Multielectron semiconductor quantum dots (QDs) provide a novel platform to study the Coulomb interaction-driven, spatially localized electron states of Wigner molecules (WMs). Although Wigner-molecularization has been confirmed by real-space imaging and coherent spectroscopy, the open system dynamics of the strongly correlated states with the environment are not yet well understood. Here, we demonstrate efficient control of spin transfer between an artificial three-electron WM and the nuclear environment in a GaAs double QD. A Landau–Zener sweep-based polarization sequence and low-lying anticrossings of spin multiplet states enabled by Wigner-molecularization are utilized. Combined with coherent control of spin states, we achieve control of magnitude, polarity, and site dependence of the nuclear field. We demonstrate that the same level of control cannot be achieved in the non-interacting regime. Thus, we confirm the spin structure of a WM, paving the way for active control of correlated electron states for application in mesoscopic environment engineering.
However, most studies have focused on the spectroscopic confirmation of WM formation, and studies on the open system dynamics using correlated states have not been reported to date.

Here, we demonstrate the formation of a WM in semiconductor QDs, which helps achieving efficient spin environment control. We use gate-defined QDs in GaAs and exploit the quenched energy spectrum of the WM ($E_{ST} \sim 0.9 \text{ h-GHz}$) to enable mixing between different spin subspaces within $B_0 < 0.3 \text{ T}$. Furthermore, we demonstrate DNP by pulsed-gate control of the electron spin states. Leakage spectroscopy and Landau–Zener–Stuckelberg (LZS) oscillations confirm a sizable bidirectional change in $B_{\text{nuc}} \sim 80 \text{ mT}$ and the spatial Overhauser field gradient $\Delta B_{\text{nuc}} \sim 35 \text{ mT}$ due to the long nuclear spin diffusion time $T_N \sim 62 \text{ s}$. Further, we demonstrate on-demand control of $B_{\text{nuc}}$ combined with coherent LZS oscillations, providing a new route for realizing controllable DNP using correlated electron states.

**Results**

Figure 1a shows a gate-defined QD device fabricated on a GaAs/AlGaAs heterostructure, where a 2D electron gas (2DEG) is formed 70 nm below the surface (see Methods). We focus on the left double QD (DQD) containing three electrons. We designed the $V_2$ gate to form an anisotropic potential, which is predicted to promote WM formation. An electrostatic simulation of the electric potential at the QD site near $V_2$ shows an oval-shaped confinement potential with anisotropy.

**Fig. 1** | Wigner molecule formation in a GaAs double quantum dot. a Scanning electron microscope image of a GaAs quantum dot (QD) device similar to the one used in the experiment. Green dots denote the double QD defined for Wigner molecule (WM) formation which is aligned along the [110] crystal axis (black arrow). The inner plunger gate $V_2$ is designed to have anisotropic confinement potential as shown in the right panel to facilitate the localization of the electronic ground state. Yellow circle: a radio-frequency (rf) single-electron transistor (rf-SET) charge sensor for rf-reflectometry. External magnetic field $B_0$ is applied along the direction denoted by the yellow arrow. b Charge stability diagram of the double QD near the three-electron region spanned by $V_1$ and $V_2$ gate voltages. Green-shaded region: the energy-selective tunneling (EST) position for the state readout and initialization. c Landau–Zener–Stückelberg (LZS) oscillation of the WM at $B_0 = 0 \text{ T}$. The relative phase evolution between the excited doublet ($D_T$) and the ground doublet ($D_S$) results in the oscillation captured by the EST readout. Red-dashed curve in the fast Fourier transformed (FFT) map shows energy dispersion calculated from the toy-model Hamiltonian. The calculation yields quenched orbital energy spacing of the inner dot $\delta_L \sim 100 \text{ h-GHz}$ and the ground doublet ($D_S$), results in the oscillation captured by the EST readout. Red-dashed curve in the fast Fourier transformed (FFT) map shows energy dispersion calculated from the toy-model Hamiltonian. The calculation yields quenched orbital energy spacing of the inner dot $\delta_L \sim 0.9 \text{ h-GHz}$.
exceeding 3 (Fig. 1a, right panel). This potential can be tuned by the gate voltage, allowing the controlled electron correlation and localization of the ground state wavefunction within the DQD.\textsuperscript{[24, 26, 27]} The yellow dot in Fig. 1a denotes a radio-frequency single-electron transistor (rf SET) charge sensor utilized for quantum state readout.\textsuperscript{41} The device was operated in a dilution refrigerator with a base temperature of \(-40\ \text{mK}\), an electron temperature \(T_e = 150\ \text{mK}\) (Supplementary Note 1), and a variable \(B_0\) applied to the direction shown in Fig. 1a.

The three-electron DQD results in two spin doublets and one spin quadruplet state. Without the magnetic field in the (2,1) [(1,2)] charge configuration, doublet-singlet states \(D_s(2,1)\) \([D_s(1,2)]\) with total spin \(S = 1/2\) form the ground state where the two electrons in the left [right] QD form a spin singlet state and fill the orbital ground in the left [right] QD. Here, \(n\) \([m]\) denotes the number of electrons in the left [right] QD by \(n\). When the two electrons in the left [right] QD form a spin triplet state and fill the excited orbital in the left [right] QD, the three-electron state result in either the doublet-triplet state \(D_t(2,1)\) \([D_t(1,2)]\) with \(S = 1/2\) or the quadruplet state \(Q_s(2,1)\) \([Q_s(1,2)]\) with \(S = 3/2\). Because of the orbital splitting, the \(D_t\) and \(Q_s\) states usually have higher energy compared to \(D_s\) states.\textsuperscript{[14, 16]} If a finite magnetic field is applied, doublet states with \(S = 1/2\) are split into \(n_s = 1/2, +1/2\) and \(S = 2\) states whereas quadruplet states with \(S = 3/2\) are split into \(n_s = -1/2, +1/2, -3/2, +3/2\) states. Here \(n_s\) is the spin quantum number related to the \(z\) component of the electron spin angular momentum. The explicit spin structures are shown in the Methods. Hereafter, \((n, m; \Delta s, \Delta m)\) notation is used to describe both the charge configuration and spin angular momentum of a state.

First, we show the spectroscopic evidence of the WM at \(B_0 = 0\ \text{T}\) by probing \(E_{ST}\) in the right QD \(S\). Figure 1b shows a charge stability diagram around (2,1) and (1,2). The green shaded region near the boxes, followed by adiabatically driving the state through the anti-crossing, which can be tuned by the nuclear spin \(I = 1\). Unlike the electrons in GaAs, nuclei have positive g-factors.\textsuperscript{[20]} Therefore, the \(\Delta n_{\text{nuc}}\) polarization effect is still visible. Thus, \(n_s > 10\ \text{s}\), as discussed below. Moreover, as the anti-crossing position is a sensitive function of \(B_{\text{nuc}} = B_0 + B_{\text{nuc}}\) over 100–300 mT, it can be used to measure \(B_{\text{nuc}}\).

We now show bidirectional DNP combined with coherent control of doublet states at \(B_0 = 230\ \text{mT}\). Figure 3a (top panel) shows the three primary paths through the anticrossings, which can flip the electron spins deterministically by adiabatic passage.\textsuperscript{[26, 27, 31]} Paths \(P_1\) and \(P_3\) describe the S-polarization that flips the electron spin with \(\Delta n_{\text{nuc}} = +1\). This is enabled by initializing the state to \(D_t(1,2; 1/2)\) \([Q_s(1,2; 3/2)]\) at the EST position and then by non-adiabatically pulsing beyond the first anticrossings near the (2,1) charge configuration (Fig. 3a, yellow boxes), followed by adiabatically driving the state through the anticrossing to \(Q(1,2; 1/2)\) \([Q(1,2; 3/2)]\), which accompanies \(\Delta n_{\text{nuc}} = -1\) (Fig. 3a, blue arrows). Because the spin state is initialized to the doublet-singlet state before the adiabatic spin-flip passage, the sequence is named S-polarization. The \(Q(1,2; 1/2)\) \([Q(1,2; 3/2)]\) state is diabatically driven back to the EST position, and one electron quickly tunnels out to the reservoir. Reloading an electron from the reservoir reinitializes one of the \(D_s\) states completing the polarization cycle. Both the \(D_t(1,2; -1/2)\) and \(D_t(1,2; 1/2)\) initial states contribute to the S-polarization. Path \(P_2\) denotes the T-polarization (\(\Delta n_{\text{nuc}} = -1, \Delta n_{\text{nuc}} = +1\)).
which is possible by driving \( D_T(1,2;1/2) \) adiabatically to \( D_S(1,2;−1/2) \) (Fig. 3a, red arrow). To prepare \( D_T(1,2;1/2) \), we apply a \( π \)-pulse to \( D_S(2,1;1/2) \) before the adiabatic passage (Fig. 3a, bottom panel).

Because we prepare the doublet-triplet state before the adiabatic spin-flip passage, the sequence is called T-polarization. The T-polarization is possible only when the state is initialized to \( D_S(2,1;1/2) \) at the EST position. Combining the S- and T-polarizations, we measure the change in \( B_{\text{nuc}} \) (\( δB_{\text{nuc}} \)), where the repeated polarization pulse sequence (Fig. 3a, bottom panel) with variable \( τ_{\text{evol}} \) and a repetition rate of \( ~20 \text{ kHz} \) is applied for \( 10 \) s before each line scan. For Fig. 3b, a waiting time \( ~10 \) min was added after each sweep to allow the polarized nuclei to diffuse and minimize the polarization effect in the next sweep. As shown in Fig. 3b, \( δB_{\text{nuc}} \) oscillates with \( τ_{\text{evol}} \), which is anti-

![Fig. 2 | Leakage spectroscopy and probabilistic nuclear polarization with the Wigner molecule.](https://i.imgur.com/3jz5.jpg)

**Fig. 2 | Leakage spectroscopy and probabilistic nuclear polarization with the Wigner molecule.** **a** Left panel: schematics of the energy levels for different external magnetic fields \( B_0 > 0 \text{ T} \). Crossings between different \( m_S \) states become anticrossings aided by the transverse nuclear Overhauser field. Right panel: schematic of the pulse sequence for leakage spectroscopy and probabilistic dynamic nuclear polarization (DNP). The pulse diabatically drives the initialized \( D_S(2,1;1/2) \) [\( D_S(2,1;−1/2) \)] to \( (1,2) \), and hold \( r \) for \( 100 \text{ ns} ≫ T_2^* \). Upon the coincidence of the pulse detuning and the anti-crossing, the state probabilistically evolves to \( Q(1,2;3/2) \) [\( Q(1,2;1/2) \)] and flips the electron spin \( Δm_S = +1 \) which accompanies \( Δm_N = −1 \). The scale bars on the bottom axis (\( ε \) axis) denote 50 μeV, and the scale bars on the left axis (Energy axis) denote 1 h·GHz. **b** Leakage spectroscopy of the Wigner molecule (WM) state as a function of \( B_0 \) and the pulse amplitude \( A_p \). Black (Red) dotted curve shows the calculated energy splitting between \( D_T \) (\( Q \)) and \( D_S \) at \( B_0 = 0 \text{ T} \). Measurement-induced nuclear field shifts the dispersion opposite to the direction of \( B_0 \). **c, d** Leakage measurement with an additional probabilistic polarization pulse with amplitude \( A_p' \) applied before each line sweep. The \( A_p' \) is fixed to 370 (450) mV, and the additional distortion in the leakage spectrum is shown as red circles near a pulse amplitude of 370 (450) mV. Black arrows denote the magnetic field sweep direction.
correlated with the LZS oscillation that represents the population of $D_1(1,2;1/2)$. This confirms that the net polarization rates can be controlled by adjusting $\tau_{\text{evol}}$. Accordingly, we calibrate $\tau_{\text{evol}} = 0$ (0.62 ns) for S (T)-polarization. We also calibrate the duration of the adiabatic spin transfer $\omega_R$. Figure 3c shows the maximum nuclear field change $B_{\text{max}}$ achievable as a function of $\omega_R$, where both S- and T-polarizations are ineffective for short $\omega_R$ because of negligible adiabatic transfer probability $P_{\text{Zener}}$. $B_{\text{max}}$ reaches a maximum around $\omega_R = 0.8$ ps, after which the maximum efficiency is retained for the S-polarization sequence. In the case of T-polarization, however, for long $\omega_R$, $B_{\text{max}}$ decreases because of $D_1$ relaxation during the adiabatic passage.

By tuning $\delta R$ via the dc gate voltages and performing similar S-polarization experiments, we find that $B_{\text{max}}$ decreases with increasing $\delta R$ (Fig. 3d, see Supplementary Note 4). As is discussed subsequently, we find that the nuclear diffusion time scale exceeds 60 s regardless of $\delta R$, but the Overhauser field change per electron flip $B_{\text{O}}$ is strongly suppressed with increasing $\delta R$. Ultimately, the observation implies that the pulsed-gate-based nuclear control becomes inefficient in the non-interacting regime. We suspect the degree of electronic waveform localization which depends on the Wigner parameter may be affecting the contact hyperfine interaction between the electron and the nuclear spins and altering the DNP efficiency as a result.

Returning to the condition $\delta R = 0.9$ hHz, we demonstrate on-demand DNP. Figure 3e, f shows the result of optimized S (T)-polarization with $\tau_{\text{evol}} = 0$ ns, $\omega_R = 1000$ ns ($\tau_{\text{evol}} = 0.62$ ns, $\omega_R = 600$ ns). Although the local fluctuations of the nuclear spins lead to random drift of the anti-crossing positions without the polarization pulse, $B_{\text{max}}$ builds toward (opposite to) the $B_0$ direction faster than the nuclear spin diffusion timescale when the polarization pulse is applied before each line scan. $B_{\text{max}}$ rises to $B_{\text{max}} = 80$ mT (–40 mT) until a dynamic equilibrium is reached. Because only the $m_s = 1/2$ states contribute to the T-polarization, $|B_{\text{max}}|$ for the T-polarization is about half of that for the S-polarization, implying that the state initialize to both $m_s$ states with nearly equal probability at the EST position.

We also demonstrate bidirectional DNP by adjusting $\tau_{\text{evol}}$ in Fig. 3g. Figure 3h illustrates the control of $B_{\text{max}}$ by adjusting the adiabatic sweep amplitude $A_0$ of the S-polarization sequence. Under the S-polarization, $B_{\text{max}}$ builds in the $B_0$ direction and drives the anti-crossing to deeper $\epsilon$ (more to (1,2) charge configuration). Because the pulse cannot have a finite polarization effect if the anti-crossing position is driven beyond $\epsilon$ reachable with $A_0$, $A_0$ serves as the limiting factor of $B_{\text{max}}$. Thus, a self-limiting DNP protocol, where the DNP field is limited by experimental parameters used in the pulse shape rather than the interpulse between pumping rate and nuclear diffusion, can be realized. This self-limiting property can be useful in future DNP experiments as the steady state DNP field can be simply controlled by adjusting the pulse amplitude.
Using a simple rate equation, we simulate the polarization-probe sequence (red-dashed curve in Fig. 3e, see Methods and Supplementary Note 5) and obtain $r_\Delta \approx 62$ s and $b_\Delta \approx 2.58 \times 10^{-9}$ kHz ($g\mu_B$) from the fit. In contrast, the DNP effect is negligible in our device with the two-electron ST$_0$ qubit$^9$ under the same repetition rate as in the WM regime (see Supplementary Note 6). We further find our DNP is still effective when the repetition rate is as low as 5 kHz (see Supplementary Note 7) showing that the Wigner molecule allows sizable DNP that cannot be achieved with conventional QDs. Through optimization of the magnitude and direction of $B_\Delta$, $b_\Delta \approx 3 \times 10^{-9}$ kHz ($g\mu_B$)$^5$ can be achieved with an ST$_0$ qubit in GaAs$^{6,8}$. However, the obtained result shows that robust nuclear control can be achieved with WMs even in the regime where the same level of control cannot be achieved with an ST$_0$ qubit. In addition, residual polarization $\sim 21.5$ mT exists after turning off the polarization sequence (Fig. 3e), which diffuses within $\sim 30$ min. The large Knight shift gradient originating from the non-uniformly broadened WM wavefunction may be a possible cause of the long $r_\Delta$. However, the newly observed phenomena in this study, including the dependence of $D_0$ on the tuning condition, require further investigations$^{7,9}$.

Furthermore, the WM’s coherent LZS dynamics provide a novel approach to measure the spatial Overhauser field gradient $\Delta B_z$ between QDs. When $\Delta B_z$ is larger than the exchange splitting between $D_1(1,2;1/2)$ and $Q_1(1,2;1/2)$, the eigenstates are expected to become $D_1(1,2;1/2)\Delta B_z \approx \{ |i\rangle \}$ and $D_0(1,2;1/2)\Delta B_z \approx \{ |f\rangle \}$. Because both states can tunnel-couple to $D_1(1,2;1/2)$ and $D_0(1,2;1/2)$, the LZS oscillation reveals the $D_1$–$D_0$ energy splittings. As can be inferred from the Hamiltonian (see Supplementary Note 8), although the $D_1$–$D_0$ splitting is independent of the $\Delta B_z$ and $B_z$, the $D_1$–$D_0$ splitting is modulated by $\Delta B_z$ depending on the sign of $\Delta B_z$ and $m_z$ providing the direct measure of $\Delta B_z$. Because the states can initialize to both $D_1(1,2;1/2)$ and $D_0(1,2;1/2)$ at the EST position, the LZS oscillation captures the dynamics of both $m_z=1/2$ and $m_z=-1/2$ subspaces.

Figure 4a (4b) illustrates the LZS oscillation measurement of the WM multiplet states at $B_\Delta = 230$ mT in the time (frequency) domain with the $S$-polarization turned on and off at specific laboratory times. The FFT spectrum exhibits three different branches corresponding to the $D_1$–$D_0$, $D_1$–$D_1$ (black and black-dashed arrows) where the $S$-polarization induces changes in $\Delta B_z$. Two different $D_1$–$D_0$ branches correspond to different $m_z$ subspaces, where the sign of $\Delta B_z$ should be known to distinguish the $m_z$ for each branch. The $D_1$–$D_1$ splitting is the same for both $m_z$ subspaces and is displayed as a single branch (red arrow). Figure 4c, d shows the simulated time (frequency) domain signal of the same LZS oscillation, which agrees well with the experimental result (see Supplementary Note 9). As expected, the $D_1$–$D_0$ splitting is constant regardless of $\Delta B_z$, whereas the $D_1$–$D_1$ splitting is modulated along the polarization sequence.

The $D_1$–$D_1$ splitting without the polarization sequence implies the built-in $\Delta B_z \approx 200$ kHz ($g\mu_B$)$^5$ (35 mT), which is also confirmed by the ST$_0$ oscillation (see Supplementary Note 6). $\Delta B_z$ increases to $400$ kHz ($g\mu_B$)$^5$ (70 mT) with the S-polarization and decreases to $200$ kHz ($g\mu_B$)$^5$ after turning off the polarization off. Thus, we conclude that the S-polarization yields the asymmetric pumping effect ($\Delta B_{nuc} \approx 200$ kHz ($g\mu_B$)$^5$) about the QD sites, whereas the $\Delta B_{nuc}$ direction can be experimentally checked, for example, via single-spin electric-dipole spin resonances$^{86}$. Furthermore, the $D_1$–$D_0$–$D_1$ splitting comprises the decoherence-free subspace for the qubit operations resilient to magnetic noises, where the coherent microwave control combined with the large polarization may enable leakage-free and state-selective transitions.

**Discussion**

The present work uncovers the spin and energy structure of the WM states and explores the central-spin problem with strongly correlated WM states in semiconductor QDs. With the energy splitting of the WM - 0.9 GHz, we confirm the controllable DNP of $B_{nuc}$ ($\Delta B_{nuc}$) reaching (but not limited to) 80 mT (35 mT) via leakage spectroscopy and LZS oscillations. The $T_1$ exceeds 60 s, which, together with bidirectional polarizability, is beneficial for stabilizing the nuclear bath fluctuation and realizing long-lived nuclear polarization$^{10,11}$.

We anticipate several developments for further developments and applications of WM-enabled DNP. Similar experiments with a larger $\delta T_1/T_1$ ratio can enable high-fidelity single-shot readout for a faster measurement of the dynamics of nuclear polarization. This would further enable feedback loop control$^{10}$ and tracking$^{12,15}$ of nuclear environments in multielectron QDs which can be utilized to narrow the nuclear field distribution for electron coherence enhancement. The real-time Hamiltonian estimation also improves frequency resolution for measuring instantaneous $\Delta B_{nuc}$, which may enable measurements of the degree of spatial localization within WMs.

We expect more asymmetric QD geometry would allow a smaller energy gap and thereby the broader electronic wavefunction. Along with the tunability of the energy gap via the gate voltage, as shown here, this may facilitate the investigation of the spatial noise characteristics within a single QD which has been impractical with the typical QD geometries. Furthermore, DNP becomes inefficient with increasing $E_{ST}$ of the WM, as discovered herein. This implies that the pulsed-gated electron–nuclear flip-flop probability is a strong function of the Wigner parameter, the microscopic origin of which requires more rigorous investigations.

**Methods**

**Device fabrication**

A quadruple QD device was fabricated on a GaAs/AlGaAs heterostructure with a 2DEG formed ~70 nm below the surface. The transport property of the 2DEG showed mobility $\mu = 2.6 \times 10^5$ cm$^2$/Vs with electron density $n = 4.0 \times 10^{11}$ cm$^{-2}$ at temperature $T = 7-4$ K. Electronic mesa around the QD site was defined by the wet etching technique, and thermal diffusion of a metallic stack of Ni/Ge/Au was used to form the ohmic contacts. The depletion gates were deposited on the surface using standard e-beam lithography and metal evaporation of 5 nm Ti/30 nm Au. The lithographical width of the inner QD along the QD axis direction was designed to be 10% wider than the outer dot to facilitate WM formation. The QD array was aligned to the [110] crystal axis, as shown in Fig. 1a. Although the magnetic field $B_\parallel$ was intended to be applied perpendicular to the [110] axis to minimize the effect of spin-orbit interaction, the angular deviation was not strictly calibrated.

**Measurement**

The device was placed on a -40 mK plate in a commercial dilution refrigerator (Oxford Instruments, Triton-500). Ultra-stable dc-voltages were generated by battery-powered dc-sources (Stanford Research Systems, SIM928). They were then combined with rapid voltage pulses from an arbitrary waveform generator (AWG, Keysight MS895A) with a sample rate up to 65 GSa/s via homemade wideband (10–100 GHz) bias tees to be applied to the metallic gate electrodes. An LC-tank circuit with a resonant radio frequency (rf) of ~120 MHz was attached to the ohmic contact near the SET charge sensor to enable high-bandwidth ($\Delta f_{\text{BW}} > 1$ MHz) charge detection$^{33,34,35,36}$. The reflected rf-signal was first amplified by 50 dB using two-stage low-noise cryo-amplifiers (Caltech Microwave Research, C1TL2 F <2 in series) at a 4 K plate. Next, it was further amplified by 25 dB at room temperature using a homemade low-noise rf-amplifier. The signal was then demodulated by an ultra-high-frequency lock-in amplifier (Zurich Instruments, UHFLI), which was routed to the boxcar integrator built in the UHFLI. Trigger signals with a repetition period of 51 ms were generated by a field-programmable-gate array (FPGA, Digilent, Zedboard) to synchronize the timing of the AWG and the boxcar integrator for the CDS$^{36}$. 

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Table 1 | Three-electron spin states

| State       | Spin structure                  |
|-------------|---------------------------------|
| Q(1,2; m_s=3/2) | |(↑)(↓)                  |
| Q(1,2; m_s=1/2) | +(√2/2)(↑)(↑)(↑)                  |
| Q(1,2; m_s=-1/2) | -(√2/2)(↑)(↑)(↑)                  |
| Q(1,2; m_s=-3/2) | +(√2/2)(↑)(↑)(↑)                  |
| D_(1,2; m_s=1/2) | |(↑)(S)                  |
| D_(1,2; m_s=-1/2) | |(↓)(S)                  |
| D_(1,2; m_s=-3/2) | |(↓)(S)                  |

Here, T<sub>0</sub>, T<sub>1</sub>, and T<sub>2</sub> denote the three triplet states (S = 1) with m<sub>s</sub> = 0, +1 and −1 respectively and S indicates the spin singlet state (S = 0).

Fig. 4 | Field gradient control and measurement. Landau–Zener–Stückelberg (LZS) oscillation of the Wigner molecule (WM) states at B<sub>0</sub> = 230 mT in a the time domain and b the frequency domain with the S-polarization sequence. The oscillation reveals the relative phase oscillation of the D<sub>11</sub> − D<sub>12</sub> (black arrow, black dotted arrow) and D<sub>10</sub> − D<sub>12</sub> (red arrow) of both the m<sub>s</sub> = 1/2 and m<sub>s</sub> = −1/2 states. The D<sub>10</sub> − D<sub>12</sub> splitting is constant regardless of the magnetic field gradient ∆B<sub>z</sub>, whereas the D<sub>11</sub> − D<sub>12</sub> energy spacing is modulated by the ∆B<sub>z</sub> depending on the sign of ∆B<sub>z</sub> and m<sub>s</sub>. The resultant beating is visible in e, f the time (frequency) domain line-cut when the polarization is on (green arrow in a) and off (blue arrow in a). The line cuts in the time domain are numerically fitted to the sum of three sine functions (solid lines in e) with different amplitudes. Three separate peaks are visible in the frequency domain (f) when the ∆B<sub>z</sub> is largely polarized in the bottom panel (blue line) in (f) Simulated LZS oscillation in e the time domain and d the frequency domain with the ∆B<sub>z</sub> in the inset of (d). The simulation in the frequency domain reproduces the branches shown in (b).

### Eigenstates of three-electron spin states

Three-electron spin-multiplet structure consists of eight different eigenstates, which are two D<sub>0</sub> states, two D<sub>T</sub> states, and four quadruplet states. For simplicity, we show only the spin states with (1,2) charge configuration in Table 1, where the spin state in the first (second) bracket indicates the single- (two-) electron spin state in the left (right) QD.

### Rate equation

Nuclear spin polarization and the diffusion process were phenomenologically modeled using a rate equation:

$$\frac{dB_{nuc}}{dt} = -\frac{B_{nuc}}{\tau_n} + b_0 P_{flip}/\tau_{rep},$$  (1)
where $\tau_N$ is the nuclear spin diffusion time, $\beta_N$ is the Overhauser field change per electron spin-flip, $P_{\text{sp}}$ is the nuclear spin flop probability obtained from the Landau–Zener transition probability $P_{\text{LZ}}$, and $T_{\text{rep}}$ is the pulse repetition period. Using Eq. (1), we simulated the polarization-probe sequence shown in Fig. 3 with the experimental parameters including the time required for the amplitude sweep in the leakage probe step.

### Data availability
The data that support the findings of this study are available from the corresponding author upon request.

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Author contributions
D.K. and W.J. conceived the project. W.J. performed the measurements and analyzed the data. J.K. and H.Jung fabricated the device. J.P., M.C., H.Jang, and S.S. built the experimental setup and configured the measurement software. G.K. and B.K. provided the numerical simulation of the Wigner molecules. V.U. synthesized and provided the GaAs heterostructure. All the authors contributed to the preparation of the manuscript.

Competing interests
The authors declare no competing interests.

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