is largely independent of the value of $\gamma_s$. Thus, the estimate from their ratio only reflects the $T_2^*$ right at the dark state where nuclear field locking is not taking effect, since the two-photon Raman Resonance (TPR) is a metastable configuration for the DNP. At the trion absorption peak where nuclear field locking is the most effective, the spin $T_2^*$ is expected to be much longer. Second, unaccounted homogeneous or inhomogeneous broadening mechanisms, if any,
FIG. 1: (color online) Dark state spectra from 2-beam measurements with various pump intensities. The corresponding Rabi frequencies (marked on each spectrum) are estimated by comparing the intensities with that of Fig. 3c in the main text in which a Rabi frequency of 0.9 GHz can be determined from the fit. The red curves on top of the data are the theoretical fits assuming the nuclear chirp functions shown on the right. The spin decoherence rates from the fitting are respectively: \( \gamma_s/2\pi = 60\text{MHz}, 90\text{MHz}, 150\text{MHz}, 185\text{MHz}, \) and 260MHz (from top to bottom), while the pump detuning is fixed at zero and the trion decoherence rate is fixed at 400MHz throughout. For each spectrum, a green curve is also shown, which is the plot assuming the same nuclear chirp function, pump detuning, and trion linewidth as used for the red curve, but with spin decoherence rate held at the thermal value \( \gamma_s/2\pi = 360\text{MHz} \). The minimum absorption at the dip \( \alpha_{\text{dip}} \) and the maximum absorption at the trion peak \( \alpha_{\text{peak}} \) are also illustrated in the figure.

could only make the actual absorption at the trion peak smaller than the prediction of Eq.(4), and make the actual absorption at the dark state dip larger than the prediction of Eq.(3). The effects of these unaccounted broadenings on increasing the dip-to-peak ratio could have been “mistakenly” counted as part of spin decoherence. This will always result in an underestimate of spin \( T_2^* \). The last cause of underestimation comes from the round off effect on the dark state dip due to the lack
of resolution in the measurement. Theoretically, the DNP effect on the dark state dip is to make it narrower but not shallower, since the absorption calculated in Eq.(3) is independent of the nuclear field chirp function one the TPR. In reality, the sharp and narrow dip will inevitably be rounded off by the lack of resolution in the laboratory due to the interlaser jitter and finite step size for the laser scanning. (See section I B for more details). This round off effect also increases the dip-to-peak ratio, and leads to some underestimation of $T_2^*$ as we will see in I B.

Nevertheless, even from these underestimated values of $T_2^*$, the trend that $T_2^*$ increases with the increasing pump intensity is clearly shown (see Fig. 3a in main text). The overestimated spin decoherence rate $\gamma_s$ is well below the thermal value of $\sim 360$ MHz.

**B. Fitting by modeling of the nuclear field chirp function**

We also fit the complex lineshapes in Fig. S1 by modeling the nuclear field chirp function $\Delta(\delta\omega_p)$ based on theory. From Fig. 4 in the main text, we see that the simulation based on the DNP equation (see section II for details) reproduces the key features of the spectrum, and shows the nuclear field as a chirped function of probe frequency (Fig. 4(e)). However, the theoretical model is not sufficiently complete to quantitatively match all details of the spectrum. Therefore, we approach the fitting of the data by modeling the chirp function within the qualitative framework that can be determined from the self-locking features of the DNP effect.

From Fig. 1 and Fig. 4 in the main text, we see that when the laser frequency scans towards a trion maximum, DNP shifts the maximum to meet the laser frequency, leading to the rising edge in the absorption spectrum. As the probe laser tries to move past the trion maximum, DNP shifts the trion resonances to follow the laser scan to maintain the maximum probe absorption, which explains the broadened and rounded top of the trion peak. After a sufficiently large nuclear spin polarization has been established, as the laser scans away from the absorption maximum, nuclear spin relaxation will dominate the nuclear field dynamics. At this point, the trion maximum can no longer follow the laser frequency and instead rebounds to its equilibrium position, moving away from the probe laser frequency. This leads to falling edges in the spectrum.

These qualitative behavior from the self-locking feature largely determines the shape of the nuclear chirp function, as schematically illustrated in Fig. S2 where the scan is going from negative to positive $\delta\omega_p$. First, the two red sections of the chirp function stay on the two lines $\Delta = \delta\omega_p - \frac{1}{2}\Omega_{\text{pump}}$ and $\Delta = \delta\omega_p + \frac{1}{2}\Omega_{\text{pump}}$, which are the locking positions for the system to maintain the
FIG. 2: DNP induced spectrum broadening due to nuclear field chirping

maximum trion excitation. The lengths of these two sections correspond to the broadening of the two trion peaks. Second, the horizontal distance between the two turning points L and R (black dots) are equal to the width of the dark state dip. Third, the three blue sections of the chirp function correspond to the rising and falling edges. The nuclear field build-up process accelerates as the probe frequency approaches the turning points (A and R in the figure), and also in leaving the turning points (L and B). So the three blue sections qualitatively look like the arcs shown in Fig. S2. These arcs are steeper in slower scans: for $\delta \omega_p$ to change by the same amount it takes a longer time during which a larger nuclear field shift can accumulate (see Fig. 4(e) in the main text). The intersection point (red dot) of the middle blue curve with the line $\Delta = \delta \omega_p$ corresponds to the tip of the dark state dip.

With properly chosen nuclear chirp functions based on the above consideration, the theoretical plots using Eq.(2) qualitatively match the measured spectra at different pump intensities (see red curves in Fig. S1). The fitting takes into account the finite resolution effect in the laboratory. Specifically, the probe frequency is continuously changing at the rate of 4 MHz per millisecond. For every 10 ms interval, data collection lasts for 3 ms and the remaining 7 ms is a waiting time. This effectively corresponds to a 40MHz resolution in the measured spectrum. The theoretical curves (red) are obtained by coarse-graining Eq.(2) with this resolution. The extracted spin $T_2^*$ values from these fittings are shown in Fig. 3a in the main text, which are in quantitative agreement
with the estimations based on dip-to-peak ratio. The spin $T_2^*$ values obtained from these fittings are also the lower bound estimation, limited by the same facts as in the dip-to-peak ratio estimation.

The green curves are the plots of Eq.(2) assuming the same chirp functions, but holding the spin decoherence rate $\gamma_s/2\pi$ at the thermal value of 360 MHz. Clearly, the increase of $T_2^*$ has resulted in a qualitative difference in the probe spectrum at high pump intensities.

II. THEORY

In section II A, we review the hyperfine coupling between the nuclear spin and the heavy-hole spin. In section II B and II C, we show that the nuclear field self-locking effect observed in the experiments (see main text) turns out to be a natural consequence of the dynamic nuclear polarization (DNP) by the hole spin constitution of the trion, which is resonantly excited in the intermediate steps. By running multiple simulations, we can find a best match between the simulated and experimental spectra for both fast and slow scans. DNP rates extracted from the best matched simulation agree reasonably well with the microscopic calculated rates. In section II G, we investigate the possible DNP effects caused by the electron spin and show that they can not give rise to the observed self-focusing DNP effect for the current experimental configuration.

A. Anisotropic Hyperfine interaction between hole spin and nuclear spins

The complete hyperfine interaction between an electron spin and nuclear spins is,

$$H_{en} = -\frac{\mu_0}{4\pi}\gamma_N\gamma_e \sum_k \hat{I}_k \cdot \left[ \frac{8\pi}{3} \delta (r_k) - \frac{r_k^2 \hat{S} - 3r_k \left( \hat{S} \cdot r_k \right)}{r_k^5} + \frac{\hat{L}}{r_k^3} \right]$$

(5)

where $\gamma_e$ and $\gamma_N$ are the electron and nuclear gyromagnetic ratio respectively. $r_k$ is the electron coordinate measured from the $k$th nuclear spin. $\hat{S}$ and $\hat{L}$ are the spin and angular momentum of the electron. The three terms are respectively the isotropic Fermi contact hyperfine interaction, the anisotropic dipole-dipole like interaction, and the coupling of electron orbital angular momentum to the nuclear spin.

It can be shown that for $s$-type Bloch states with slowly-varying envelope functions, the dipolar hyperfine interaction is much weaker than the contact Fermi interaction. Therefore, the hyperfine interaction between the conduction band electron with nuclear spins is dominated by the isotropic
contact part

\[ H_{e-n} = \sum_k A_{e,k} (S_x^e I_x^k + S_y^e I_y^k + S_z^e I_z^k), \]

where \( A_{e,k} \equiv A_{e,\alpha_k} \mid f_e (R_k) \mid^2 c_0^3 / 4 \). \( f_e (R_k) \) is the normalized envelope function of the quantum dot and \( c_0 \) is the lattice constant. \( A_{e,\alpha} \) is the hyperfine constant of the material: \( A_{e,\text{As}} = 46 \mu \text{eV} \) and \( A_{e,\text{In}} = 56 \mu \text{eV} \). Note that each primitive unit cell of volume \( c_0^3 / 4 \) contains one \( \text{In} \) nuclear spin and one \( \text{As} \) nuclear spin. The total number of nuclear spins \( N \) is proportional to the quantum dot volume \( V \): \( N = 8Vc_0^{-3} \). The coupling strength of the electron with an individual nuclear spin is on the order of \( \frac{2}{N} A_{e,\alpha} \), inversely proportional to the quantum dot size.

For holes in the valance band, the contact hyperfine interaction vanishes for \( p \)-type Bloch states, and the hole-nuclear spin interaction is dominated by the other two terms. Strictly speaking, the nuclei not only interact with the fraction of the hole in the same primitive unit cell (‘on-site’ interaction), but also with the probability density localized at more distant unit cells (long-ranged interactions). Nevertheless, it has been shown that the long-range part only leads to corrections on the order of 1% relative to the on-site interaction, and hence can be neglected\(^2\)\(^7\). In the present study, we are interested in the hyperfine interaction between the heavy hole and nuclear spins. We use \( \frac{1}{2} \) pseudo-spin \( S_h \) to denote the heavy hole subspace.

For a small quantum dot, heavy-light hole mixing is a non-negligible effect. The two ‘heavy hole’ states after mixing are \( |S_z^h = \frac{1}{2} \rangle \equiv |J_z = \frac{3}{2} \rangle - \eta |J_z = -\frac{1}{2} \rangle \) and \( |S_z^h = -\frac{1}{2} \rangle \equiv |J_z = -\frac{3}{2} \rangle - \eta |J_z = \frac{1}{2} \rangle \), where \( \hat{z} \) is assumed to be the growth direction. The amount of mixing \( \eta \) is \( \sim 0.2 \) for the self-assembled quantum dot used in the experiment, which is identified using polarization dependent absorption spectroscopy\(^8\). Assuming that the hole envelope function \( f_h (R_k) \) varies slowly on the length scale of a primitive cell, when the time reversal symmetry is broken by the magnetic field, one finds that the hyperfine interaction between the heavy hole spin and the nuclear spins is\(^2\)\(^7\),

\[ H_{h-n} = \sum_k A_{h,k} \left[ S_h^x I_x^k + O (\eta) (S_h^y I_y^k + S_h^z I_z^k) + O (\eta^2) (S_h^y I_y^k + S_h^z I_z^k) \right] \]

where \( A_{h,k} \equiv A_{h,\alpha_k} \mid f_h (R_k) \mid^2 c_0^3 / 4 \). We have \( A_{h,k} \sim \frac{2}{N} A_{h,\alpha_k} \), which is similar to the electron case. For \( \text{In} \) and \( \text{As} \), experiments and calculations lead to the estimation \( A_h \sim 0.1 - 0.2 A_e \)\(^2\)\(^7\).

Unlike the electron, the hole spin couples to the nuclear spins through the anisotropic hyperfine interaction. This results in significantly different dynamic polarization effect on the nuclear spins. In a magnetic field applied in the \( \hat{x} \) direction, we define the nuclear spin raising and lowering
operators $I^\pm_k = I^y_k \pm iI^z_k$. For the electron, the terms that flip the nuclear spin eigenstates in the field direction are $S^e I^\pm_k$, which always involve the simultaneous flip of the electron spin eigenstate. For the hole, we have unique terms like $S^h I^\pm_k$, which flip the nuclear spin eigenstates without changing the hole spin. This type of processes is of direct relevance for the dynamics of the nuclear Overhauser shift observed in this experiment. If the magnetic field is entirely in the $x-y$ plane, such processes arise from the $O(\eta^2)(S^z_h I^z_k + S^y_h I^z_k)$ term in Eq. (7). If the magnetic field has some out-of-plane component, such processes can also arise from the $S^z_h I^z_k$ term in Eq. (7).

**B. Dynamic Nuclear Polarization by resonantly excited hole spin in a laser driven $\Lambda$ system**

The relevant states in the quantum dot form a three-level $\Lambda$ system: two single electron spin states and a trion state which is composed of an electron spin singlet and a heavy hole spin aligned along the magnetic field direction $\hat{x}$ (see Fig.1A in the main text). The two transitions $H1$ and $V2$ are coupled respectively to the pump and probe lasers. For the trion state, the electron spin do not couple to the nuclear spins since they are in a singlet configuration, but the nuclear spins feel the hyperfine interaction from the heavy hole constitution in the form of Eq. (7).

The role of the heavy hole spin in DNP stands out as its hyperfine interaction with the nuclear spins has terms like $S^x_h I^\pm_k$ which flip the nuclear spin without changing the hole spin state. The energy cost for such processes is only the nuclear Zeeman energy $\hbar \omega_N$ which is $\sim 0.01$ GHz in an external magnetic field of 1 Tesla. Since the trion state has a homogeneous broadening $\sim 0.1 - 1$ GHz, the energy cost for flipping a nuclear spin can be compensated by the energy uncertainty of the trion state. Therefore, the hole-nuclear hyperfine coupling $S^x_h I^\pm_k$ can flip the nuclear spin through a first order process.

We note that a more extensively investigated mechanism for DNP effects is the electron-nuclear flip-flop hyperfine coupling $S^\pm_e I^\mp_k$. To flip the nuclear spin by such hyperfine couplings, phonon-assistance or photon-assistance is needed to compensate for the energy change for the simultaneous flip of the electron. The phonon-assisted electron-nuclear flip-flop is at least a second order process involving a coupling matrix element between electron spin and phonon which is generally weak\(^5,6\). The photon-assisted electron-nuclear flip-flop is at least a third order process involving two optical coupling matrix elements (see section II.G). While the strength of the hole-nuclear hyperfine coupling term $S^z_h I^z_k$ is much weaker as compared to the electron-nuclear flip-flop coupling term $S^\pm_e I^\mp_k$, its role may not be any less significant since it is a lower order process. As we show below
and in section II C, the nuclear field self-focusing effect observed in the experiments turns out to be a natural consequence of the DNP by the hole spin which is resonantly excited in the intermediate steps.

We assume that the three-level $\Lambda$ system is always in the instantaneous steady-state, determined by the instantaneous value of the laser frequencies and the nuclear Overhauser field. This is because the optical interaction is many orders faster than the nuclear spin processes and the scan of the laser frequencies. We define the instantaneous steady-state being $|\Psi_i\rangle$ ($|\Psi_f\rangle$) before (after) a nuclear spin is flipped by the $S_k^z I_k^z$ term. The rate of this flip transition can be estimated using the Fermi’s Golden rule,

$$r_{D,k}^\pm \simeq \frac{2\pi}{\hbar} |\langle\Psi_f| O(\eta^2) A_{h,k} S_k^z I_k^z |\Psi_i\rangle|^2 D\pm \hbar \omega_N \big),$$

$$= \frac{2\pi}{\hbar} O(\eta^4) A_{h,k}^2 |\langle\Psi_f| S_k^z |\Psi_i\rangle|^2 |\langle\Psi_f| I_k^z |\Psi_i\rangle|^2 D\pm \hbar \omega_N \big),$$

$$= \frac{\pi}{2} \rho_{t,i} \rho_{t,f} O(\eta^4) A_{h,k}^2 |j \pm m_k(j \mp m_k + 1)D(\pm \hbar \omega_N \big) \big),$$

(8)

where $\Psi_e$ ($\Psi_N$) is the electronic (nuclear) part of the wavefunction and $\rho_{t,i}$ ($\rho_{t,f}$) is the trion population in the initial (final) steady-state configuration. $m_k$ is the eigenvalue of $I_k^z$ of the initial nuclear state. $j = 3/2$ for As and $j = 9/2$ for In. We have used $r_{D}^+$ ($r_{D}^-$) to denote the rate nuclear spin $k$ is flipping up (down) and $D(\pm \hbar \omega_N \big)$ is the density-of-state of the final state at $\pm \hbar \omega_N$ measured from the initial state energy. $D$ is inversely proportional to the energy uncertainty if the latter quantity is much larger than $\hbar \omega_N$. The energy broadening comes from the trion portion in the steady-state, which is $\sim \rho_t \gamma_t$ where $\gamma_t$ is the homogeneous broadening of the trion. In the current experimental configuration, $\rho_t$ is $\sim 0.1 - 0.2$ at the two absorption peak at the two sides of the dark state dip and we have $D \sim 10/\pi$. The last equality in Eq. (8) is by the fact that the hole spin operator $S_h^z$ has non-zero matrix elements only between the trion components of the electronic wavefunction. If $|\Psi_{i(f)}\rangle = a_{-i(f)}|x-\rangle + a_{+i(f)}|x+\rangle + a_{t,i(f)}|T-\rangle$, then $|\langle\Psi_{f}^\pm| S_h^z |\Psi_i\rangle|^2 = \frac{1}{4} |a_{t,i}|^2 |a_{t,f}|^2 \equiv \frac{1}{2} \rho_{t,i} \rho_{t,f}$, which leads to the final expression of $r_{D,k}^\pm$ in Eq.(8). It can be easily shown that the final expression of $r_{D,k}^\pm$ holds even when the steady-state of the three-level $\Lambda$ system is a mixed state.

There are probabilities for each nuclear spin to flip up or flip down and both rates $r_{D,k} \propto \rho_{t,i} \rho_{t,f}$. $\rho_t$ is determined by the probe detuning $\delta$ from the two-photon-resonance condition. $\delta \equiv \omega_{\text{probe}} - \omega_{\text{pump}} - g\mu_B B - \Delta$, where $\omega_{\text{pump}}$ and $\omega_{\text{probe}}$ are respectively the frequency of the pump and probe laser, $g\mu_B B$ is the electron Zeeman energy in external magnetic field, and $\Delta$ is the Overhauser shift by the nuclear field. The flip of nuclear spin $k$ changes the electron Zeeman energy by $\pm A_{e,k}$ and hence $\delta$ is changed by the same amount. Thus, $\rho_{t,f} = \rho_{t,i} \pm \frac{\partial \delta}{\partial A_{e,k}} A_{e,k}$. The DNP for nuclear
spin $k$ is

$$\frac{d}{dt} m_k = -\gamma_n m_k + r^{+}_{D,k} - r^{-}_{D,k}. \quad (9)$$

In the above we have taken $m_k$ as a continuous number, which is an approximation well justified since the quantum dot contains a sufficiently large number of nuclei. $\gamma_n$ is the nuclear spin relaxation rate, and

$$r^{+}_{D,k} - r^{-}_{D,k} \approx \pi \rho_t A^2_{e,k} O(\eta^4) D \left[ \frac{\partial \rho_t}{\partial \delta} A_{e,k}(j^2 - m_k^2 + j) + \rho_t m_k \right]. \quad (10)$$

C. Equation of motion for the Overhauser shift

Since the DNP rate $r_D$ depends on the probe detuning $\delta$ through $\rho_t$ and $\frac{\partial \rho_t}{\partial \delta}$, it is convenient to work with the total Overhauser shift: $\Delta \equiv \sum_k m_k A_{e,k}$. From Eq. (9), we have the equation-of-motion,

$$\frac{d}{dt} \Delta = -\gamma_n \Delta + \pi \rho_t O(\eta^4) D \sum_k A_{e,k} A^2_{h,k} \left[ \frac{\partial \rho_t}{\partial \delta} A_{e,k}(j^2 - m_k^2 + j) + \rho_t m_k \right]. \quad (11)$$

One can show that $\left| \frac{\partial \rho_t}{\partial \delta} A_{e,k}(j^2 - m_k^2 + j) \right| \gg |\rho_t m|$ if the nuclear polarization $p \ll 10\%$ in the quantum dot being studied. The magnitude of nuclear polarization established can be estimated from the frequency shift of the trion resonance. From the experimental data, one finds that the maximum $p$ ever established is $\sim 1\%$. Therefore, Eq. (11) becomes

$$\frac{d}{dt} \Delta = -\gamma_n \Delta + \alpha \rho_t \frac{\partial \rho_t}{\partial \delta} \quad (12)$$

where

$$\alpha = \pi O(\eta^4) D \sum_k A^2_{e,k} A^2_{h,k} (j^2 - m_k^2 + j)$$

$$\simeq \pi O(\eta^4) D A^2_{e,k} A^2_{h,k} \left( \frac{2}{N} \right)^4 \sum_k (j^2 - m_k^2 + j) \quad (13)$$

A unique feature of the DNP process described by Eq. (12) is that it tends to increase the trion population by adjusting the nuclear Overhauser field. As shown in Fig. 2A in the main text, the trion population in the steady state has two maxima at $\delta \sim \pm \Omega_{pump}/2$ which also correspond to the maxima of the probe absorption. Here $\Omega_{pump}$ is the Rabi frequency of the pump laser. Once the $\Lambda$ system is brought into either one of the two peak-regions, DNP by trion starts to take effect. DNP always tends to adjust the nuclear Overhauser field to evolve $\rho_t$ into a local maximum.
Therefore, the two maxima located at $\delta \sim \pm \Omega_{\text{pump}}/2$ should be the stable configurations for DNP. The fluctuations of the Overhauser field which shift the electron spin Zeeman splitting can be suppressed via the feedback process assisted by DNP. This self-focusing mechanism well explains all important behaviors observed.

The pump-probe spectroscopy with forward and backward scans of the probe frequency can be simulated using Eq. (12) which only has two parameters. Assuming $\alpha = 50$ (MHz)$^3$ and $\gamma_n = 2.5S^{-1}$, one finds the simulation results reproduces the fast scan experimental data reasonably well (see Fig. 4c in the main text). Assuming $\alpha = 2.4$ (MHz)$^3$ and $\gamma_n = 1.5S^{-1}$, one finds the simulation results reproduce the slow scan experimental data reasonably well (see Fig. 4d in the main text). We also point out that the absorption lineshapes depend on the pump detuning. For example, if the pump is detuned, the lineshapes between the forward and backward scans are not symmetric, as shown by Fig. 2(a) in the main text. Assuming $\alpha = 2.4$ (MHz)$^3$ and the pump is -1 GHz detuned to the trion transition, the numerical simulation in Fig. S1(a) qualitatively shows this asymmetric feature.

One can extract the magnitude of the hole-nuclear hyperfine coupling strength using Eq. (13). For the quantum dot being studied, we estimate $D \sim \frac{10}{\gamma_t} \sim 25\text{GHz}^{-1}$, $\eta = 0.2$, and $N \sim 10^4$. $j^2 - m^2 + j = 2.5$ for As and $j^2 - m^2 + j = 17$ for In. We find $A_h = 20\mu \text{eV} = 0.25A_e$ from the fast scan, and $A_h = 3\mu \text{eV} = 0.055A_e$ from the slow scan, in reasonable agreement with the estimation $A_h = 0.1 - 0.2A_e$ in Ref. 2 and 7. Since the number of nuclear spin $N$ appears in Eq. (10) to a 4th power, its deviation will have the largest effect on the estimation of $A_h$. Considering these possible parameter ranges, we expect that the extracted $A_h$ can be up to several times larger or smaller than the number we give above. For example, if we use $N \sim 2 \times 10^4$ instead, we estimate $A_h \sim A_e$ from the slow scan fit and $A_h \sim 0.2A_e$ from fast scan fit. If we use $N \sim 0.6 \times 10^4$ instead, we estimate $A_h \sim 0.1A_e$ from the slow scan fit and $A_h \sim 0.02A_e$ from fast scan fit. Our estimation suggest that slow scan fits tend to overestimate the value of $A_h$, while our fast scan fits tend to underestimate $A_h$. This suggests that, while our simple theoretical model well explains all key experimental features, it may not be sophisticate enough to account for all the precise numbers. Further theoretical studies are anticipated.
D. Further explanation of the data by invoking the self-locking effect

We can use this self-locking mechanism to further explain our experimental observations described in the main text. When the laser frequency scan is moving towards a trion maximum, DNP shifts the maximum to meet the laser frequency, leading to rising edges in the absorption spectrum. As the probe laser tries to move past the trion maximum, DNP shifts the maximum to follow the laser scan to maintain the maximum probe absorption, which explains the broadened and rounded top of the trion peak. After a sufficiently large nuclear spin polarization has been established, as the laser scans away from the absorption maximum, nuclear spin relaxation will dominate the nuclear field dynamics. At this point, the trion maximum can no longer follow the laser frequency and instead rebounds to its equilibrium position, moving away from the probe laser frequency. This leads to sharp falling edges in the spectrum.

The self-locking DNP mechanism naturally explains the switching behaviors in Fig.2 of the main text, including a subtle feature in Fig. 2d: there is no dip observed in the partial forward scan (blue curve) compared to the full forward scan (black curve). This is a signature that the system jumps from the low absorption hysteresis state I into the stability region II (Fig.4a in the main text). Since region II is already on the blue side of the dark state, there is no dark state formed during the partial forward scan. We can also expect that spectra of the partial (blue line) and full (black line) forward scans should be shifted from each other by a small amount, seen in Fig. 2d of the main text.

E. Control of the nuclear Overhauser field: tuning the mean value and narrowing the inhomogeneous broadening

The above hole spin assisted DNP has a unique feature. Once the probe laser is nearly resonant with the trion transition V2, the DNP starts to take effect to adjust the electron Zeeman energy. The electron Zeeman energy is always changed in the direction so that the steady-state trion population (and probe absorption) are increased. Once \( \delta \equiv \omega_{\text{probe}} - \omega_{\text{pump}} - g\mu_B B - \Delta \) reaches the value where the probe absorption is a maximum as a function of \( \delta \), a stable configuration is reached where DNP stops to take effect. This is confirmed by the good agreement of the theory and simulation with the observed spectral features, and from the time-dependent measurements. As shown in Fig. 2C and 2(F) in the main text, the high-absorption configuration is indeed the stable
FIG. 3: (color online) (a) The numerical simulation of the probe absorption spectra with pump -1 GHz detuned to the trion transition. (b) Calculated the change of DNP rate (slope) at the locking position with various pump Rabi frequencies. At each pump Rabi frequency, the calculation uses the extracted $T_2^*$ value as shown in Fig. 3a of the main text. Larger slope means better locking effect. Negative value of the slope corresponds to DNP acting as a restoring force to the locking position.

There are two stable configurations located at $\delta \sim \pm \Omega_{\text{pump}}/2$, which may be denoted as I and II respectively. After the system reaches either stable configuration, if we change the frequency of the probe laser by a small amount $\delta \omega$, DNP will automatically adjust the nuclear Overhauser field by the same amount so that the system remains in I (II), and hence the electron Zeeman frequency is determined ONLY by the pump and probe laser frequencies.

This DNP process provides a powerful tool to tune the electron Zeeman frequency on demand. In the slow scan, the range of this tunability on the electron Zeeman frequency is $\sim$ GHz. Since the nuclear field relaxes on an extremely slow timescale, this tunability may be used to pre-adjust the resonances of the QD electron spin qubit before the optically controlled logic operations. More significantly, narrowing the inhomogeneous broadening of the nuclear Overhauser field becomes possible by the same processes. For time ensemble measurements performed on a single dot, by preparing the system in a stable configuration with the same laser frequencies in every run of the measurement, the electron Zeeman frequency is set to the same value independent of the initial nuclear field. The inhomogeneous broadening due to different initial nuclear configurations is therefore eliminated.

Indeed, the $T_2^*$ plotted in Fig. 3a of main text does not correspond to the ultimate nuclear field locking that can be achieved at a given pump power. This is because in the two-beam experiment,
both the nuclear field locking and the probe of the coherent dark state are realized by the same pair of pump and probe lasers. First of all, an accurate measurement of the dark state dip requires a fast probe scan to reduce the effect of the continuing DNP which shifts the resonance during the measurement. This point is confirmed from the scan rate dependence shown in Fig. 1c of main text. However, in the fast scan measurement shown in Fig. 3a and 3b, since the locking position of the nuclear field changes as the probe laser scans at a rate much faster than the equilibration of DNP feedback, the suppression effect we obtained there is obviously a lower bound of the capability of this nuclear field locking technique.

The quantitative enhancement of $T_2^*$ by the self-locking mechanism is determined by the slope of the DNP rate as a function of detuning at the locking points, i.e. the two circled positions in Fig. 4b of main text, where $\delta = \pm \Omega_{pump}/2$. A larger value (more negative) means a stronger restoring force, and hence better a locking effect. Figure S3(b) plots the calculated slope of the DNP rate at the locking position as a function of the pump Rabi frequency. The negative sign means the DNP acts as a restoring force to the locking positions. The plot shows that the absolute value of slope increases as we increase the pump Rabi frequency. Therefore, the locking effect is enhanced by increasing the pump intensity, which is consistent with the experimental observations. However, from the discussion in the preceding paragraph and the comparison of Fig. 3b and 3c in the main text, we note that the $T_2^*$ as a function of pump power shown in Fig. 3a can only be viewed as a very loose lower bound of the enhancement effect.

F. Comparison between the present work and Ref. 18 in the main text

A comparison of the results in this paper with the data presented in Xu et al., Nat Phys 4, 692-695 (2008) (cited as Ref. 18 in main text) is called for. The results in Ref. 18 show coherent spin trapping in the dark state, but the signature of hysteresis reflecting the presence of strong
DNP was not observed under those experimental conditions. The experimental configuration was different: the optical field polarizations for the pump and probe are reversed leading possibly to a different level of excitation of hole spin states. Also, the trion transition driven by the pump has a stronger transition moment that the one studied in this paper. In particular, the pump intensity used in Ref. 18 ranges from a factor of 5 less than the minimum pump intensity to an intensity that is comparable with the minimum intensity in the current paper. The transition moment for the pump transition in the previous paper is also larger than the other transition coupling to the probe, but this transition is already saturated. The smaller transition moment coupling to the probe produces a weaker excitation as evidenced by a smaller absorption signal under comparable conditions, and a smaller trion excitation level. Since the DNP effect varies as the square of the trion excitation, this may be part of the explanation as to the absence of a strong DNP effect. Nevertheless, there is some evidence for a weak DNP effect since the trion linewidth is slightly broadened over the intrinsic linewidth, and the spin decoherence rate extracted from the data analysis is also smaller than the thermal value. Other signatures of spectrum servoing, however, are not significant under the conditions for that paper. Specifically, the onset of significant DNP induced spectrum servoing and hysteresis in the configuration of Ref. 18 was not observed until we reached a higher pump intensity in that arrangement (the maximum pump intensity used in Ref. 18 was 35 W/cm² and significant DNP induced spectrum servoing is not observable under the conditions of that experiment until we reach an intensity of 70 W/cm²). Work on this issue is continuing.

G. Relevance of the DNP by the electron spin

DNP effects can also be induced by the electron spin, which is more extensively investigated under other experimental setups. In the isotropic electron-nuclear hyperfine coupling, the terms that can flip a nuclear spin eigenstate are \( S^\pm I^{\pm} \). The spin state of the electron has to be simultaneously flipped with the nuclear spin, which costs the energy of the electron Zeeman energy \( g\mu_B B \). This energy (\(~ 7 \) GHz in a field of 1 Tesla) can be compensated by emission or absorption of phonons\(^6\). Alternatively, in the presence of resonant optical excitation, the energy change for flipping the electron can also be compensated in a two-photon assisted process as shown in Fig. S4\(^9\).

For the phonon-assisted process, we note that the experiment is performed at a temperature of 5K, where \( k_B T \gg g\mu_B B \). Therefore, the phonon bath is equally efficient in assisting the electron-
For the photon-assisted process, we note that the pump and probe are both strong, so that stimulated emission always dominates over spontaneous emission. Therefore, a two-photon assisted process involving $S^+_e I^-_k$ is always accompanied with an equally efficient two-photon assisted process involving $S^-_e I^+_k$ (see Fig. S4). So which one of $S^+_e I^-_k$ and $S^-_e I^+_k$ dominates is again determined by the steady state electron spin polarization.

Therefore, in DNP by both the phonon-assisted and photon-assisted electron nuclear flip-flops, the sign of the DNP rate is determined by the sign of the electron spin polarization only. In the present pump-probe setup, numerical calculation shows that the electron spin polarization is nearly a constant throughout the entire spectral range and is the same for forward scan and backward scan (see Fig. S5). Therefore, the nuclear spin can only be polarized in one direction, i.e. the Overhauser field can only be in one direction, regardless of the frequency scan direction of the probe laser. This is clearly in contradiction with the scan-direction dependent DNP effect observed in the experiments, e.g. the spectral position of the dark state is pushed into opposite directions between the forward and backward scans.

On the other hand, since the electron spin polarization remains nearly a constant value in the entire spectral range being studied, we expect the DNP by electron spin to contribute a constant background nuclear spin polarization $p_0$. This background nuclear spin polarization will be established right after the probe frequency scan begins, at the blue side or red side of the spectrum while still far away from the trion peak (see Fig. S5). DNP by the electron spin then only induces

FIG. 5: Calculated steady-state electron spin polarization as a function of two-photon Raman detuning $\delta$ in the pump probe spectroscopy.
an overall shift in the probe spectrum independent of the scan direction, but spectral lineshape of
the trion peak will not be affected. The magnitude of $p_0$ can be determined by changing the pump-
probe configuration so that the steady-state electron spin polarization has an overall sign change.
From the experimental data (not shown), we do find such a background nuclear polarization $p_0$, in
the order of several percent, corresponding to an overall frequency shift of $\sim 2$ GHz.

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