Great experimental progress in the last decade enabled the concentration, initialization, and readout of single spins in quantum dots [1]. Controlled coherent single-spin rotations—a key ingredient for quantum manipulation—were demonstrated recently using the electron spin resonance (ESR) [2–6]. The weak hyperfine coupling of the electron spin to the nuclear spins in the host material appeared to be of great importance in this field. It was identified as the main source of qubit decoherence and provides a significant hybridization of the spin states [7,8]. This has stimulated extensive theoretical and experimental research focusing on nuclear spin dynamics in quantum dots [9–14].

Overhauser pointed out already in the 1950s [15] that ESR may provide the buildup of significant nuclear spin polarization. Indeed, most ESR experiments on quantum dots, aimed at demonstrating electron spin rotations, also clearly demonstrated dynamical nuclear spin polarization (DNSP) [2–5].

For ESR driving of a single spin in an almost isolated quantum dot, or an ensemble of such dots, the scenario is similar to that of the usual Overhauser effect: The direction of DNSP is parallel to the spin of the excited electrons [15,16]. Recent ESR experiments on self-assembled quantum dots have confirmed this picture [5], and a similar reasoning holds for spin experiments with optically pumped dots [17]. In some cases, a bistability has been observed: Under the same conditions, the nuclear spins in the dot can be either polarized or unpolarized [11].

However, several issues can complicate the situation. In recent ESR experiments in double quantum dots [2–4] (i) electrons participate in transport during ESR driving, and (ii) there can be different nuclear spin dynamics in the two dots. Furthermore, a driving magnetic field is in practice accompanied by an electric field which modulates the electron-nuclear spin coupling at the resonance frequency [4]. All this makes a straightforward extension of existing models [16] impossible and promises richer and more interesting physics, which we indeed reveal.

In this Letter, we report a study of ESR in a double quantum dot focusing on DNSP. We have observed multiple stable states of nuclear polarization (up to four states), not seen in single-dot experiments, nuclear self-tuning to the ESR condition over a large range of magnetic fields (≥100 mT), and a sign of DNSP opposite to that following from the Overhauser argument. We identify the most probable mechanism governing DNSP and present a theoretical model explaining our findings. The results reported enable applications of this self-tuning effect, including accurate manipulation and control of the nuclear polarization [18] and use of this for improving the electron spin coherence time, possibly by orders of magnitude.

The double quantum dot system is electrostatically defined in a two-dimensional electron gas, located 90 nm below the surface of a GaAs/AlGaAs heterostructure, by applying negative voltages to metal surface gates. The dots are tuned to the Pauli spin blockade regime [19], where the transport sequence of charge states is \((1,1) \rightarrow (0,2) \rightarrow (0,1) \rightarrow (1,1)\), \((n, m)\) denoting the charge state with \(n(m)\) excess electrons in the left(right) dot. The current through the double dot depends on the spin orientation of the electrons in the \((1,1)\) state since the only accessible \((0,2)\) state is a spin singlet (Fig. 1).

Magnetic spin resonance is achieved by sending an alternating current through a coplanar stripline (CPS) which lies on top of the surface gates, separated by a thin dielectric layer. This current produces a small oscillating magnetic field \(B_1 \approx 1\) mT perpendicular to the external magnetic field \(B_0 \approx 100\) mT. The experimental data are obtained with the same device and in the same measurement run as the data presented in Ref. [2]. The difference is that the device is tuned to a higher interdot tunnel coupling and coupling to the right lead.

When we apply a continuous wave RF current with fixed frequency \(\omega\) to the CPS and sweep the external magnetic field \(B_0\) passing the resonance condition \(B_0 = B_{\text{res}} = \hbar \omega / g \mu_B\), we make a remarkable observation. One would expect that the resonance manifests itself as a peak in the current [2]. Indeed, if the external field is swept from low to high values, the current jumps up upon achieving the resonance condition. Unexpectedly, this resonant response extends over a wide range of magnetic fields, that exceeds \(B_{\text{res}}\) by a factor of 2 [see Fig. 2(a) upper panel]. If the field...
magnetic field $B_N$ acting on the electron spin [15]. Where high current is observed in the hysteresis region, this extra field is such that the total field $B_0 + B_N = B_{\text{res}}$, i.e., the nuclear field “tunes” the system to the resonance condition [16]. Low current indicates that $B_0 + B_N$ significantly deviates from $B_{\text{res}}$. The nuclei are unpolarized. Both polarized and unpolarized states are stable in the interval of hysteresis. Fluctuations of any kind could provide spontaneous switching between stable states, leading to the RTS.

A number of experimental details does not fit into this simple picture. First, there are multiple values of the current observed, three are clearly visible in Fig. 2(b) (labeled A–C). This implies multiple stable states of nuclear polarization with a total field close to $B_{\text{res}}$. Actually, we think that the RTS traces provide evidence for the existence of a fourth state. There is a number of current dips observed (labeled D) too big to be statistical fluctuations. We interpret those dips as signatures of a fourth state that decays on the scale of a second, i.e., different from state A, which decays on a larger time scale. Second, switching between the different current levels is rather fast. The nuclear spin dynamics are known to be slow, with a typical relaxation time $\tau_n \sim 15$ s [7,12,13]. If the current is a direct measure of the nuclear polarization, then why is the duration of the switching events so short? A third point is the sign of the polarization. Usually, in ESR experiments the dominating mechanism of DNSP is described by the Overhauser effect: The ESR excitation drives the electron spin(s) out of equilibrium, and hyperfine-induced electron-nuclear spin exchange is one of the mechanisms contributing to electron spin relaxation. As reasoned by Overhauser, on grounds of spin conservation, the direction of nuclear polarization should be parallel to the spin of excited electrons, whatever its orientation is with respect to the magnetic field applied.

This is the case for most DNSP experiments, e.g., [2,5,11]. Given the negative $g$ factor and positive hyperfine coupling in GaAs [20], this would give a $B_N$ parallel to $B_0$ [16]. In

![FIG. 1 (color online). Double dot setup. (a) The double quantum dot is coupled to two leads. Because of a voltage bias, electrons can only run from the left to the right lead. (b) Energy diagram. The four possible (1,1) states differ in spin projections on the quantization axes (red arrows). Under ESR conditions the axes can be different in the two dots and do not coincide with the direction of the external magnetic field. These states are coherently coupled (green arrow) to the (0,2) singlet that decays quickly (broadened line), leaving the system in (0,1).](image)

Another unexpected observation is made at fixed $B_0 \approx B_{\text{res}}$. Instead of a single value of the current corresponding to the maximum value of the ESR satellite peak, we observe clearly distinguishable multiple stable values of the current. Switching between these values gives rise to a random telegraph signal (RTS) with time scales ranging from seconds to minutes. Typical time-resolved measurements of the RTS are presented in Fig. 2(b) for three different values of the energy level detuning $\Delta_{LR}$ (Fig. 1).

We associate both the hysteresis and RTS with DNSP induced by the nonequilibrium electron spin dynamics under conditions of ESR and transport in the dots. Nuclear polarization is known to provide an effective

![FIG. 2 (color online). (a) Magnetic field sweeps for $\omega$ fixed at 350 MHz. Upper panel: Magnetic field sweep from low to high values resulting in an ESR peak width exceeding 100 mT. Lower panel: Sweep in the opposite direction, showing a much narrower ESR peak [23]. The nominal resonance condition $B_{\text{res}} = \hbar \omega / g \mu_B$ is met at $B_0 = 71$ mT for $\omega = 350$ MHz and $g = 0.35$ [2] (see dashed line). Note that in both traces the nuclear bath is unpolarized at the onset of electron spin resonance [22]. (b) Multiple values of the current through the double dot approximately at resonance. The current switches between at least three stable states on a time scale of seconds to minutes. The three panels correspond to three different values of the energy level detuning $\Delta_{LR}$ (increasing from the bottom to the upper panel). The values given for $\Delta_{LR}$ may have a constant offset, as photon assisted tunneling processes broaden the interdot transition which makes it difficult to separate resonant and inelastic transport. In both (a) and (b) the lowest value of current was subtracted as offset. The data in (b) were taken for a larger $\Gamma_{\text{in}}$ and $\Gamma_{\text{out}}$ than the data in (a).](image)
our experiment, its direction is clearly opposite, as high current is seen for B_0 > B_{res}. All three points are captured by the theory given below.

The electron spin \( \mathbf{S} \) and nuclear spins \( \mathbf{I}_k \) in each dot are coupled by hyperfine interaction [20] \( \mathbf{H}_{hf} = \frac{1}{2} \sum_k A_k (2\mathbf{S} \cdot \mathbf{I}_k + \mathbf{S^+} \mathbf{I}_k + \mathbf{S^-} \mathbf{I}_k^-) \), where the sum runs over \( N \approx 10^8 \) nuclei. The energy \( A_k \) is proportional to the probability to find the electron at the position of nucleus \( k \), \( A_k \approx 10^{-10} \) eV. With an external field applied in the \( z \) direction, the “flip-flop” terms \( \mathbf{S} \cdot \mathbf{I}_k \) provide spin exchange between the electrons and nuclei. Owing to energy conservation, these transitions must be second-order processes involving a mechanism supplying or absorbing the excess Zeeman energy. Conventionally, the spin exchange is due to the time-independent hyperfine coupling \( A_k \). However, as recently has been pointed out [2,4], in this setup a significant ac electric field moves the electrons in the dots with respect to the nuclei. This can be accounted for by introducing a time-dependent component in the hyperfine coupling \( A_k \rightarrow A_k + A_k e^{i\omega t} + A^*_k e^{-i\omega t} \). We estimate that under the present conditions \( A_k / A_0 \approx 0.1 \) [4,21].

We have considered six candidate mechanisms for DNSP [22], assuming a saturated ESR. We concluded that the dominant one involves the time-dependent hyperfine coupling, which allows for photon assisted flip-flops. These flip-flops do not have a preferred direction set by a nuclear spin, their scale set by \( \hbar / C_{0,2} \), much faster than the sweep rate in Fig. 2(a), [4,21]. The functions \( P \) and \( R \) are dimensionless functions giving the functional dependence of the resonant nuclear spin pumping (P) and resonantly enhanced nuclear spin relaxation (R) on the mixing angles and on \( \Gamma_\perp / \xi \Gamma_\parallel \), and have a maximum \( \sim 1 \). While \( R \) is roughly Lorentzian shaped, the function \( P \) is zero far from resonance \( \theta \rightarrow \{0, \pi\} \), reaches maximum at \( |f| \approx \tilde{B} \), and falls off to zero again at the resonance \( \theta = \pi/2 \). This resonant dip is due to the vanishing of electron spin polarization at the saturated resonance. In Eq. (1), the terms proportional to \( -B_N \) give nuclear spin relaxation: The first term presents the usual \( \tau_n \) while the second term gives a resonant enhancement owing to spin exchange with electrons. Nuclear spin pumping is given by \( \Gamma_1 B_{ov} P \sim 50 \) mT/s, much faster than the sweep rate in Fig. 2(a), with a sign opposite to that following from the Overhauser reasoning: Spin exchange under conditions of electron transport is mostly due to electrons polarized along the

\[
\frac{dB_N}{dt} = -\Gamma_1 B_{ov} P(\theta_{L,R}) - \left\{ \frac{1}{\tau_n} + \Gamma_2 R(\theta_{L,R}) \right\} B_N, \tag{1}
\]

and the equation for \( B_N^R \) is obtained by permutation of \( L \) and \( R \). The field \( B_{ov} \) is the Overhauser field of full polarization, \( B_{ov} = 5 \) T for GaAs. The functions \( P \) and \( R \) are dimensionless functions giving the functional dependence of the resonant nuclear spin pumping (P) and resonantly enhanced nuclear spin relaxation (R) on the mixing angles and on \( \Gamma_\perp / \xi \Gamma_\parallel \), and have a maximum \( \sim 1 \). While \( R \) is roughly Lorentzian shaped, the function \( P \) is zero far from resonance \( \theta \rightarrow \{0, \pi\} \), reaches maximum at \( |f| \approx \tilde{B} \), and falls off to zero again at the resonance \( \theta = \pi/2 \). This resonant dip is due to the vanishing of electron spin polarization at the saturated resonance. In Eq. (1), the terms proportional to \( -B_N \) give nuclear spin relaxation: The first term presents the usual \( \tau_n \) while the second term gives a resonant enhancement owing to spin exchange with electrons. Nuclear spin pumping is given by \( \Gamma_1 B_{ov} P \sim 50 \) mT/s, much faster than the sweep rate in Fig. 2(a), with a sign opposite to that following from the Overhauser reasoning: Spin exchange under conditions of electron transport is mostly due to electrons polarized along the

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Equation (1) determines the interval of hysteresis as been observed in experiment [18]. When subsequently third unpolarized stable state (Fig. 3, blue curve), giving as isolated from the other stable states: If the system switches to e, it will never switch back [see Fig. 4(c)]. This also has been observed in experiment [18]. When subsequently sweeping back from high to low field, the barrier for switching back from e to a high-current state is again gradually lowered. When the typical switching time becomes comparable to the time scale of the sweep, the current switches to a high value [Fig. 2(a), lower panel].

From Eq. (1) we construct a Fokker-Planck equation to study the stochastic properties of the polarizations in more detail [16,22]. Importantly, due to the accelerated dynamics, the fluctuations around all polarized states are suppressed as \( \langle (\Delta B_N) \rangle^2 / \Omega^2 = (B_1 / B_N^{\text{max}})^2 \), \( \Omega^2 ≡ (A_1 / g \mu_B)^2 N \) being the field variance in the unpolarized state.

To conclude, we have observed multiple nuclear polarization states and locking of the ESR condition over a large range of magnetic fields in a double quantum dot under ESR. We presented a theoretical model that captures the existence of these phenomena and their unusual features as fast switching and a ”wrong” sign of DNSP.

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[23] Another peak is observed at zero field which manifests the mixing of the four (1, 1) states by the fluctuating nuclear fields [2,7]. An offset of ~7 mT of \( B_0 \) due to the superconducting magnet is compensated for in both traces.