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Phys. Rev. B **99**, 085203 — Published 7 February 2019
DOI: 10.1103/PhysRevB.99.085203

Photo-generation of a single electron from a single Zeeman-resolved light-hole exciton with preserving angular momentum

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Quantum state transfer from a single photon to a single electron following selection rule can only occur for a spin-resolved light hole excitation in GaAs quantum dots; However, these phenomena are 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 photo-excitation 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.

The selection rules of inter-band optical transitions in a GaAs/AlGaAs hetero-structures have long been studied, as they form the basis for the quantum interface between an absorbed photon polarization and a photo-electron 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 sub-levels split 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, photo-generation of a single electron in a laterally gated quantum dot (QD) has been achieved but 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 photo-generated 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 photo-electron; effectively working as a spin projection measurement along the field. The successive photo-generation of an electron with a parallel spin 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. As a preceding study, the blockade effect has indeed been observed for photo-electrons 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 photo-electron, because the HH can not provide a superposition state of two opposite spin orientations.

Here we perform spin-selective photo-excitation of single HH and LH excitons in a laterally gated GaAs QD sample and use a charge sensing technique to measure the probability of finding single photo-electrons in the dot that are created in line with the optical selection rules. We, at 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 photo-excitation of a pair of an electron and one of the spin-resolved LHs is prohibited by optical spin blockade for the case of the photo-generated 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 photo-electron with a double QD (DQD) [11]. We detect the spin orientation (up or down) of the photo-generated 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 that the angular momentum is preserved via photo-excitation

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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 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:

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

For the LH excitation in the $N_e = 0$ SQD, which is represented by Eq. (1), the H (V)-polarization 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-polarization 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-polarization light is forbidden for $N_e = 1$.

We first study optical spin blockade in single photo-electron generation by selective photo-excitation 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 [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)):

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

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 the confinement direction 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 the similar discussion with the LH excitation, the optical transition expressed in the first square bracket of Eq. (2) is prohibited for the excitation of the electron-HH pair in the $N_e = 1$ SQD. Therefore, the photo-electron trapping probability for $N_e = 1$ is reduced to half the value compared to $N_e = 0$.

Fig. 1(c), and (d) are the calculated spectra of the HH and LH excitation by the photo-electron trapping probability for the V-, and H-polarized light, respectively, based on the preceding discussion. The calculation procedure is explained in detail in the supplementary information 6. N_e in the figures is the electron number initially prepared in the SQD, i.e. for the V(H)-polarization light. The black, and red (blue) curves indicate the spectrum for the $N_e = 0$, and 1 SQD, 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 (Supplementary information 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 [13], allowing fast read-out of the photo-generated 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 neighbouring 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. 2(c) shows the typical photo-response, conductance shift of the charge sensor $\Delta G_{\mathrm{sensor}}$. In the Fig. 2 (c) we observe an abrupt change of $\Delta G_{\mathrm{sensor}}$ at t = 0, indicating a photo-electron trapping event. The large photo-response in (c) is assigned to a single photo-electron trapping event in the dot. The photo-generated electron eventually tunnels out of the dot and ΔG_{sensor} returns to the original value. The time resolution of ΔG_{sensor} is 250 μsec: shorter than the electron spin lifetime, and therefore, we are able to detect the orientation of the photo-generated 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 photo-luminescence (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 ($\Delta\nu_{\rm photon}$=0.6 meV) and the Zeeman splitting LH- or LH+ state is well resolved. The Zeeman energy is estimated to be 162 μeV ($< \Delta\nu_{\rm photon}$) for electrons, assuming the g-factor of -0.4 [14], and 3 meV ($> \Delta\nu_{\rm photon}$) for LHs, assuming the g-factor of -3.5 [15, 16] under a large in-plane magnetic field, $B_{//}$ = 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 $B_{//}$ = 7 T are shown in Fig. 3(a) and Fig. 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 case 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 (Supplementary information 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 [15, 16]. 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 ± 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 ± 30 %. We suspect 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 (Supplementary information 5). We estimate the band to band excitation to be 7 to 8 meV (HH exciton binding energy) above the HH exciton peak [17, 18]. The edge of the excitation energy of the band to band transition (grey) is indicated by the broken line in both figures of Fig. 3(a) and Fig. 3(b). Therefore, we are focusing 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 ± 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-polarization light excitation in Fig. 3(a) and (b) even though they should be the same as the expected results depicted in Fig. 1(c) and (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-polarization 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 qualitative 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 photo-electron created from the LH+ state has the spin opposite to one from the LH- state. We continue to perform photo-electron 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)) and photon-trapping spectrum (See Fig. SI1(d) and (e)).

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 number in the left and right dot) with the right electron spin parallel to the exteranl 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 photo-electron can tunnel to the right dot. Especially, when two singlet states of S(1,1) and S(0,2) are energetically aligned, the inter-dot electron tunneling of 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 inter-dot electron tunneling is blocked by Pauli exclusion. Consequently, the difference of the photo-generated spin configuration, parallel or antiparallel, can be distinguished in a single-shot measurement of the charge change in the DQD (Supplementary information 4)[11]. Note that a photo-electron can be created in either dot of the DQD, and therefore we only used the post-selected events of photo-electron trapping in the left dot to derive the probability of finding the parallel or antiparallel spin configuration.

Fig. 4 shows a typical charge sensor photo-response obtained for the V-, and H-polarized photon excitation in (a), and (b), respectively at in-plane magnetic field $B_{//}$ = 7 T. We observe inter-dot oscillations between (1,1) and (0,2) upon the photo-electron trapping in (a), antiparallel but not in (b), parallel configuration. The probability of finding inter-dot oscillations (or antiparallel spin configuration) is 53±13 % for the V-photon excitation but 0±0 % for the H-photon excitation. The probability for the H-polarization is as expected, however, for the V-polarization 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 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 light-hole 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 photo-electron 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 provide 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 and generation of quantum entanglement between a photon and an electron spin in a QD. This will in turn open a path way towards advanced quantum technology of quantum media conversion and quantum communication based on quantum teleportation.

ACKNOWLEDGMENTS

This work was partially supported by Grant-in-Aid for Young Scientific Research (A) (No. JP15H05407), Grant-in-Aid for Scientific Research (A) (No. JP16H02204, No. JP25246005), Grant-in-Aid for Scientific Research (S) (No. JP26220710, No. JP17H06120), Grant-in-Aid for Scientific Research (B) (No. JP18H01813) from Japan Society for the Promotion of Science, JSPS Research Fellowship for Young Scientists (No. JP16J03037), JSPS Program for Leading Graduate Schools (ALPS) from JSPS, Japan Society for the Promotion of Science (JSPS) Postdoctoral Fellowship for Research Abroad Grant-in-Aid for Scientific Research on Innovative Area, "Nano Spin Conversion Science" (No.26103004), Grant-in-Aid for Scientific Research on Innovative Area, CREST (No. JPMJCR15N2). A.D.W., A.L. and S.R.V acknowledge gratefully support of DFG-TRR160, BMBF-Q.com-H 16KIS0109, and the DHF/UFA CDFA-05-06. The main text is checked and edited by Ivan Valerievich Borzenets.

References

[1] R. Vrijen and E. Yablonovitch, Physica E: Low-Dimensional Systems and Nanostructures **10**, 569 (2001).
[2] H. Kosaka, H. Shigyou, Y. Mitsumori, Y. Rikitake, H. Imamura, T. Kutsuwa, K. Arai, and K. Edamatsu, Physical Review Letters **100**, 096602 (2008).
[3] H. Kosaka, T. Inagaki, Y. Rikitake, H. Imamura, Y. Mitsumori, and K. Edamatsu, Nature **457**, 702 (2009).
[4] T. Inagaki, H. Kosaka, Y. Mitsumori, and K. Edamatsu, Physical Review B **89**, 085311 (2014).
[5] H. Kosaka, D.S. Rao, H.D. Robinson, P. Bandaru, K. Makita, and E. Yablonovitch, Physical Review B **67**, 045104 (2003).
[6] D. S. Rao, T. Szkopek, H. D. Robinson, E. Yablonovitch, and H. W. Jiang, Journal of Applied Physics **98** (2005).
[7] M. Kuwahara, T. Kutsuwa, K. Ono, and H. Kosaka, Applied Physics Letters **96**, 10 (2010).
[8] A. Pioda, E. Totoki, H. Kiyama, T. Fujita, G. Allison, T. Asayama, A. Oiwa, and S. Tarucha, Physical Review Letters **106**, 146804 (2011).
[9] T. Fujita, H. Kiyama, K. Morimoto, S. Teraoka, G. Allison, A. Ludwig, A. D. Wieck, A. Oiwa, and S. Tarucha, Physical Review Letters **110**, 266803 (2013).
[10] A. Högele, M. Kroner, S. Seidl, K. Karrai, M. Atatüre, J. Dreiser, A. Imamoğlu, R. J. Warburton, A. Badolato, B. D. Gerardot, and P. M. Petroff, Applied Physics Letters **86**, 221905 (2005).
[11] T. Fujita, K. Morimoto, H. Kiyama, G. Allison, M. Larsson, A. Ludwig, S. R. Valentin, A. D. Wieck, A. Oiwa, and S. Tarucha, , arXiv:1504.03696 (2015).
[12] X. Marie, T. Amand, P. Le Jeune,M. Paillard,

P. Renucci,L. E. Golub, V. D. Dymnikov, and E. L. Ivchenko, Physical Review B **60**, 5811 (1999).
[13] C. Barthel, M. Kjærgaard, J. Medford, M. Stopa, C. M. Marcus, M. P. Hanson, and A. C. Gossard, Physical Review B - Condensed Matter and Materials Physics **81**, 161308 (2010).
[14] P. Le Jeune, D. Robart, X. Marie, T. Amand, M. Brousseau, J. Barrau, V. Kalevich, and D. Rodichev, Semiconductor Science and Technology **12**, 380 (1997).
[15] V. B. Timofeev, M. Bayer, A. Forchel, and M. Potemski, , 57 (1996).
[16] M. V. Durnev, Physics of the Solid State **56**, 1416 (2014).
[17] G. Bastard, E. E. Mendez, L. L. Chang, and L. Esaki, Physical Review B **26**, 1974 (1982).
[18] L. Hrivnák, Journal of Applied Physics **72**, 3218 (1992).

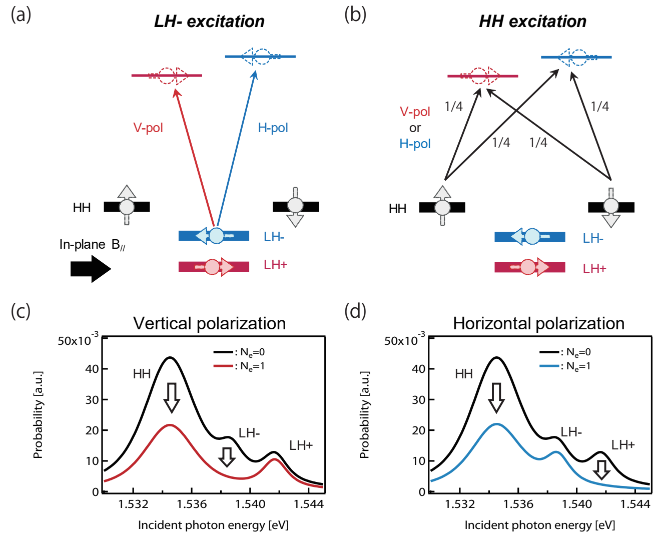

FIG. 1. (Color online). (a), (b)Schematics of spin-selective optical transitions between the electron and hole states for both the LH and HH excitation. For the $N_e = 0$ dot, optical excitation of both an electron spin parallel and antiparallel to an in-plane magnetic field are allowed. On the other 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 because of the Pauli exclusion rule in the dot. (c), (d) Calculations of the photo-electron trapping probability in the SQD as a function of the incident photon energy. The peak shapes are estimated from the photo-electron trapping spectrum of the second wafer depicted in Fig. SI1 (d). 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

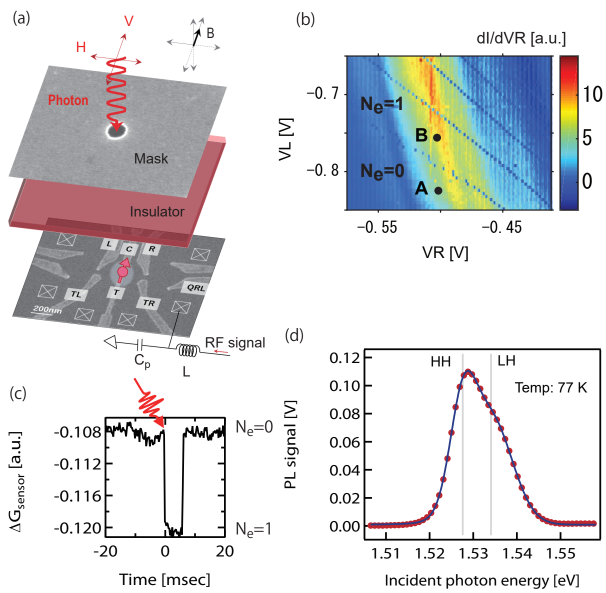

FIG. 2. (Color online). (a) Schematic showing the device layout and directions of light polarization and external magnetic field. Photons are irradiated onto the dot passing through a 500 nm diameter aperture in a thick gold mask placed on top of the dot. All the cross marks indicate Ohmic contacts between 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 state, respectively. The diagonal lines indicate the charge state transitions. (c) Typical time trace of photo-response at the single QD. The time trace is measured in the $N_e = 0$ Coulomb blockade region and measured with the sampling rate of 4 kHz. A pulsed photon is irradiated at t = 0 msec. ΔG_{sensor} drops just after the photon irradiation and returns to the original level after some time, indicating a photo-electron is generated and trapped on the QD and then tunnels out. (d) Photo-luminescence 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 to LH excitation.

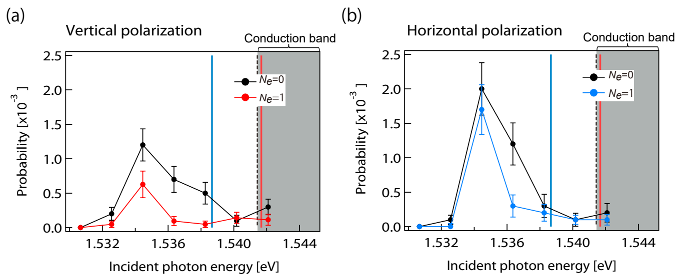

FIG. 3. (Color online). Photo-electron trapping probability per photon passing through the aperture measured with the V-polarized light in (a) and H-polarized light in (b) as a function of excitation photon energy. Black dots indicate for $N_e = 0$ and red (blue) dots for $N_e = 1$ with the V(H)-polarized 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 LHs 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.

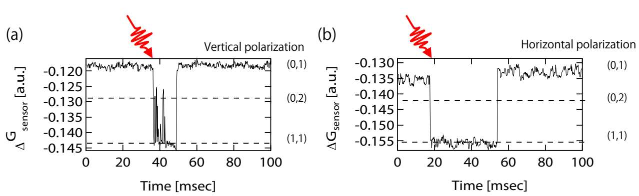

FIG. 4. (Color online). Typical time traces of a photo-electron trapping on the resonant DQD. (a) Photo-electron trapping signal with V-polarization photon excitation. The signal level of the charge sensor shows the oscillation between (0,2) and (1,1) charge states just after the photo-electron trapping, indicating that the photo-generated 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.