

# Collective nuclear stabilization in single quantum dots by noncollinear hyperfine interaction

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We present a theory of efficient suppression of the collective nuclear spin fluctuation, which prolongs the electron spin coherence time through the noncollinear hyperfine interaction between the nuclear spins and the hole spin. This provides a general paradigm to combat decoherence by direct control of environmental noise, and a possible solution to the puzzling observation of symmetric broadening of the absorption spectra in two recent experiments [Xu *et al.*, *Nature (London)* **459**, 1105 (2009) and Latta *et al.*, *Nature Phys.* **5**, 758 (2009)].

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## I. INTRODUCTION

Electron spins in semiconductor quantum dots (QDs) are promising candidates as solid-state quantum bits.<sup>1</sup> A critical obstacle is the short electron spin coherence time due to the fluctuating collective nuclear field from the nuclear spins of the host lattice.<sup>2</sup> Combating electron spin decoherence is of paramount importance for quantum information. For this purpose, two major approaches are under rapid development. One aims at decoupling a general qubit from the environments by frequently applying additional pulses on the qubit.<sup>3</sup> The other aims at directly prolonging the electron spin coherence time by stabilizing (i.e., suppressing the fluctuation of) the nuclear field through a nonlinear feedback loop driven by a steady-state nuclear field. The latter approach, referred to as nuclear stabilization (NS), has the merit that the prolongation of the electron spin coherence time persists for a long time, so that NS can be temporally separated from subsequent qubit operations. Intensive research efforts have led to successful NS in QD ensembles<sup>4</sup> and suppression of the fluctuation of the nuclear field difference between two coupled QDs.<sup>5</sup>

Recently, three groups<sup>6–8</sup> reported significant NS in single QDs. Xu *et al.*<sup>6</sup> observed prolongation of electron spin coherence time by NS upon optical pumping of trion in the Voigt geometry. Latta *et al.*<sup>7</sup> observed NS upon optical pumping of trion and blue exciton in the Faraday geometry. Vink *et al.*<sup>8</sup> theoretically deduced NS upon microwave pumping of electron spin resonance. A key observation in these experiments is the maintenance (i.e., locking) of resonant absorption over a wide range of pump frequency away from resonance. This locking behavior arises from the shift of the electron energy level from off-resonance to resonance by the steady-state nuclear field that drives the NS. A striking observation by both Xu *et al.* and Latta *et al.* is that the locking occurs nearly symmetrically on both sides of the resonance, in sharp contrast to the observation by Vink *et al.*, where the locking only occurs on the red side. This symmetric locking reveals that the steady-state nuclear field is antisymmetric across the resonance. However, this feature cannot be produced by the two cornerstone NS mechanisms: the Overhauser and/or the reverse Overhauser effect,<sup>7–10</sup> both of which are based on the flip of nuclear spins by the electron through the isotropic contact hyperfine interaction (HI). Xu *et al.*<sup>6</sup> gave a very inspiring clue by attributing their observation to the flip of nuclear spins by the hole (a vacancy in the valence band generated by optical pumping) through the noncollinear

dipolar HI,<sup>11,12</sup> but the mechanism proposed there flips the nuclear spins without a preferential direction and hence cannot produce any steady-state nuclear field to lock the resonance. To date, the symmetric locking remains an open problem.

In this paper, we provide a general mechanism capable of establishing the desired antisymmetric nuclear field and hence the symmetric locking. The key process of this mechanism is the flip of the nuclear spins through their noncollinear HI with the hole excited by nonresonant pumping (instead of resonant pumping<sup>6,11</sup> or through the isotropic contact HI with the ground-state electron under resonant pumping).<sup>7–10</sup> This process has a preferential direction giving rise to an antisymmetric steady-state nuclear field, which in turn drives a nonlinear feedback loop leading to efficient NS. This provides an avenue for electron spin coherence time prolongation through the noncollinear HI, in addition to the widely explored isotropic contact HI.

## II. GENERAL THEORY

Our model consists of many QD nuclear spins coupled to an electron spin state  $|0\rangle$  (with energy zero) and a hole spin state  $|1\rangle$  (with energy  $\omega_0$ ) under nonresonant pumping with detuning  $\Delta \equiv \omega_0 - \omega$  [Fig. 1(a)]. The excited hole state  $|1\rangle$  is coupled to the nuclear spins through the noncollinear HI  $\hat{\sigma}_{11} \sum_j a_{j,\text{nd}} (\hat{I}_j^+ + \hat{I}_j^-)$  ( $\hat{\sigma}_{ji} \equiv |j\rangle\langle i|$ ),<sup>6,11</sup> which flips the nuclear spins without changing the hole spin state. The ground electron state  $|0\rangle$  is coupled to the nuclear field  $\hat{h} = \sum_j a_{j,\text{d}} \hat{I}_j^z$  to be stabilized through the diagonal part  $\hat{\sigma}_{00} \hat{h}$  of the contact HI. An external magnetic field gives rise to the nuclear Zeeman term  $\sum_j \omega_{j,N} \hat{I}_j^z$ . The total density matrix  $\hat{\rho}(t)$  obeys the quantum Liouville equation, with the dephasing of  $|1\rangle$  (total rate  $\gamma_2$ ) and spontaneous emission  $|1\rangle \rightarrow |0\rangle$  (rate  $\gamma_1$ ) incorporated in the Lindblad form. Finally, we emphasize that our theory is equally applicable to the noncollinear HI between the electron spin and the nuclear spins (see Appendix A for details).

Before going into details, we outline the physical picture of the NS driven by our mechanism. It is a two-step feedback loop: the induction of a steady-state nuclear field by the hole through our mechanism and the back action of this nuclear field on the electron. First, through the noncollinear HI, a virtual hole generated by the nonresonant pumping flips the nuclear spins, leading to antisymmetric steady-state nuclear field. Second, through the interaction  $\hat{\sigma}_{00} \hat{h}$ , the nuclear field

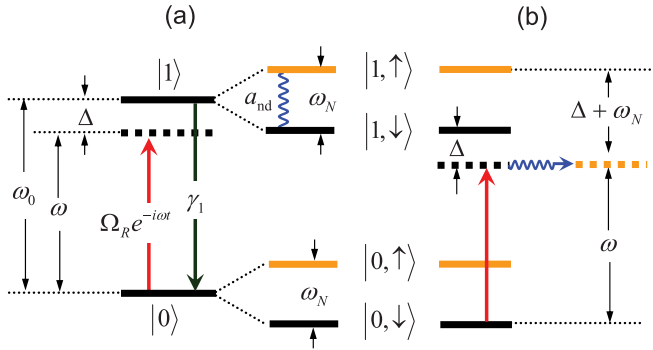


FIG. 1. (Color online) (a) Electron and hole spin states  $|0\rangle$  and  $|1\rangle$  coupled to a typical nuclear spin-1/2 by optical pumping. (b) Down-to-up nuclear spin-flip channel (solid arrow followed by wavy arrow) for  $\Delta > 0$ .

shifts the electron energy by  $\hat{h}$  and hence changing the detuning from  $\Delta$  [Fig. 1(a)] to  $\hat{\Delta} \equiv \Delta - \hat{h}$ . The antisymmetric steady-state nuclear field in the first step enables the feedback loop to produce the desired symmetric locking and efficient NS (i.e., suppression of the fluctuation of  $\hat{h}$ ). All this information is contained in the diagonal part  $\hat{P}(t)$  of the nuclear spin density matrix  $\text{Tr}_{eh} \hat{\rho}(t)$ .

The dynamics of  $\hat{P}(t)$ , which occurs on a very long time scale compared with other variables, is derived from the quantum Liouville equation of  $\hat{\rho}(t)$  by applying the adiabatic approximation.<sup>13</sup> Up to  $O(a_{\text{nd}}^3)$ , we obtain (see Appendix B)

$$\dot{\hat{P}} = - \sum_j [\hat{I}_j^-, \hat{I}_j^+ W_{j,+}(\hat{\Delta}) \hat{P}] - \sum_j [\hat{I}_j^+, \hat{I}_j^- W_{j,-}(\hat{\Delta}) \hat{P}], \quad (1)$$

where  $W_{j,+}(\hat{\Delta})$  [ $W_{j,-}(\hat{\Delta})$ ] is the rate of the flip of the  $j$ th nuclear spin that increases (decreases)  $\hat{I}_j^z$  by one. They are obtained from the steady-state hole fluctuation  $a_{j,\text{nd}}^2 \int_{-\infty}^{\infty} dt e^{\mp i\omega_{j,N}t} \langle \hat{\sigma}_{11}(t) \hat{\sigma}_{11} \rangle$  in the absence of nuclear spins by replacing  $\Delta$  with  $\hat{\Delta}$ . Equation (1) is the starting point of our subsequent discussions. It can be easily understood from the fluctuation-dissipation relation:<sup>14</sup> the hole fluctuation  $\langle \hat{\sigma}_{11}(t) \hat{\sigma}_{11} \rangle$  drives the irreversible nuclear spin flip leading to a steady-state nuclear field, which in turn acts back on the electron by replacing  $\Delta$  with  $\hat{\Delta}$ .

To keep the exposition simple, we consider uniform couplings  $a_{j,d} = a_d$  and  $a_{j,\text{nd}} = a_{\text{nd}}$  to identical nuclei  $I_j = I$  and  $\omega_{j,N} = \omega_N$ . Then  $W_{j,\pm}(\hat{\Delta})$  becomes  $W_{\pm}(\hat{\Delta})$  and the nuclear field  $\hat{h} = h_{\text{max}} \hat{s}$ , where  $h_{\text{max}} \equiv Na_d I$ ,  $N$  is the number of QD nuclei, and  $\hat{s} \equiv (1/N) \sum_{j=1}^N \hat{I}_j^z / I$ . Hereafter we also refer to  $\hat{s} = \hat{h} / h_{\text{max}}$  as the nuclear field.

First we demonstrate the desired antisymmetric behavior of the steady-state nuclear field. To avoid complication, we drop the back action, so  $W_{\pm}(\hat{\Delta})$  becomes  $W_{\pm} \equiv W_{\pm}(\Delta)$ . In this case, Eq. (1) shows that on the coarse-grained time scale of  $\hat{P}(t)$ , the faster nuclear spin decoherence causes the dynamics of different nuclear spins to be independent, and it suffices to consider only one nuclear spin  $\mathbf{I}$ . For a nuclear spin-1/2 with two Zeeman sublevels  $|\uparrow\rangle$  and  $|\downarrow\rangle$ , Eq. (1) reduces to  $\dot{P}_{\uparrow\uparrow} = W_+ P_{\downarrow\downarrow} - W_- P_{\uparrow\uparrow}$ . The nuclear field  $s = P_{\uparrow\uparrow} - P_{\downarrow\downarrow}$  obeys  $\dot{s} = -\Gamma_p(s - s_0)$  [i.e., a finite steady-state nuclear field

$s_0 = (W_+ - W_-)/(W_+ + W_-)$  is established within a time scale  $1/\Gamma_p$ ], where  $\Gamma_p \equiv W_+ + W_-$ . For a nucleus with spin  $I \geq 1/2$ , Eq. (1) gives  $\dot{s} = -\Gamma_p(s - s_0^{(I)})$  for the nuclear field  $s \equiv \langle \hat{I}^z(t) \rangle / I$ , where  $s_0^{(I)} \approx 2(I+1)s_0/3$  for  $|s_0^{(I)}| \ll 1$ . Straightforward evaluation of  $\langle \hat{\sigma}_{11}(t) \hat{\sigma}_{11} \rangle$  gives

$$s_0 \approx -\frac{2\Delta\omega_N}{\Delta^2 + \gamma_2^2} F \sim -\frac{\Delta\omega_N}{\gamma_2^2}, \quad (2)$$

$$\Gamma_p \approx \frac{4\tilde{a}_h^2}{\gamma_1} \frac{W\gamma_1^2}{(\gamma_1 + 2W)^3} c_1 \sim \frac{\tilde{a}_h^2}{\gamma_1} \quad (3)$$

up to leading order of  $|\omega_N|/\gamma_{1,2}$ , where  $F$  and  $c_1$  are positive  $O(1)$  quantities (see Appendix B) and  $W \equiv 2\pi(\Omega_R/2)^2 L^{(\gamma_2)}(\Delta)$  is the pumping rate, with  $L^{(\gamma_2)}(\Delta) \equiv (\gamma_2/\pi)/(\Delta^2 + \gamma_2^2)$  being the energy conserving delta function broadened by hole dephasing.  $s_0$  displays two distinguishing features: the antisymmetric behavior  $s_0|_{\Delta \rightarrow -\Delta} = -s_0$  and the weak dependence on the pumping rate  $W$  (since  $F$  is weakly dependent on  $W$ ). This weak dependence is very different from the two existing NS mechanisms: the Overhauser effect<sup>9</sup>  $|s_0|$  is maximal for saturated pumping  $W \gg \gamma_1$  and the reverse Overhauser effect<sup>10</sup>  $|s_0|$  is maximal for weak pumping  $W \ll \gamma_1$ .

The above distinguishing features of our mechanism can be explained qualitatively as the competition between two coherent nuclear spin-flip channels  $|0, \downarrow\rangle \xrightarrow{\Omega_R e^{-i\omega t}} |1, \downarrow\rangle \xrightarrow{a_{\text{nd}}} |1, \uparrow\rangle$  (down-to-up channel) and  $|0, \uparrow\rangle \xrightarrow{\Omega_R e^{-i\omega t}} |1, \uparrow\rangle \xrightarrow{a_{\text{nd}}} |1, \downarrow\rangle$  (up-to-down channel). Each channel consists of two steps: the generation of a virtual hole by the nonresonant pumping followed by the nuclear spin flip by this virtual hole through the noncollinear HI. For the down-to-up channel [Fig. 1(b)], the first step (solid arrow) has an energy mismatch from the detuning  $|\Delta|$ , but the second step (wavy arrow) is nontrivial: the hole in its initial state  $|1, \downarrow\rangle$  is a virtual hole with energy  $\omega$  (instead of a real hole with energy  $\omega_0$ ), while the hole in its final state  $|1, \uparrow\rangle$  is a real hole with energy  $\omega_0$ . Therefore, the energy mismatch of this step (i.e., the difference between the final state energy  $\omega_0 + \omega_N/2$  and the initial state energy  $\omega - \omega_N/2$ ) is  $|\omega_N + \Delta|$ . Similarly, for the up-to-down channel, the energy mismatch is  $|\Delta|$  in the first step and  $|\omega_N - \Delta|$  in the second step. The different energy mismatches  $|\omega_N \pm \Delta|$  (instead of the pumping)<sup>7-10</sup> sets a  $\Delta$ -dependent preferential direction for the nuclear spin flip. This is the origin of the antisymmetric behavior  $s_0|_{\Delta \rightarrow -\Delta} = -s_0$  and the weak dependence of  $s_0$  on the pumping rate  $W$ . For  $|\omega_N| \ll \gamma_{1,2}$  and  $|\Delta| \ll \gamma_2$ , the small energy mismatches  $|\Delta|, |\omega_N \pm \Delta| \ll \gamma_2$  of both channels are easily compensated by the strong dephasing (with rate  $\gamma_2$ ), so the nuclear spin flip is nearly resonant, in contrast to the off-resonant Overhauser effect.<sup>9</sup> This resonant nature makes the strength of our mechanism comparable with the detrimental nuclear spin depolarization even if the noncollinear HI is weak.<sup>6,11</sup>

The physics of our mechanism depicted above differs qualitatively from previous theories.<sup>6-11</sup> First, the nuclear spin flip is not accompanied by any change of the hole state, in contrast to the pairwise electron-nuclear spin flip flop.<sup>7-10</sup> Second, the nuclear spin is flipped by a virtual hole in a coherent channel (resembling cotunneling).<sup>15</sup> This is the origin of  $s_0|_{\Delta \rightarrow -\Delta} = -s_0$  responsible for symmetric

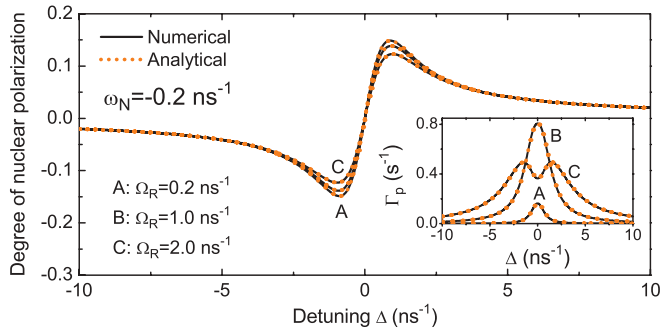


FIG. 2. (Color online) Comparison of our analytical results (dotted lines) for  $s_0$  and  $\Gamma_p$  (inset) with the numerical solution to the quantum Liouville equation (solid lines) involving one nuclear spin-1/2.

locking and efficient NS. By contrast, in an incoherent channel (resembling sequential tunneling),<sup>15</sup> the nuclear spin flip by a real hole<sup>6,11</sup> has no preferential direction<sup>6,11</sup> to lock the resonance, while the nuclear spin flip by a real electron<sup>8–10</sup> has  $\Delta$ -independent preferential direction leading to unidirectional locking.<sup>16</sup> Third, the nuclear Zeeman splitting  $\omega_N$ , considered as negligible in all previous theories,<sup>6–11</sup> plays a critical role in our mechanism.

Now we analyze the entire feedback loop by including the back action. We are interested in the nuclear field  $\hat{s} = \hat{h}/h_{\max}$  in the steady state: its average value  $s^{(ss)} = h^{(ss)}/h_{\max}$  is responsible for the symmetric locking and its fluctuation  $\sigma$  quantifies the NS. This information is contained in the distribution function  $p(s, t) \equiv \text{Tr} \hat{P}(t) \delta(s - \hat{s})$  of  $\hat{s}$ . Using Eq. (1), the equation of motion of  $p(s, t)$  is derived and solved straightforwardly (see Appendix C). The steady-state solution  $p^{(ss)}(s)$  shows one or more sharp Gaussian peaks, each of which corresponds to a stable nuclear spin state: the position  $s_\alpha^{(ss)}$  (standard deviation  $\sigma_\alpha$ ) of the  $\alpha$ th peak corresponds to the average nuclear field (nuclear field fluctuation) in the  $\alpha$ th stable state. For  $|s_0^{(I)}| \ll 1$  or  $I = 1/2$ , we obtain the analytical

result for the fluctuation in the  $\alpha$ th stable state

$$\sigma_\alpha = \sigma^{\text{eq}} \sqrt{\frac{1 - s_0^2}{1 + h_{\max} (ds_0^{(I)}/d\Delta)|_{\Delta \rightarrow \Delta - h_{\max} s_\alpha^{(ss)}}}}, \quad (4)$$

where  $\sigma^{\text{eq}} \equiv [(I + 1)/(3NI)]^{1/2}$  is the thermal equilibrium fluctuation. Equation (4) shows that the NS driven by our mechanism is efficient since  $s_0$  and hence  $s_0^{(I)} \approx [2(I + 1)/3]s_0$  are very sensitive to  $\Delta$  on the resonance  $\Delta = 0$  [see Eq. (2)], where the strength  $\Gamma_p$  of our mechanism is maximal [see Eq. (3)].

### III. APPLICATION TO EXPERIMENTS

Now we apply our model to describe the single-pump experiment of Xu *et al.*<sup>6</sup> and the trion pump experiment of Latta *et al.*<sup>7</sup>, which correspond to  $|0\rangle$  being the spin-up electron state and  $|1\rangle$  being the spin-up trion state (consisting of two inert electrons in a spin singlet and a spin-up hole). In this case, the diagonal part  $\sum_j a_{j,e} \hat{S}_e^z \hat{I}_j^z$  of the electron-nuclear contact HI gives the diagonal coupling  $\hat{\sigma}_{00} \hat{h}$  with  $a_{j,d} = a_{j,e}/2$ . The non-collinear hole-nuclear dipolar HI<sup>6,11</sup>  $\sum_j O(\eta^2) a_{j,h} \hat{S}_h^z (\hat{I}_j^+ + \hat{I}_j^-)$  gives the noncollinear HI with  $a_{j,nd} = O(\eta^2) a_{j,h}$ . For specificity, we consider a typical InAs QD under a magnetic field of 2–3 T and use the following realistic parameters<sup>2,6,7,17</sup> (with  $\hbar = 1$  understood) unless specified:  $N = 10^4$ ,  $I = 9/2$ ,  $\Omega_R = \gamma_1 = \gamma_2 = 1$  ns<sup>-1</sup>,  $\omega_N = \pm 0.2$  ns<sup>-1</sup>,  $a_e = 10^{-2}$  ns<sup>-1</sup>,  $a_h = 10^{-3}$  ns<sup>-1</sup>, and  $a_{nd} = 0.04 a_h$  (based on experimentally reported hole mixing  $|\eta| = 0.2\text{--}0.7$ ).<sup>18</sup>

Figure 2 shows  $s_0$  and  $\Gamma_p$  (inset) in the absence of the back action. First, our analytical results (dotted lines) from Eqs. (2) and (3) agree well with the numerical solution (solid lines) to the original quantum Liouville equation involving one nuclear spin-1/2. Second, the feature  $s_0|_{\Delta \rightarrow -\Delta} = -s_0$  and its weak dependence on the pumping rate  $W \propto \Omega_R^2$  are obvious.

Figure 3 shows the stable [the peaks of  $p^{(ss)}(s)$ ] and unstable [the dips of  $p^{(ss)}(s)$ ] nuclear fields, the absorption spectra, and

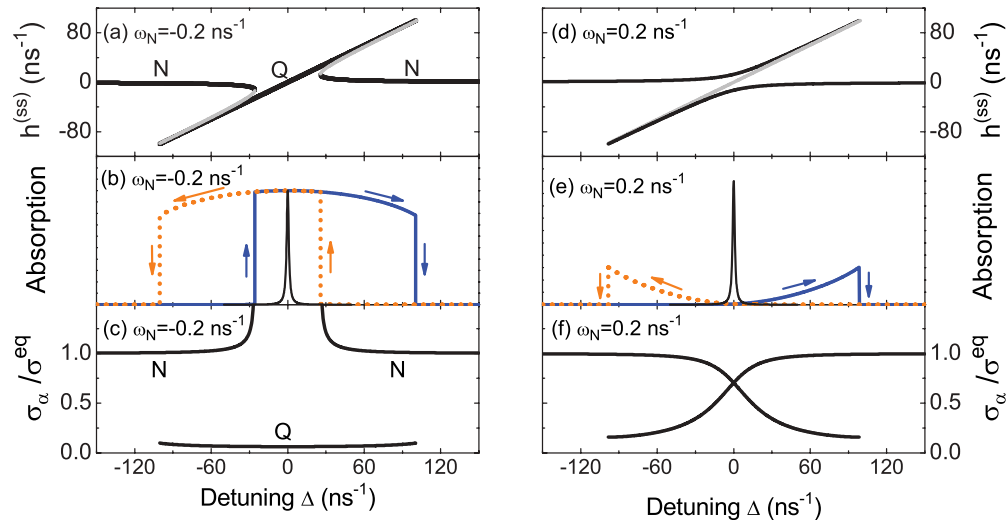


FIG. 3. (Color online) (a), (d): Stable (black lines) and unstable (gray lines) nuclear field  $h^{(ss)} = h_{\max} s^{(ss)}$ . (b), (e): Absorption spectra obtained by sweeping  $\Delta$  in different directions (indicated by the arrows). The sharp Lorentzian peaks at  $\Delta = 0$  are absorption spectra in the absence of the nuclei. (c), (f): NS  $\sigma_\alpha / \sigma^{\text{eq}}$  in stable states (N and Q branches).

the NS in the presence of the back action. First, the steady-state nuclear field  $s^{(ss)}$  is bistable and antisymmetric  $s^{(ss)}|_{\Delta \rightarrow -\Delta} = -s^{(ss)}$ , leading to hysteretic symmetric locking<sup>6,7</sup> for  $\omega_N < 0$ , as shown in Fig. 3(b). For  $\omega_N > 0$  [Fig. 3(d)], the absorption peak is shifted to finite detunings [Fig. 3(e)], as observed recently.<sup>19</sup> Second, a maximal degree of NS  $\sim 15$  appears in the  $Q$  branch of Fig. 3(c).

So far we have neglected the detrimental effect of nuclear depolarization. For a direct comparison with the experiment, we include a uniform depolarization rate  $\Gamma_{\text{dep}}$ , which changes  $W_{j,\pm}(\hat{\Delta})$  in Eq. (1) to  $W_{j,\pm}(\hat{\Delta}) + \Gamma_{\text{dep}}/2$ . It reduces  $s_0^{(I)}$  and hence the extent of the locking [Fig. 3(b)] by a factor  $\Gamma_p/(\Gamma_p + \Gamma_{\text{dep}})$  and the NS by a factor  $\sim [\Gamma_p/(\Gamma_p + \Gamma_{\text{dep}})]^{1/2}$ . The typical  $\Gamma_{\text{dep}} \sim 1 \text{ s}^{-1}$  in the experiments<sup>6,7</sup> is comparable to the typical  $\Gamma_p \sim 1 \text{ s}^{-1}$  (inset of Fig. 2). Note that  $\Gamma_p \propto O(\eta^4)$  depends strongly on the hole mixing coefficient  $\eta$ . For the InAs QD under consideration, a relatively strong depolarization  $\Gamma_{\text{dep}} \sim 10\Gamma_p$  would reduce  $s_0^{(I)}$  from  $\sim 40\%$  to  $\sim 4\%$  and the NS [Fig. 3(c)] from  $\sim 15$  to  $\sim 5$ , in order-of-magnitude agreement with Xu *et al.*<sup>6</sup> For an InGaAs QD as used by Latta *et al.*,<sup>7</sup> the extent  $\sim 30 \text{ ns}^{-1}$  of the observed locking for trion corresponds to  $\Gamma_p \sim \Gamma_{\text{dep}}$ . Similarly, the symmetric locking for blue exciton<sup>7</sup> can also be reproduced with  $\Gamma_p \sim \Gamma_{\text{dep}}$  by identifying  $|0\rangle$  with the vacuum state and  $|1\rangle$  with the blue exciton state.

#### IV. CONCLUSION

In summary, we have provided a general mechanism for generating steady-state nuclear spin polarization through the noncollinear hyperfine interaction between the hole spin and the nuclear spins. The antisymmetric behavior of the nuclear spin polarization provides a possible solution to the puzzling observation of symmetric broadening of the absorption spectra in two recent experiments.<sup>6,7</sup> This mechanism also drives a nonlinear feedback loop leading to efficient suppression of the nuclear spin fluctuation and hence provides an avenue for electron spin coherence time prolongation through the noncollinear HI, in addition to the widely explored isotropic contact HI.

Finally, we mention two possible directions for future research. First, parameter-free fitting of existing experiments<sup>19</sup> and inclusion of other interactions and effects beyond our simple model [e.g., the nuclear quadrupole interaction<sup>20</sup> and the frequency focusing effect<sup>4,13</sup> (which may be involved in the two-pump setup of Xu *et al.*<sup>6</sup>)]. Second, our theory can also be applied to the electron spin interacting with the nuclear spins through the noncollinear HI<sup>19</sup> (see Appendix A for details) (e.g., in phosphorus donor in silicon<sup>12</sup> or conventional QDs with strong spin-orbit coupling in the conduction band).

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#### APPENDIX A: ELECTRON-NUCLEAR AND HOLE-NUCLEAR INTERACTIONS AND NUCLEAR SPIN-FLIP MECHANISMS

First we briefly summarize all identified HIs between the nuclear spins  $\{\hat{\mathbf{I}}_j\}$  and the  $S_e = 1/2$  electron spin  $\hat{\mathbf{S}}_e$  or the  $S_h = 3/2$  hole spin  $\hat{\mathbf{S}}_h$  in a quantum dot (QD) grown along the  $z$  axis:

(i) The electron spin  $\hat{\mathbf{S}}_e$  is coupled to the nuclear spins  $\{\hat{\mathbf{I}}_j\}$  through the isotropic contact HI

$$\hat{V}_{eN} = \sum_j a_{j,e} \hat{\mathbf{S}}_e \cdot \hat{\mathbf{I}}_j.$$

In an external magnetic field along the  $z$  axis,  $\hat{V}_{eN}$  can be decomposed into the sum of the Ising-like term

$$\hat{V}_{eN}^{(\text{Ising})} \equiv \sum_j a_{j,e} \hat{S}_e^z \hat{I}_j^z$$

and the pairwise flip-flop term

$$\hat{V}_{eN}^{(\text{flip-flop})} \equiv \sum_j a_{j,e} (\hat{S}_e^x \hat{I}_j^x + \hat{S}_e^y \hat{I}_j^y).$$

(ii) The dipolar HI between the nuclear spins and the  $S_h = 3/2$  hole spin  $\hat{\mathbf{S}}_h$  assumes a complicated form<sup>17</sup> because the envelope function of the heavy hole is different from that of the light hole. If this difference is neglected, then this interaction becomes

$$\hat{V}_{hN} = \sum_j a_{j,h} \hat{\mathbf{S}}_h \cdot \hat{\mathbf{I}}_j,$$

where  $|a_{j,h}| \sim 0.1|a_{j,e}|$  is the interaction strength. It assumes the same form as the electron-nuclear contact HI. In an external magnetic field along the  $z$  axis,  $\hat{V}_{hN}$  can be decomposed into the sum of the Ising-like term

$$\hat{V}_{hN}^{(\text{Ising})} \equiv \sum_j a_{j,h} \hat{S}_h^z \hat{I}_j^z$$

and the pairwise flip-flop term

$$\hat{V}_{hN}^{(\text{flip-flop})} \equiv \sum_j a_{j,h} (\hat{S}_h^x \hat{I}_j^x + \hat{S}_h^y \hat{I}_j^y).$$

(iii) Both the electron-nuclear contact HI and the hole-nuclear dipolar HI could contribute to the noncollinear HI  $\hat{\sigma}_{11} \sum_j a_{j,\text{nd}} (\hat{I}_j^+ + \hat{I}_j^-)$  as utilized in our mechanism. For the state  $|1\rangle$  being a nondegenerate electron spin state, if its quantization axis  $\langle 1|\hat{\mathbf{S}}_e|1\rangle \equiv \mathbf{n}$  deviates from the  $z$  axis (this could happen in the presence of spin-orbit coupling), then  $\hat{V}_{eN}$  would give rise to the noncollinear coupling

$$\begin{aligned} \hat{V}_{eN}^{(\text{noncollinear})} &= |1\rangle\langle 1| \sum_j a_{j,e} (n_x \hat{I}_j^x + n_y \hat{I}_j^y) \\ &\longrightarrow |1\rangle\langle 1| \sum_j \frac{\sqrt{n_x^2 + n_y^2} a_{j,e}}{2} (\hat{I}_j^+ + \hat{I}_j^-), \end{aligned}$$

where the last step is obtained by a nuclear spin rotation along the  $z$  axis (which does not change the dynamics of  $\{\hat{I}_j^z\}$  and hence the nuclear field). Similarly, for the state  $|1\rangle$  being a nondegenerate heavy hole state, if its quantization axis  $\langle 1|\hat{\mathbf{S}}_h|1\rangle$  deviates from the  $z$  axis (this usually happens



due to the heavy-light hole mixing), then  $\hat{V}_{hN}$  would also give rise to a noncollinear coupling  $\hat{V}_{hN}^{(\text{noncollinear})}$ . For example, due to heavy-light hole mixing, the lowest hole state  $|1\rangle \approx |3/2, \pm 3/2\rangle + O(\eta)|3/2, \mp 1/2\rangle + O(\eta^2)|3/2, \pm 1/2\rangle$  is the mixture of the dominant heavy-hole components  $|3/2, \pm 3/2\rangle$  and a small amount of light-hole components  $|3/2, \pm 1/2\rangle$ , where  $\eta$  is the hole mixing coefficient.<sup>18</sup> In this case,  $\hat{V}_{hN}$  gives rise to the noncollinear coupling<sup>6</sup>

$$\begin{aligned}\hat{V}_{hN}^{(\text{noncollinear})} &= |1\rangle\langle 1| \sum_j O(\eta^2) a_{j,h} (\hat{I}_j^+ + \hat{I}_j^-) \\ &\equiv |1\rangle\langle 1| \sum_j \tilde{a}_{j,h} (\hat{I}_j^+ + \hat{I}_j^-).\end{aligned}$$

The strength  $\tilde{a}_{j,h} \equiv a_{j,h} O(\eta^2)$  of the hole-nuclear noncollinear HI depends strongly on the hole-mixing coefficient  $\eta$ . Values of  $|\eta| = 0.2\text{--}0.7$  have been reported by a series of experimental measurements<sup>18,21</sup> and an atomistic pseudopotential calculation,<sup>22</sup> which explained successfully the experimental fine structure of excitons in self-assembled InGaAs/GaAs dots. The precise value of  $\eta$  depends strongly on the geometry of the quantum dot. Hole mixing can be strong in the presence of in-plane anisotropy and/or strain,<sup>21</sup> while it decreases significantly when the quantum dot becomes flatter.<sup>23</sup> For a hole mixing  $|\eta| = 0.1$ , the noncollinear hole-nuclear HI  $\tilde{a}_{j,h}$  is weaker than the electron-nuclear contact HI  $a_{j,e}$  by a factor  $\sim 10^3$ . Finally we mention that in silicon, the electron-nuclear HI contains a large noncollinear term  $\propto \hat{S}_e^z (\hat{I}_j^+ + \hat{I}_j^-)$ .<sup>12</sup>

Below we summarize existing nuclear spin-flip mechanisms based on the different parts of the electron-nuclear and hole-nuclear interactions. For specificity we consider an external magnetic field  $B = 2\text{--}3$  T along the  $z$  axis, where the magnitudes of the Zeeman splittings  $\omega_e$ ,  $\omega_h$ , and  $\omega_N$  of the electron, the hole, and the nuclear spins are  $|\omega_e| \sim 100$  ns<sup>-1</sup>,  $|\omega_h| \sim 100$  ns<sup>-1</sup>, and  $|\omega_N| \sim 0.1$  ns<sup>-1</sup>, respectively.

(i) *Overhauser effect.*<sup>9</sup> The nuclear spin is flipped by the electron spin through  $\hat{V}_{eN}^{(\text{flip-flop})}$ , accompanied by an electron spin flip. This process has a large energy mismatch  $|\omega_e| \sim 100$  ns<sup>-1</sup>, which is dissipated by a thermal bath. The nuclear spin flip has a preferential direction determined by the deviation of the electron spin polarization away from thermal equilibrium. Obviously,  $\hat{V}_{hN}^{(\text{flip-flop})}$  could induce a similar effect, but the strength is much weaker because  $|a_h| \sim 0.1|a_e|$ .

(ii) *Reverse Overhauser effect.*<sup>10</sup> The nuclear spin is flipped by the electron spin through  $\hat{V}_{eN}^{(\text{flip-flop})}$ , accompanied by an electron spin flip. This process has a large energy mismatch  $|\omega_e| \sim 100$  ns<sup>-1</sup>, which is compensated by an ac electric field. The nuclear spin flip has a preferential direction determined by the steady-state electron spin polarization. Obviously,  $\hat{V}_{hN}^{(\text{flip-flop})}$  could also induce a similar effect, but the strength is much weaker.

(iii) *The effect proposed by Xu et al.*<sup>6</sup> and subsequently elaborated by Ladd et al.<sup>11</sup> The nuclear spin is flipped by a real, optically excited hole spin through the noncollinear HI  $\hat{V}_{hN}^{(\text{noncollinear})}$  without any accompanying hole spin flip. The nuclear spin flip in both directions has the same, small energy mismatch  $|\omega_N| \sim 0.1$  ns<sup>-1</sup> (so the nuclear spin flip has no preferential direction), which is dissipated by spontaneous emission. For a hole mixing  $\eta \sim 0.1$ , we have  $\tilde{a}_h \sim 10^{-3}a_e$

and  $|\omega_N| \sim 10^{-3}|\omega_e|$ . Therefore, the strength  $\sim \tilde{a}_h^2/\omega_N^2$  of this effect<sup>6,11</sup> may be comparable with the strength  $\sim a_e^2/\omega_e^2$  of the Overhauser effect.<sup>9</sup> However, due to the absence of a preferential nuclear spin-flip direction, this mechanism cannot establish any steady nuclear field to lock the resonance [i.e., Eq. (9) and subsequent equations in the supplementary materials of Ref. 6 are ungrounded].

In the present work, we focus on the nuclear spin dynamics driven by an optically excited virtual hole through the noncollinear HI  $\hat{V}_{hN}^{(\text{noncollinear})}$ . We drop the flip-flop term  $\hat{V}_{eN}^{(\text{flip-flop})}$  and  $\hat{V}_{hN}^{(\text{flip-flop})}$ , since the nuclear spin dynamics (e.g., Overhauser<sup>9</sup> or reverse Overhauser<sup>10</sup> effect) due to such interactions has already been intensively investigated and can be trivially incorporated by introducing the corresponding Lindblad operators.

## APPENDIX B: DERIVATION OF EQ. (1)

In the frame rotating with the pump frequency  $\omega$ , the Hamiltonian is

$$\begin{aligned}\hat{H} &= -(\Delta - \hat{h})\hat{\sigma}_{00} + \frac{\Omega_R}{2}(\hat{\sigma}_{10} + \hat{\sigma}_{01}) + \sum_j \omega_{j,N} \hat{I}_j^z \\ &\quad + \hat{\sigma}_{11} \sum_j a_{j,\text{nd}}(\hat{I}_j^+ + \hat{I}_j^-),\end{aligned}\quad (\text{B1})$$

where  $\Delta \equiv \omega_0 - \omega$  is the nominal pump detuning. The evolution of the density matrix  $\hat{\rho}$  of the whole system is governed by the master equation

$$\dot{\hat{\rho}} = -i[\hat{H}, \hat{\rho}] + \dot{\hat{\rho}}_{\text{damp}}, \quad (\text{B2})$$

where the Lindblad dissipative part

$$\begin{aligned}\dot{\hat{\rho}}_{\text{damp}} &= -\gamma_1 \left( \frac{\hat{\sigma}_{11}\hat{\rho} + \hat{\rho}\hat{\sigma}_{11}}{2} - \hat{\sigma}_{01}\hat{\rho}\hat{\sigma}_{10} \right) \\ &\quad - 2\gamma_2^\varphi \left( \frac{\hat{\sigma}_{11}\hat{\rho} + \hat{\rho}\hat{\sigma}_{11}}{2} - \hat{\sigma}_{11}\hat{\rho}\hat{\sigma}_{11} \right)\end{aligned}$$

includes the spontaneous emission  $|1\rangle \rightarrow |0\rangle$  with rate  $\gamma_1$  and hole pure dephasing with rate  $\gamma_2^\varphi$ . The total hole dephasing rate is  $\gamma_2 \equiv \gamma_1/2 + \gamma_2^\varphi \geq \gamma_1/2$ .

For clarity we introduce the nuclear spin basis  $|\mathbf{m}\rangle \equiv \otimes_j |m_j\rangle_j$ , where  $|m_j\rangle_j$  is the Zeeman eigenstate of the  $j$ th nuclear spin,  $\hat{I}_j^z|m_j\rangle_j = m_j|m_j\rangle_j$ , where  $m_j = -I, -I+1, \dots, I$ . In the product basis  $\{|0, \mathbf{m}\rangle, |1, \mathbf{m}\rangle\}$  of the electron and hole states  $\{|0\rangle, |1\rangle\}$  and the nuclear spin states  $\{|\mathbf{m}\rangle\}$ , there are seven relevant density matrix elements:  $\rho_{0\mathbf{m},0\mathbf{m}}$ ,  $\rho_{1\mathbf{m},1\mathbf{m}}$ ,  $\rho_{1\mathbf{m},0\mathbf{m}}$ ,  $\rho_{0\mathbf{m}+1\mathbf{j},0\mathbf{m}}$ ,  $\rho_{1\mathbf{m}+1\mathbf{j},1\mathbf{m}}$ ,  $\rho_{1\mathbf{m}+1\mathbf{j},0\mathbf{m}}$ , and  $\rho_{0\mathbf{m}+1\mathbf{j},1\mathbf{m}}$ , where  $|\mathbf{m}+1\mathbf{j}\rangle \equiv |m_j+1\rangle \otimes_{k \neq j} |m_k\rangle$ . Their equations of motion follow directly from Eq. (B2). In particular, an arbitrary element  $P_{\mathbf{m},\mathbf{m}} = \rho_{0\mathbf{m},0\mathbf{m}} + \rho_{1\mathbf{m},1\mathbf{m}}$  of the diagonal part  $\hat{P}(t)$  of the nuclear spin density matrix obeys

$$\dot{P}_{\mathbf{m},\mathbf{m}} = 2 \sum_j a_{j,\text{nd}} [\eta_{m_j} \text{Im} \rho_{1\mathbf{m}+1\mathbf{j},1\mathbf{m}} - (\mathbf{m} \rightarrow \mathbf{m}-1\mathbf{j})], \quad (\text{B3})$$

where  $\eta_m = \sqrt{(I-m)(I+m+1)}$ . The dynamics of  $\hat{P}(t)$  or equivalently its matrix elements  $\{P_{\mathbf{m}',\mathbf{m}}(t)\}$  is singled out if we can express  $\rho_{1\mathbf{m}+1\mathbf{j},1\mathbf{m}}$  as a function of  $\{P_{\mathbf{m}',\mathbf{m}}(t)\}$ . This

is achieved through the adiabatic approximation, which is justified by the very long time scale for the dynamics of  $\hat{P}(t)$  (characterized by a time scale  $T_1^N \sim 1$  s)<sup>6,7</sup> compared with the much shorter time scales of other density matrix elements (including  $\rho_{1\mathbf{m}+1\mathbf{j},1\mathbf{m}}$ ):

(i) The dephasing dynamics (characterized by a time scale  $1/\gamma_2 \sim 1$   $\mu$ s – 1 ns) of the electron and hole coherences  $\langle 1|\hat{\rho}|0\rangle$  and  $\langle 0|\hat{\rho}|1\rangle$ , including  $\rho_{1\mathbf{m},0\mathbf{m}}$ ,  $\rho_{1\mathbf{m}+1\mathbf{j},0\mathbf{m}}$ , and  $\rho_{0\mathbf{m}+1\mathbf{j},1\mathbf{m}}$ .

(ii) The relaxation dynamics (characterized by a time scale  $1/\gamma_1 \sim 1$  ms – 1 ns) of the electron and hole population  $\langle 0|\hat{\rho}|0\rangle - \langle 1|\hat{\rho}|1\rangle$ , including  $\rho_{0\mathbf{m}+1\mathbf{j},0\mathbf{m}} - \rho_{1\mathbf{m}+1\mathbf{j},1\mathbf{m}}$  and  $\rho_{0\mathbf{m},0\mathbf{m}} - \rho_{1\mathbf{m},1\mathbf{m}}$ .

(iii) The dephasing dynamics (characterized by a time scale  $T_2^N \sim 1$  ms)<sup>24</sup> of the first-order off-diagonal coherences  $\text{Tr}_{eh}(\mathbf{m} + 1\mathbf{j}|\hat{\rho}|\mathbf{m}) = \rho_{0\mathbf{m}+1\mathbf{j},0\mathbf{m}} + \rho_{1\mathbf{m}+1\mathbf{j},1\mathbf{m}}$  due to the fluctuation of the electron spin (through the electron-nuclear interaction) or the fluctuation of surrounding nuclear spins (through the nuclear-nuclear interaction).

Therefore, we can identify  $\{P_{\mathbf{m}',\mathbf{m}'}(t)\}$  as slow variables and other density matrix elements as fast variables and then apply the adiabatic approximation, which assumes that the responses of the fast variables to the slow variables are instantaneous [this introduces an error of the order  $O(\dot{P}) = O(a_{\text{nd}}^2)$ ]:

*Step 1.* In the equations of motion of the fast variables, the slow variables  $\{P_{\mathbf{m}',\mathbf{m}'}(t)\}$  are regarded as constants and the steady-state responses  $\rho_{1\mathbf{m},0\mathbf{m}}^{(\text{sr})}(\{P_{\mathbf{m}',\mathbf{m}'}(t)\})$ ,  $\rho_{1\mathbf{m}+1\mathbf{j},0\mathbf{m}}^{(\text{sr})}(\{P_{\mathbf{m}',\mathbf{m}'}(t)\})$ , ... of the fast variables as functions of  $\{P_{\mathbf{m}',\mathbf{m}'}(t)\}$  are obtained by setting their derivatives to zero.

*Step 2.* In the equation of motion Eq. (B3) of the slow variables  $\{P_{\mathbf{m}',\mathbf{m}'}\}$ , we replace the fast variable  $\rho_{1\mathbf{m}+1\mathbf{j},1\mathbf{m}}(t)$  by its steady-state response  $\rho_{1\mathbf{m}+1\mathbf{j},1\mathbf{m}}^{(\text{sr})}(\{P_{\mathbf{m}',\mathbf{m}'}(t)\})$ , so that Eq. (B3) becomes an effective equation of motion for the slow variables  $\{P_{\mathbf{m}',\mathbf{m}'}\}$ :

$$\dot{P}_{\mathbf{m},\mathbf{m}} = 2 \sum_j a_{j,\text{nd}} [\eta_{m_j} \text{Im} \rho_{1\mathbf{m}+1\mathbf{j},1\mathbf{m}}^{(\text{sr})}(\{P_{\mathbf{m}',\mathbf{m}'}(t)\}) - (\mathbf{m} \rightarrow \mathbf{m} - 1\mathbf{j})]. \quad (\text{B4})$$

Following the above prescription, under the typical condition  $|\omega_N| \gg |a_d|$ , straightforward calculation yields Eq. (1). When the back action is dropped, we obtain Eqs. (2) and (3), with  $c_1 \equiv 1 + [\gamma_1/(2\gamma_2)]f + W/\gamma_1$ ,  $f \equiv (\gamma_2^2 - \Delta^2)/(\gamma_2^2 + \Delta^2)$ ,  $c_0 \equiv 1/2 + \gamma_2/\gamma_1 + f + W/\gamma_1$ , and  $F \equiv [\gamma_1/(2\gamma_2)](c_0/c_1)$  being always positive since  $c_0$  and  $c_1$  are always positive.

### APPENDIX C: DERIVATION OF EQ. (4)

This is achieved straightforwardly by substituting Eq. (1) into the equation of motion  $\dot{p}(s,t) = \text{Tr} \delta(\hat{s} - s) \hat{P}(t)$ . The

result is

$$\begin{aligned} \frac{\partial}{\partial t} p(s,t) = & -NI \{ W_+(\Delta - h_{\text{max}}s) \text{Tr} \delta(\hat{s} - s)(\hat{K} - s) \hat{P}(t) \\ & - W_+[\Delta - h_{\text{max}}(s - a)] \text{Tr} [\hat{K} - (s - a)] \\ & \times \delta[\hat{s} - (s - a)] \hat{P}(t) \} \\ & - NI \{ W_-(\Delta - h_{\text{max}}s) \text{Tr} \delta(\hat{s} - s)(\hat{K} + s) \hat{P}(t) \\ & - W_-[\Delta - h_{\text{max}}(s + a)] \text{Tr} [\hat{K} + (s + a)] \\ & \times \delta[\hat{s} - (s + a)] \hat{P}(t) \}, \end{aligned}$$

where  $a \equiv 1/(NI)$  is the change of  $\hat{s}$  by each nuclear spin flip, and  $\hat{K} \equiv 1/(NI) \sum_j (\hat{I}_{j,x}^2 + \hat{I}_{j,y}^2)$ . For  $I = 1/2$ ,  $\hat{K}$  reduces to unity. For  $I \geq 1/2$  but weak nuclear polarization  $|s_0^{(I)}| \ll 1$ , the transverse fluctuation of each individual nuclear spin is not significantly influenced by the nuclear spin polarization, so we can approximate  $\hat{K}$  by a constant  $2(I + 1)/3$ . In either case, we obtain a closed equation for  $p(s,t)$ ,

$$\begin{aligned} \frac{\partial}{\partial t} p(s,t) = & -[G_+(s)p(s,t) - G_+(s - a)p(s - a,t)] \\ & - [G_-(s)p(s,t) - G_-(s + a)p(s + a,t)], \end{aligned}$$

where  $G_{\pm}(s) \equiv NI W_{\pm}(\Delta - h_{\text{max}}s)[2(I + 1)/3 \mp s]$ . For  $N \gg 1$  and hence  $a \ll 1$ , we expand the above equation up to the second order of the small quantity  $a$  and obtain the Fokker-Planck equation

$$\frac{\partial}{\partial t} p(s,t) = \frac{\partial}{\partial s} \left[ \frac{\partial}{\partial s} D(s)p(s,t) - v(s)p(s,t) \right],$$

where  $D(s) = (a^2/2)[G_+(s) + G_-(s)]$  is the diffusion coefficient and  $v(s) = a[G_+(s) - G_-(s)]$  is the drift coefficient. Note that  $D(s) = O(a^2)$  is much smaller than  $v(s)$  and  $\partial D(s)/\partial s$ , which are of the order  $O(a)$ . The steady-state solution is

$$p^{(\text{ss})}(s) = \frac{D(s^*)}{D(s)} p^{(\text{ss})}(s^*) \exp \left( \int_{s^*}^s \frac{v(s')}{D(s')} ds' \right),$$

where  $s^*$  is an arbitrary constant. Here  $v(s)$  vanishes at each stable state  $s_{\alpha}^{(\text{ss})}$ , so we expand  $v(s)$  around  $s_{\alpha}^{(\text{ss})}$  to the first-order  $v(s) \approx [dv(s)/ds]_{s=s_{\alpha}^{(\text{ss})}}(s - s_{\alpha}^{(\text{ss})})$  and obtain a Gaussian distribution [the influence of the factor  $D(s_{\alpha}^{(\text{ss})})/D(s)$  on the shape of the distribution is negligible for our case]

$$p^{(\text{ss})}(s) \approx \frac{D(s_{\alpha}^{(\text{ss})})}{D(s)} p^{(\text{ss})}(s_{\alpha}^{(\text{ss})}) \exp \left( -\frac{(s - s_{\alpha}^{(\text{ss})})^2}{2\sigma_{\alpha}^2} \right),$$

with a standard deviation

$$\sigma_{\alpha} = \sqrt{\frac{D(s_{\alpha}^{(\text{ss})})}{|dv(s)/ds|_{s=s_{\alpha}^{(\text{ss})}}}}.$$

For  $I = 1/2$ , the above equation coincides with previous theories.<sup>9,10,25</sup> By substituting the explicit expressions of  $D(s)$  and  $v(s)$  into the above equation, we obtain Eq. (4).

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