Photogeneration of a single electron from a single Zeeman-resolved light-hole exciton with preserved angular momentum

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Quantum state transfer from a single photon to a single electron following selection rules can only occur for a spin-resolved light-hole excitation in GaAs quantum dots; however, these phenomena have yet to be experimentally realized. Here, we report on single-shot readout of a single electron spin via the Zeeman-resolved light-hole excitation using an optical spin blockade method in a GaAs quantum dot and a Pauli spin blockade method in a double GaAs quantum dot. The observed photoexcitation probability strongly depends on the photon polarization, an indication of angular momentum transfer from a single photon to an electron. Our demonstration will open a pathway to further investigation of fundamental quantum physics and applications of quantum networking technology.

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The selection rules of interband optical transitions in GaAs/AlGaAs heterostructures have long been studied, as they form the basis for the quantum interface between an absorbed photon polarization and a photoelectron spin created in the conduction band. In this process the quantum state must be preserved between the particles (quantum state transfer). It is known in GaAs quantum dots (QDs) that only Zeeman split sublevels of a light-hole (LH) exciton can generate a spin product state of an electron and a LH. This is not the case for a heavy-hole (HH) exciton (described later) [1]. Indeed coherence in the LH excitation process has been previously demonstrated using a Kerr rotation technique that optically measures the spin orientation [2]. Tomographic Kerr measurements were also performed for Zeeman-resolved LH excitons but only for large ensembles in GaAs quantum wells (QWs) [3,4]. Recently, photogeneration of a single electron in a laterally gated quantum dot (QD) has been achieved, but it featured neither the LH excitation nor spin readout [5–9]. To demonstrate the photon-to-spin quantum state transfer requires challenging experiments to directly read out the spin state of the photogenerated electron via the spin-resolved LH excitation.

Since a single electron spin in a QD is polarized along an external magnetic field, the initially trapped electron in the ground state results in Pauli spin blockade of the generated photoelectron, working effectively as a spin projection measurement along the field. The successive photogeneration of an electron with a spin parallel to the initially trapped electron is prohibited. (We refer to the prohibition under this condition as optical spin blockade). Photon absorption efficiency observed for a single QD (SQD) strongly depended on the polarizations of an incident laser, reflecting the optical spin blockade effect. In a preceding study, the blockade effect was indeed observed for photoelectrons excited by the spin-resolved HH excitation in a singly charged InGaAs/GaAs self-assembled QD [10]. However, that experiment did not demonstrate the quantum state transfer from a photon to a photoelectron, because photoexcitation process involving HH and electron pair can not provide a superposition state of two opposite spin orientations.

Here we perform spin-selective photoexcitation of HH and LH excitons in a laterally gated GaAs QD sample and use a charge sensing technique to measure the probability of finding single photoelectrons in the dot that are created in line with the optical selection rules. We first use a SQD having zero or one electron, $N_e = 0$ or 1, and pump just one electron in the dot by vertical (V) or horizontal (H) linearly polarized light. We show that the photoexcitation of a pair of an electron and one of the spin-resolved LHs is prohibited by optical spin blockade for the case of the photogenerated electron spin being parallel to the residing electron spin in the $N_e = 1$ SQD. Next, we compare the result with a spin readout method for the photoelectron with a double QD (DQD) [11]. We detect the spin orientation (up or down) of the photogenerated electron in the spin-resolved LH excitation using the electrical Pauli spin blockade effect. Finally, we show that the obtained results are consistent between the SQD and DQD experiments, supporting the angular momentum preservation via photoexcitation of an electron-LH spin product state.

In a Voigt geometry with an in-plane magnetic field, irradiation of linearly polarized photons selectively generates electrons with spins parallel ($|\rightarrow\rangle_e$) or antiparallel ($|\leftarrow\rangle_e$) to

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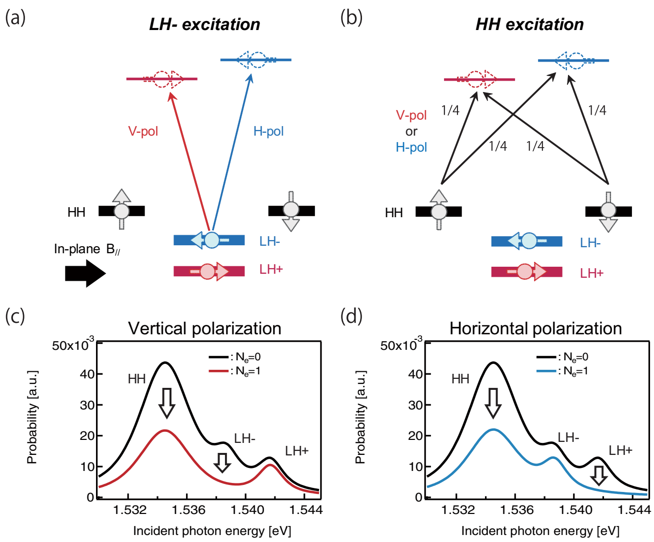

FIG. 1. (a),(b) Schematics of spin-selective optical transitions between the electron and hole states for both the LH and HH excitations. For the $N_e = 0$ dot, optical excitations of electron spins both parallel and antiparallel to an in-plane magnetic field are allowed. On the other hand, for the $N_e = 1$ dot an electron spin antiparallel to the magnetic field is initially trapped. In this configuration the optical transition to the spin parallel to the field is forbidden by the Pauli exclusion rule. (c),(d) Calculations of the photoelectron trapping probability in the SQD as a function of the incident photon energy. The peak shapes are estimated from the photoelectron trapping spectrum of the second wafer depicted in Fig. SI1(d) of the Supplemental Material [13]. N_e indicates the electron number initially trapped by the dot. The optical excitation with the V-polarized light from the upper Zeeman split LH state, LH−, is forbidden. On the other hand, for the lower Zeeman split LH state, LH+, the excitation with the H-polarized light is forbidden. For the HH excitation the trapping probability for the $N_e = 1$ SQD is suppressed to half the value for $N_e = 0$. The suppression of the trapping probability due to the optical spin blockade is indicated by the hollow arrows.

the magnetic field. Therefore, from a superposition state of two photon polarizations, $|\psi\rangle_{ph} = \alpha|H\rangle + \beta|V\rangle$ (H and V are linear polarizations which are perpendicular and parallel to the applied magnetic field, respectively), a simple product of a superposition state of $(\alpha|\leftarrow\rangle_e + \beta|\rightarrow\rangle_e)$ and a spin-resolved LH state, LH− : $|\Leftarrow\rangle_{lh}$ or LH+ : $|\Rightarrow\rangle_{lh}$ [see Fig. 1(a)], is generated:

$$\alpha|H\rangle + \beta|V\rangle \Longrightarrow (\alpha|\leftarrow\rangle_e + \beta|\rightarrow\rangle_e) \otimes |\Leftarrow\rangle_{lh}$$
$$[\text{or } (\alpha|\leftarrow\rangle_e - \beta|\rightarrow\rangle_e) \otimes |\Rightarrow\rangle_{lh}]. \quad (1)$$

For the LH excitation in the $N_e = 0$ SQD, which is represented by Eq. (1), the H(V)-polarized light generates an optical transition from LH− to $|\leftarrow\rangle_e$ ($|\rightarrow\rangle_e$) [see Fig. 1(a)]. On the other hand, for the $N_e = 1$ SQD, because the electron spin initially occupies the ground state of $|\rightarrow\rangle_e$, the optical transition to the spin state $|\rightarrow\rangle_e$ generated by the V-polarized light is forbidden by the optical spin blockade effect. Note that in contrast to the LH−, the polarization dependence is opposite for the LH+ excitation, i.e., the transition by the H-polarized light is forbidden for $N_e = 1$.

We first study optical spin blockade in single photoelectron generation by selective photoexcitation of an electron and HH or LH pairs in SQDs for $N_e = 0$ or 1. For the HH excitation the in-plane g factor is 0 in the case of crystal growth along the [001] direction [12]: both the up-spin $|\Uparrow\rangle_{hh}$ and down-spin $|\Downarrow\rangle_{hh}$ states contribute to the optical transition. For the linear polarized photons, the spin configuration of the excited electron-HH pair is therefore expressed as [see Fig. 1(b)]

$$\alpha|H\rangle + \beta|V\rangle$$
$$\Longrightarrow \frac{1}{\sqrt{2}}(\alpha+\beta)|\uparrow\rangle_e \otimes |\Uparrow\rangle_{hh} + \frac{1}{\sqrt{2}}(\alpha-\beta)|\downarrow\rangle_e \otimes |\Downarrow\rangle_{hh}$$
$$= \frac{1}{2}|\leftarrow\rangle_e \otimes [(\alpha+\beta)|\Uparrow\rangle_{hh} + (\alpha-\beta)|\Downarrow\rangle_{hh}]$$
$$+ \frac{1}{2}|\rightarrow\rangle_e \otimes [(\alpha+\beta)|\Uparrow\rangle_{hh} - (\alpha-\beta)|\Downarrow\rangle_{hh}]. \quad (2)$$

For the first row, because the in-plane g factor of a HH is 0, the easy axis of the HH is not determined by the in-plane magnetic field but by the confinement directions of the QW, $|\Uparrow\rangle_{hh}$ and $|\Downarrow\rangle_{hh}$. This is why $|\uparrow\rangle_e = |\leftarrow\rangle_e + |\rightarrow\rangle_e$ and $|\downarrow\rangle_e = |\leftarrow\rangle_e - |\rightarrow\rangle_e$ are chosen. For any values of α and β, the probability amplitude is 1/2 in all four terms of electron-hole pairs in Eq. (2). This holds for the HH excitation in the $N_e = 0$ SQD. In a similar manner, for the LH excitation, the optical transition expressed in the first square-bracket term of Eq. (2) is prohibited for the excitation of the electron-HH pair in the $N_e = 1$ SQD. Therefore, the photoelectron trapping probability for $N_e = 1$ is reduced to half the value compared to $N_e = 0$.

Figures 1(c) and 1(d) are the calculated spectra of the HH and LH excitations by the photoelectron trapping probability for V- and H-polarized light, respectively, based on the preceding discussion. The calculation procedure is explained in detail in Sec. 6 of the Supplemental Material (SM) [13]. N_e in the figures is the electron number initially prepared in the SQD, i.e., for the V(H)-polarized light. The black and red (blue) curves indicate the spectra for $N_e = 0$ and 1 SQDs, respectively. The suppressions of the trapping probability due to the optical spin blockade in the $N_e = 1$ SQD are indicated by the hollow arrows.

The QD device studied here is fabricated in a two-dimensional electron gas (2DEG) accumulated in an AlGaAs/GaAs/AlGaAs QW. Two kinds of GaAs QW wafers are used to fabricate the QD devices (SM Secs. 1 and 2). The first wafer has a well width grading between 12 and 15 nm across a quarter of a 2 inch wafer, which is used to fabricate the SQD for the optical spin blockade experiment. Therefore the actual well width of the SQD depends on the wafer position used for the fabrication, and we roughly estimate the width of 13 ± 0.5 nm. The DQD for the Pauli spin blockade experiment is fabricated using the second wafer with a fixed well width of 15 nm. The QDs are defined by applying appropriate voltages to the surface gates. A quantum point contact (QPC) is formed on the right of the dot by gates TR and QRL and used as a charge sensor [all the gate labels are depicted in Fig. 2(a)]. The QPC sensor is embedded in a radio-frequency (rf) impedance-matched circuit with resonance frequency of 214.5 MHz [14], allowing fast readout of the photogenerated

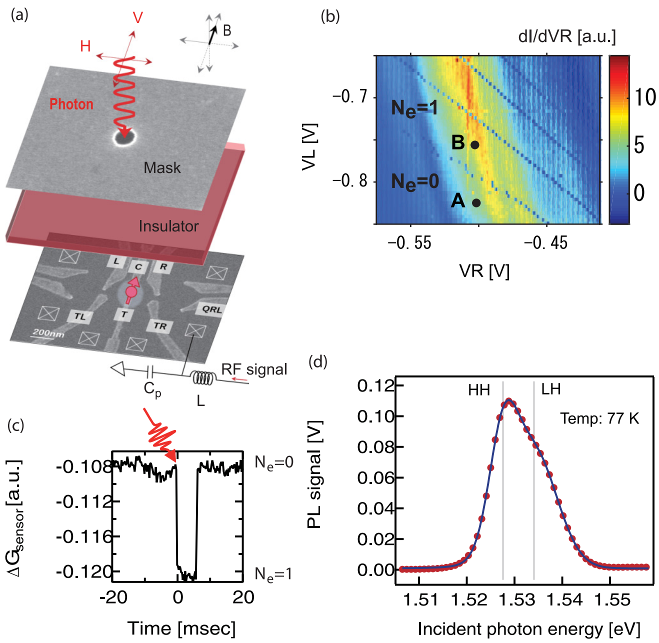

FIG. 2. (a) Schematic showing the device layout and directions of light polarization and external magnetic field. Photons are irradiated onto the dot, passing through a 500 nm diameter aperture in a thick gold mask placed on top of the dot. All the cross marks indicate Ohmic contacts between the QW and the wafer surface. (b) Charge stability diagram of the dot measured with the rf charge sensor. Point A and B corresponds to the $N_e = 0$ and $N_e = 1$ charge states, respectively. The diagonal lines indicate the charge state transitions. (c) Typical time trace of photoresponse at the single QD. The time trace is measured in the $N_e = 0$ Coulomb blockade region and measured with a sampling rate of 4 kHz. A pulsed photon is irradiated at $t = 0$ ms. ΔG_{sensor} drops just after the photon irradiation and returns to the original level after some time, indicating a photoelectron is generated and trapped on the QD and then tunnels out. (d) Photoluminescence spectrum of the wafer measured at 77 K without magnetic field. The left higher peak is assigned to HH excitation and the small shoulder located on the higher energy side of the HH peak is assigned to LH excitation.

electrons. The tunnel coupling of the dot to the right lead was carefully adjusted with gates T and TR to be in the range of 0.2 to 20 kHz, which is comparable to or lower than the charge sensor band width, and negligible to the left lead. A 100 to 200 nm thick dielectric layer of calixarene is deposited on top of the central region of the device. A 300 nm thick Ti/Au metal mask with a 500 nm diameter aperture is centered over the device.

First, the SQD device is tuned to accumulate just a few electrons in the dot. Fig. 2(b) shows the charge state stability diagram measured with the charge sensor with respect to gates L and R. The diagonal lines indicate the charge state transitions. The charge number N_e is fixed in each region of Coulomb blockade between the neighboring lines. The charge transition line of $N_e = 0$ to 1 appears jagged, owing to the dot-lead tunnel rate being lower than the gate voltage sweep rate. We set the gate bias point A (B) in the $N_e = 0$ (1) state in Fig. 2(b) for the photon-trapping experiment.

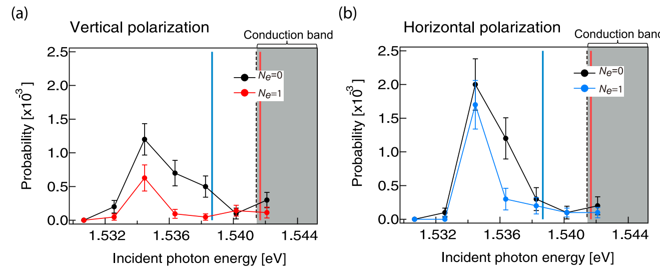

FIG. 3. Photoelectron trapping probability per photon passing through the aperture measured with V-polarized light in (a) and H-polarized light in (b) as a function of excitation photon energy. Black dots indicate results for $N_e = 0$ and red (blue) dots for $N_e = 1$ with V(H)-polarized light. The in-plane magnetic field is 7 T. A peak at around 1.5345 eV indicates the HH state excitation. The vertical blue and red lines are LH peak energies expected from previously reported values. The gray region indicate that the excitation to the conduction band appears above the vertical broken line and is overlapped with the LH+ excitation peak.

Figure 2(c) shows the typical photoresponse, conductance shift of the charge sensor ΔG_{sensor}. In Fig. 2(c) we observe an abrupt change of ΔG_{sensor} at $t = 0$, indicating a photoelectron trapping event. The large photo-response in (c) is assigned to a single photoelectron trapping event in the dot. The photogenerated electron eventually tunnels out of the dot and ΔG_{sensor} returns to the original value. The time resolution of ΔG_{sensor} is 250 μs: shorter than the electron spin lifetime, and therefore we are able to detect the orientation of the photogenerated electron spin before the spin relaxes.

In order to evaluate the HH and LH excitation energy in the dot fabricated from the QW wafer, we measured the photoluminescence (PL) spectrum at 77 K in the absence of a magnetic field [see Fig. 2(d)]. The temperature is relatively high so that both HH and LH excitons are populated. The PL spectrum is asymmetric due to the contribution from the LH excitons on the high energy side. The main peak is assigned to the HH exciton at 1.5276 eV, and the shoulder at higher energies is assigned to the LH exciton at 1.5341 eV. The exciton resonance energies are consistent with calculations for a one-dimensional finite well potential. The GaAs band gap increases with decreasing temperature, and therefore the HH and LH resonance peaks shift to higher energy by about 11 meV when the temperature is lowered to 0.1 K.

The first QW wafer used here is specially designed such that electron Zeeman energy is larger than the excitation light band width ($\Delta\nu_{\text{photon}} = 0.6$ meV) and the Zeeman splitting LH− or LH+ state is well resolved. The Zeeman energy is estimated to be 162 μeV ($<\Delta\nu_{\text{photon}}$) for electrons, assuming a g factor of -0.4 [15], and 3 meV ($>\Delta\nu_{\text{photon}}$) for LHs, assuming a g factor of -3.5 [16,17] under a large in-plane magnetic field, $B_\parallel = 7$ T. The magnetic field is chosen to be large enough to polarize the electron spin but not so large as to reduce the spin lifetime below the readout time [11]. We find that the SQD device used here has large enough HH-LH separation to be able to resolve the LH− state excitation for the photon-trapping experiment.

The obtained photon-trapping probability spectra for the V and H polarization at $B_\parallel = 7$ T are shown in Figs. 3(a) and

3(b), respectively. The estimated electron temperature is about 100 mK (or 10 μeV). This is much smaller than the electron Zeeman energy, ensuring that $N_e = 1$ QD is spin polarized up to 88%. The black closed circles and colored closed circles in both figures represent the cases for $N_e = 0$ and 1, respectively. A peak at around 1.5345 eV observed for both $N_e = 0$ and 1 for both light polarizations is assigned to the HH excitation. This peak energy is consistent with that predicted from the PL data in Fig. 2(d). No Zeeman splitting is observed for the HH exciton peak, while for $N_e = 0$ in both figures the photon trapping probability for the LH excitation shows a dip at around 1.540 eV due to the Zeeman splitting (SM Sec. 2). The blue (red) vertical line indicates the calculated LH− (LH+) exciton energy, respectively, using a value from literature of the LH exciton Zeeman energy [16,17]. We observe a feature (shoulder or kink) in the blue line, reflecting the LH− excitation and an increasing probability of the red line.

Now we compare the photon-trapping probability for $N_e = 1$ to that of $N_e = 0$ to reveal the optical spin blockade effect. For V polarization in Fig. 3(a) the peak value assigned to the HH is reduced by nearly half and the peak corresponding to the LH− has been strongly suppressed when N_e is changed from 0 to 1. The reduction of the LH− is caluculated to be about $91 \pm 9.8\%$, which is likely limited by the electron spin polarization. On the other hand, the LH+ excitation sees a reduction at $N_e = 1$ of only about $62 \pm 30\%$. We suspect that the reason for this inconsistency is that the excitation energy of the LH+ exciton allows for excitations between the HH band and the electron conduction band. So at the LH+ peak both excitations contribute to the trapping probability (SM Sec. 5). We estimate the band-to-band excitation to be 7 to 8 meV (HH exciton binding energy) above the HH exciton peak [18,19]. The edge of the excitation energy of the band-to-band transition (grey) is indicated by the broken line in both Fig. 3(a) and Fig. 3(b). Therefore, we focus on the optical spin blockade effect in the HH and LH− excitation.

For the H polarization in Fig. 3(b), the difference of the HH peak between $N_e = 0$ and 1 is not so clear compared to that for the V polarization, although it is qualitatively consistent with Fig. 3(a). The peak assigned to the HH for $N_e = 1$ is slightly reduced at the peak energy (by $85 \pm 24\%$) but strongly suppressed on the high energy side (>50%). The LH− peak is only slightly reduced, which is consistent our expectation shown Fig. 1(d).

The $N_e = 0$ photon-trapping probability spectrum, particularly for the HH excitation, is apparently different between the V- and H-polarized light excitation in Figs. 3(a) and 3(b) even though they should be the same as the expected results depicted in Figs. 1(c) and 1(d). The reason is not clear, but could possibly be due to asymmetry in the optical coupling to the dot through the metal mask aperture between the V- and H-polarized light as the dot shape is elliptic. Since the gate structure and the electrical field configuration near the QD are too complicated to be calculated, it is difficult to discuss the spectrum difference. Nevertheless, we successfully observe a qualitatively consistent influence of the optical spin blockade on the photon trapping probability in the LH− excitation.

In the preceding paragraphs we addressed the photon-to-spin conversion through the LH− excitation using the optical spin blockade effect in a SQD. Note that, according to the principle of the optical selection rules, a photoelectron created from the LH+ state has the spin opposite to one from the LH− state upon irradiating photons of the same polarization. We continue to perform photoelectron excitation from the LH+ state and detect the spin orientation using the Pauli spin blockade method in a DQD. The DQD sample is fabricated from the second wafer with a slightly larger well width, because the HH-LH separation is smaller and no large overlap of the LH+ and the conduction band excitations is assumed. Indeed, we confirm that this assumption holds from measurements of the PL spectrum [see Fig. SI1(c) of the Supplemental Material] and photon-trapping spectrum [see Figs. SI1(d) and SI1(e)].

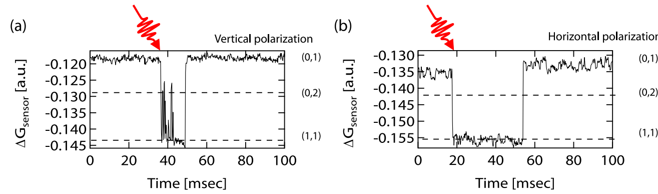

FIG. 4. Typical time traces of photoelectron trapping on the resonant DQD. (a) Photoelectron trapping signal with V-polarized photon excitation. The signal level of the charge sensor shows the oscillation between (0,2) and (1,1) charge states just after the photoelectron trapping, indicating that the photogenerated electron has a spin antiparallel to the magnetic field. (b) For the case of the H-polarized photon excitation no oscillation is observed, because the electron spin is parallel to the prepared electron spin and blocked in the left dot by the Pauli exclusion rule.

The measurement method in detail is explained in [11]. Here we briefly summarize. First, a DQD is prepared in the (0,1) state (the electron numbers in the left and right dots) with the right electron spin parallel to the external magnetic field. Specifically, when an electron with spin antiparallel to the magnetic field is generated in the left dot by the V-polarized photon, the photoelectron can tunnel to the right dot. Especially, when two singlet states of $S(1, 1)$ and $S(0, 2)$ are energetically aligned, the interdot electron tunneling of the $S(1, 1)$-$S(0, 2)$ transition repeatedly occurs as in Fig. 4(a). This is not the case for the H-polarized photon excitation, because a triplet state of $T_+(1, 1)$ with two up spins is created and the interdot electron tunneling is blocked by Pauli exclusion rule. Consequently, the difference of the photogenerated spin configuration, parallel or antiparallel, can be distinguished in a single-shot measurement of the charge change in the DQD (SM Sec. 4) [11]. Note that a photoelectron can be created in either dot of the DQD, and therefore we only used the post-selected events of photoelectron trapping in the left dot to derive the probability of finding the parallel or antiparallel spin configuration.

Figure 4 shows a typical charge sensor photoresponse obtained for the V- and H-polarized photon excitation in (a) and (b), respectively, at in-plane magnetic field $B_\parallel = 7$ T. We observe interdot oscillations between (1,1) and (0,2) upon the photoelectron trapping in (a) antiparallel but not in (b) parallel configuration. The probability of finding interdot oscillations (or antiparallel spin configuration) is $53 \pm 13\%$ for the V-photon excitation but $0 \pm 0\%$ for the H-photon excitation. The probability for the H-polarization is as expected; however, for the V-polarization it is smaller than 100%. This is likely due to unintentional misalignment of the (1,1) and (0,2) states induced by the photon irradiation. Nevertheless, the obtained result is consistent with the prediction about the coherent photon-to-spin conversion in the spin-selective LH+ excitation.

In conclusion, the quantum state transfer for a single photon to a single electron spin through spin-resolved LH-state excitation is confirmed by a combined method of single-shot charge sensing and the optical spin blockade in the SQD. We observed that the photoelectron trapping probability is strongly reduced for the V-polarized photon excitation of the $N_e = 1$ SQD from the LH− state due to the optical spin blockade effect. Additionally, we confirmed that the quantum state transfer is correctly realized with a Pauli spin blockade effect in the DQD. These results consistently show that the gated GaAs QD provides a candidate for a quantum interface between a photon and a spin, encouraging further investigation to demonstrate the quantum state transfer from a photon qubit to a spin qubit. Since we previously established an experimental technique of single photon-electron pair creation from polarization-entangled photon pairs [20], our result indicates a possibility of generation of quantum entanglement between a photon and an electron spin in a QD. This will in turn open a pathway towards advanced quantum technology of quantum media conversion and quantum communication based on quantum teleportation.

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