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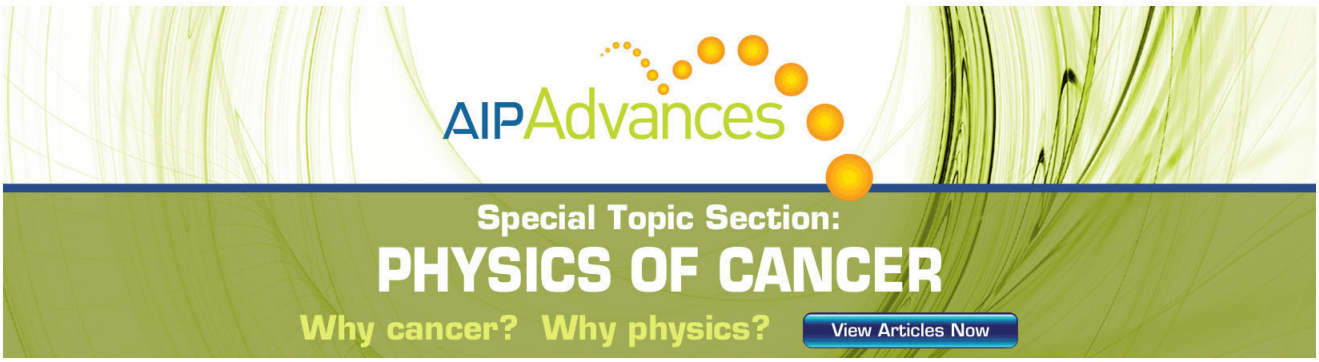

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Nuclear and excitonic spin polarization formed using cross-linearly polarized pulse pair via half-localized state in a single self-assembled quantum dot

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The oscillations of excitonic and nuclear spin polarizations in an optically pumped single self-assembled $In_{0.8}Al_{0.2}As/Al_{0.35}Ga_{0.65}As$ quantum dot (QD) were clearly observed under the excitation of a wetting layer edge at $B=5$ T. This indicates that an exciton pair with opposite spins is alternatively created via the half-localized state only by changing the delay time between cross-linearly polarized pulse pair. Furthermore, periodic modulation of Zeeman energy synchronizes the degree of circular polarization of photoluminescence from a single QD, indicating that the Overhauser field follows the optically created electron spin polarization in half-localized states of a QD, and the half-localized state in a QD consists of a confined electron in a discrete state and hole in the continuum state. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2927448]

I. INTRODUCTION

Manipulations of a nuclei-electron spin system in a semiconductor quantum dot (QD) are currently of great interest because they have many potential applications, such as memory with ultralong coherence,[1] the use of a local magnet in quantum computation,[2] and media conversion between photon qubits and electron spin qubits[3] for quantum information processing. It is difficult to manipulate nuclear spins in a QD by the standard nuclear spin resonance technique due to the small amount of nuclei ($\sim 10^5$) that are extremely well isolated. Therefore, electron-nuclear spin interaction (hyperfine interaction) has recently been used as a driving force for nuclear spin polarization because it is emphasized by the strong confinement of the electron wave function in QDs. In fact, a high rate of nuclear spin polarization was recently observed in interface GaAs QDs,[4–6] self-assembled InAlAs QDs,[7,8] and InGaAs QDs.[9–11] In the case of optical pumping, a highly polarized electron spin should be generated in the ground state to obtain a high nuclear spin polarization.[12] In general, the electron spin generated in a continuum state such as bulk material is instantaneously depolarized. Even in a QD, the observed nuclear spin polarization is small when electron-hole pairs are optically created with an excitation light having the barrier band gap energy.[7] On the other hand, when electron (hole) spin is generated in "half-localized" states[13–17] that consist of electron and hole states, one of which is confined to the ground state of the QD, the observed Overhauser field is of a few tesla,[8] because one of the carriers (possibly an electron) confined in a QD preserves the spin state with high possibility.[18] The half-localized states are significantly important to understand the practical QD physics. These states appear as a wing of the wetting layer energy band in the photoluminescence excitation (PLE) spectra. The half-localized states are one of the characteristics of the real QDs which are not observed in the ideal "artificial atoms."

In this study, we investigate the effect of the half-localized state on the excitation process in a single InAlAs QD under a magnetic field, wherein copropagating coherent visible light pulses are used. The in-phase and antiphase coherent polarization oscillations of exciton spins in a single QD are clearly observed by changing the interpulse delay time. In addition, the optically created Overhauser field in a QD is observed using orthogonally polarized optical pulses. Although it is not mainstream for spin manipulation in a QD, the half-localized state can act as an intermediate state for the Λ transition[19–21] between remote localized states, such as the stimulated Raman adiabatic passage. Moreover, the half-localized state can relax the selection rules of optical transitions between localized states in a single QD. Therefore, if the laboratory pulses have sufficient power to excite half-localized states, it will be a great advantage.

II. EXPERIMENTAL DETAILS

$In_{0.8}Al_{0.2}As$ QDs were grown on an $Al_{0.35}Ga_{0.65}As$(120 nm)/GaAs(300 nm) buffer layer on an undoped (100) GaAs substrate by molecular beam epitaxy using a RIBER-MBE32 system. The growth temperature was 620 °C for the buffer layer and 500 °C for the InAlAs QDs. The substrate temperature was lowered gradually during the growth of the buffer layers. The InAlAs QDs were grown at a low growth rate of 1.5×10^{-13} monolayers/s. For stabilizing these QDs, postannealing was performed for 90 s. Then, the substrate temperature was gradually increased to 590 °C when an AlGaAs capping layer (100 nm) was grown. For

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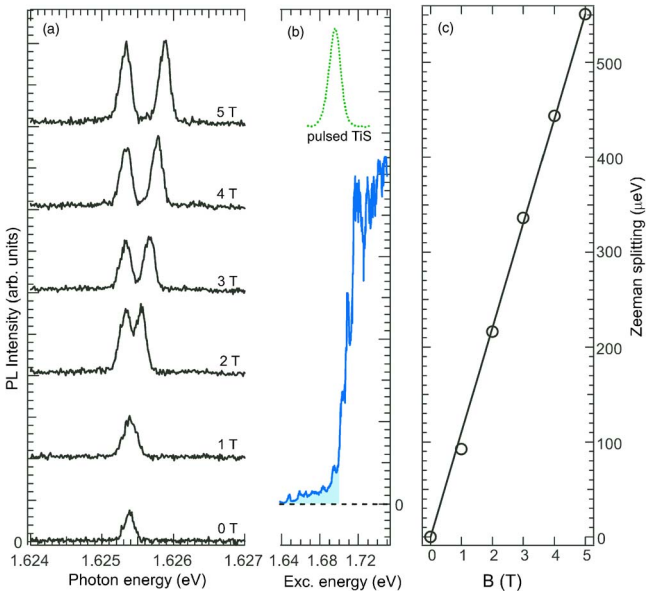

FIG. 1. (Color online) (a) PL spectra of a single QD (solid black line) recorded at different magnetic fields under cw-TIS laser excitation (720 nm). (b) Solid (blue) line indicates the PLE spectrum with a detection energy of 1.6253 eV was obtained under zero magnetic field at 4.2 K. Dotted (green) line located on top of PLE spectrum is a mode-locked TIS laser pulse spectrum. (c) Zeeman splitting of an exciton line in a InAlAs QD shown in (a). Open circles represent experimental data, and the line is fitted with $g_x\mu_B B_z$, $g_x=1.90\pm0.02$.

single QD spectroscopy, small mesa structures with the top lateral size of ~150 nm were fabricated by electron-beam lithography and wet chemical etching to limit the number of observable QDs. Since the QD density is estimated to be $\sim 2.5\times10^{10}$ cm^{-2} from the atomic force microscopy measurement of the uncapped sample under the same growth condition, some mesas have one or a few QDs due to inhomogeneous QD size distribution, resulting in well-separated sharp, single QD emissions.

Figure 1(a) shows the photoluminescence (PL) spectra of a single InAlAs QD obtained by varying the magnetic field up to 5 T at 4.2 K. The magnetic field was aligned parallel to the growth direction, and the sample was excited in a Faraday configuration. A continuous-wave Ti:sapphire (TIS) laser was used as an excitation source with a linearly polarization. The PL peak in the figure has an emission energy of 1.6253 eV (full width at half-maximum of 124 μeV) at $B=0$ T. In Fig. 1(c), Zeeman splitting is plotted against the external magnetic field. By fitting the spectra with $g_x\mu_B B_z$, we obtained an exciton g factor as $g_x=1.90\pm0.02$. A continuous-wave (cw) TIS laser with an excitation energy of 1.654–1.758 eV was used for the PLE measurements [Fig. 1(b)]. The detection energy of the PLE spectrum was adjusted to the emission energy of 1.6253 eV. The sharp features above 1.7 eV are not due to bound states and may indicate some type of resonance of a multiphonon or a mere detector noise. Note that a PL peak is located on the low-energy tails of the half-localized states, as shown in the PLE spectra. Previous studies[13–17] suggested that these continuumlike tails were due to transitions between one of the carriers in a QD and the other carrier in the wetting layer, which is a quantum well with about 1 ML thickness beneath the QD.

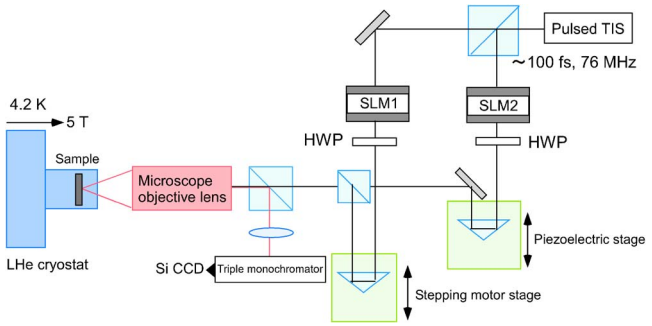

FIG. 2. (Color online) Schematic illustration of an experimental setup based on a Mach-Zehender interferometer. The setup includes two dispersion-free 4-f optical systems. The polarization and delay time between the two pump pulses are controlled by HWP angle and the piezoelectric stage, respectively.

A schematic of the experimental setup is shown in Fig. 2. A mode-locked TIS laser with a pulse repetition rate of 76 MHz was used as the light source. A sequence of pump pulses was produced by passing them through a Mach-Zehender interferometer. A 100 fs laser pulse was divided into two pump pulses with a controlled time delay using a piezoelectric stage with a resolution of 5 nm. The pump-pulse pair was spectrally shaped by removing the tail of the laser spectra using two dispersion-free 4-f optical systems in conjunction with spatial light modulators (SLMs), which have a resolution of 350 μeV. Then, two pump pulses were combined coaxially and focused onto the sample surface by a microscope objective lens. The single QD emissions collected by the same objective lens were dispersed by a triple grating spectrometer ($f=0.64$ m) and detected with a liquid N_2-cooled Si-charge-coupled device (CCD) camera, which has a resolution of 12 μeV. Next, the energies of the emission peaks were determined to be on the order of 3 μeV by spectral fitting. The mode-locked TIS laser was tuned to ~1.7 eV for the excitation of the half-localized states determined by the PLE measurements [Fig. 1(b)]. The sample was held in a liquid He-flow cryostat and maintained at 4.2 K. A magnetic field of 5 T was applied along the growth direction. The pump-pulse pair can have either cross-linear or colinear polarization by adjusting two half-wave plates (HWPs) placed after the SLMs.

III. RESULTS AND DISCUSSION

Figure 3 shows the 3D plots of the excitonic Zeeman spectra as a function of the interpulse delay time. The exciton ground state with opposite spins splits by ~550 μeV at 5 T in both cases of (a) and (b). The emission energies of a Zeeman-split exciton pair for the case of (b) are slightly large ~200 μeV compared with those for the case (a). This is mainly due to the temperature drift of the sample, CCD detector, and experimental room during the long measurement time. The energy shift due to the temperature drift becomes a crucial problem when the discrete level is resonantly excited. However, this energy shift has no effect on our study, because we use a coherent pump pulse with a bandwidth of ~9 meV, which negates the above energy shift to excite the half-localized state. With the colinearly polarized excitation, the coherent amplitude oscillation of the PLs from the exci-

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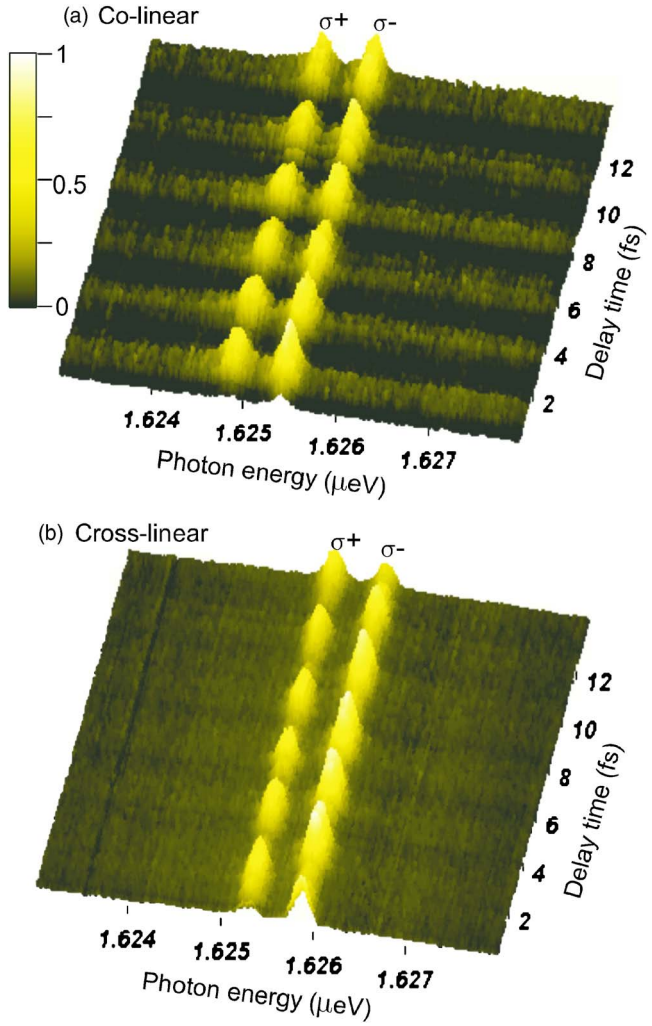

FIG. 3. (Color online) 3D image (CCD image) of a Zeeman pair as a function of the pulse delay time. Two excitation pulses can have either colinear (a) or cross-linear (b) polarization by changing the HWP angle (Fig. 2). In both cases (a) and (b), PL intensities are normalized by maximum intensity.

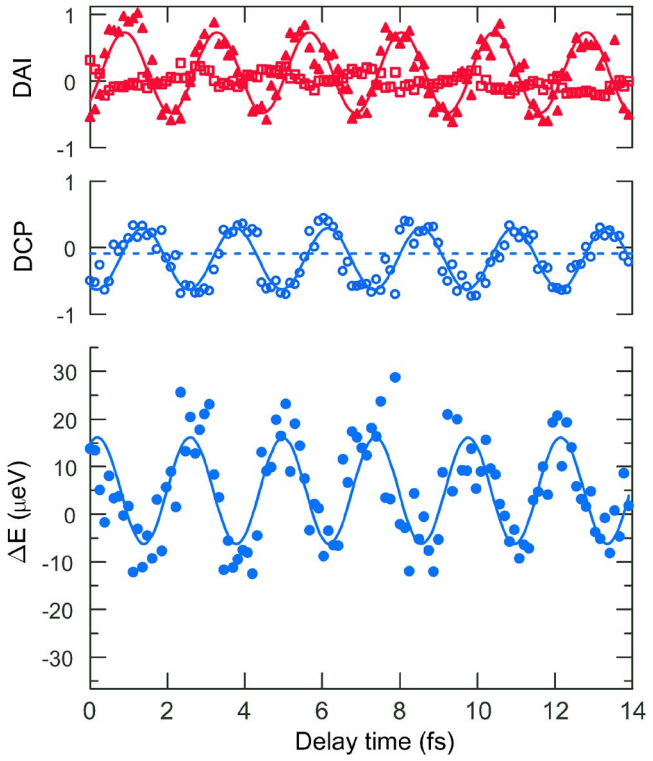

FIG. 4. (Color online) Amplitude interference effects of a Zeeman pair in colinearly (solid triangles) and cross-linearly (open boxes) polarized excitations. Solid and open circles indicate $\Delta E = E_{Ze}^{cross} - \langle E_{Ze}^{co} \rangle$ and DCP$=(I_+ - I_-)/(I_+ + I_-)$ as a function of the pulse delay in the cross-linear polarization setup. The DAI profile with colinear polarization and DCP (ΔE) with cross-linear polarization (open (solid) circles) are fitted with a cosine (red line) and sine (blue lines) function. Dashed (blue) line indicates the average values of exciton polarization rate in the colinearly polarized excitation.

ton ground states in a single QD is observed. As shown in Fig. 3(a), the time-integrated PL spectra of the Zeeman-split exciton pair oscillate *in phase* as a function of the interpulse delay time (τ) with a period ($\sim$2.4 fs) corresponding to the energy of the pump pulses ($\sim$1.7 eV). This oscillation of the Zeeman pair can be explained by the amplitude interference between first ($\mathbf{E}_1$) and second ($\mathbf{E}_2$) pulses, $\sim\cos(\omega\tau)\mathbf{e}_1\cdot\mathbf{e}_2$, where ω and $\mathbf{e}_{1,2}$ are the center frequency and polarization vector of the pump pulses, respectively. In this experiment, since the half-localized state is excited by the optical pulses, the observed PL oscillation decays within the overlapping time of the pump-pulse pair, unlike the use of the discrete excited state in the previous studies such as wavepacket interferometry.[22–24]

In the case of a pump pair with cross-linear polarizations, the amplitude interference becomes zero because the polarization of the pump pair is orthogonal, $\mathbf{e}_1\cdot\mathbf{e}_2=0$. In addition, the *antiphase* oscillations of the Zeeman split pair with the same frequency are clearly observed in the case of colinear polarization ($\mathbf{e}_1\cdot\mathbf{e}_2=1$),[25] as shown in Fig. 3(b). The polarizations formed by the orthogonally polarized pulse pair are modulated by the delay time between the first and second pulses. When τ is swept, the right (σ^+) and left (σ^-) circular polarization appear alternately with the period $\omega\tau=\pi$; $\mathrm{Re}\langle\mathbf{E}_1+\mathbf{E}_2|\sigma^{\pm}\rangle\sim\cos(\omega\tau\pm\pi/2)$. The τ-dependence of the degree of circular polarization is expressed as $\sim-\sin(\omega\tau)$. Therefore, the antiphase oscillations caused by the phase difference between the first and second pulses indicate that an exciton pair with opposite spins is created through the half-localized state in the QD.

The upper panel of Fig. 4 shows the degree of amplitude interference, which is determined from the total PL intensity of the Zeeman pair divided by the delay time average of PL intensity, DAI$=(I_+ + I_-)/\langle I_+ + I_-\rangle - 1$, where $I_{\pm}$ and $\langle I_+ + I_-\rangle$ indicate the integrated PL intensity of the corresponding peak of Zeeman pair ($\sigma^{\pm}$), and the delay time average of $I_+ + I_-$. In fact, amplitude interference is observed only in the case of the colinearly polarized excitation, indicating that the amounts of optically created exciton are modulated by the delay time between the pump pulse pair. The middle and lower panels of Fig. 4 highlight the degree of the circular polarization, DCP$=(I_+ - I_-)/(I_+ + I_-)$, with cross-linear polarizations and the energy difference between the Zeeman energies of cross-linearly and colinearly polarized excitation setup, $\Delta E = E_{Ze}^{cross} - \langle E_{Ze}^{co} \rangle$, where $\langle E_{Ze}^{co} \rangle$ is the delay time average of the Zeeman energy of colinearly polarized excitation. For the sake of comparison, the delay time average of DCP in a colinearly polarized setup is shown by the dashed line in

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the middle panel of Fig. 4. Note that the average value could not be estimated with a high degree of accuracy because the PL spectra of the Zeeman-split exciton pair becomes zero in phase, which implies that indeterminate forms are presented in DCP.

ΔE is varied up to ~30 μeV by changing the interpulse delay only in the case of cross-linearly polarized excitation. This variation from the colinearly polarized excitation can be explained by the "Overhauser field,"[12] $B_N = A\langle I_z\rangle / g_z^e \mu_B$, where A, $\langle I_z\rangle$, μ_B, and g_z^e are the hyperfine constant, averaged nuclear spin polarizations, Bohr magneton, and electron g-factor, respectively. The Overhauser field is induced by the hyperfine interaction between the electron and nuclear spins, $\mathcal{H}_{\mathrm{hf}} = A[I^+S^- + I^-S^+]/2 + AI_zS_z$, where $I^{\pm}(S^{\pm})$ and S_z are nuclear (electron) raising and lowering operators and electron spin polarization, respectively.[26] The nuclear spins are polarized by the first flip-flop term with an optically created electron spin. The electron Zeeman splitting is modulated by this optically created Overhauser field via AI_zS_z. The change of the Zeeman splitting energy from the linearly polarized excitation is known as the Overhauser shift (OHS). Therefore, the OHS is a good indicator of the electron spin polarization ($\langle S_z\rangle$) in a single QD. As shown in Fig. 4, the amplitude and direction of the Overhauser field synchronize the variation of DCP modulated by changing the delay time between the cross-linearly polarized pump pulses, $B_N \propto$ DCP.[9] Although the DCP of the emissions originating from the ground states is precisely lower than $2\langle S_z\rangle$ in the ground state due to the flip-flop process between electron and nuclear spins, the ΔE associated with the observed DCP is clearly observed, implying that observed DCP is reflected in $\langle S_z\rangle$. Therefore, the observed DCP indicates the polarization of the electron spin generated in a discrete state of a QD, because the DCP in the ground state is reflected in the exciton polarization produced in the half-localized state when the coherence of half-localized states is instantaneously destroyed by energy relaxation with the depolarization of hole generated in continuum state.

To verify the above-mentioned scenario, we compare the DCP dependence of ΔE with the calculated $\langle S_z\rangle$ dependence of OHS. The steady state OHS is expressed as

$$A\langle I_z\rangle = AQ(\langle S_z\rangle - S_0)\frac{T_{\mathrm{ND}}}{T_{\mathrm{NF}} + T_{\mathrm{ND}}}. \quad (1)$$

The flip-flop rate $1/T_{\mathrm{NF}}$ between the electron and nuclear spins is expressed as follows:[9–11]

$$\frac{1}{T_{\mathrm{NF}}} = \tau_c f\left(\frac{A}{N\hbar}\right)^2 \Big/ \left[1 + \left(\frac{\tau_c}{\hbar}\right)^2 \{g_z^e \mu_B (B_z - B_N)\}^2\right], \quad (2)$$

where $Q(=[I(I+1)]/[S(S+1)])$, τ_c, N, T_{ND}, S_0, and f indicate the momentum conversion coefficient from electron spin to nuclear spin system, correlation time of the coupled electron-nuclei system with broadening $\hbar/\tau_c$, number of nuclei in a QD, nuclear spin depolarization rate, thermal electron spin polarization, and the degree of electron existence in a QD, respectively. Note that B_N modulates only the electron Zeeman splitting energy, because the probability of a hole with a p-like wave function at the nucleus site is negligible. In Fig. 5, we show the OHS curves calculated using Eqs. (1) and (2). In addition, the following parameters are used: $A = 52.38$ μeV, $T_{\mathrm{ND}} = 5$ ms, $I(I+1) = 12.65$, $N = 23\,000$, $g_z^e = -0.3$, $\tau_c = 10.6$ ps, and $f = 0.1-1.0$ (solid curves), where A and $I(I+1)$ are weighted values for an $In_{0.8}Al_{0.2}As$ QD.[27] The asymmetrical OHS is caused by the increase in $1/T_{\mathrm{NF}}$ due to the decrease in $|g_z^e \mu_B (B_z - B_N)|$ when B_z is eliminated by B_N. The DCP dependence of ΔE is fitted by the $\langle S_z\rangle$ dependence of OHS using $f \sim 0.2$, as shown in Fig. 5, suggesting that the observed DCP is approximately $2\langle S_z\rangle$. To confirm this compatibility, we also show the reference data obtained in cw excitation measurements with the excitation energy ~1.7 eV (measurement method is shown in Ref. 7). The excitation power was set enough to saturate the PL intensity. In comparison to the reference data, the observed $|\Delta E|$ is small. This difference can be explained by the well-known excitation power dependence of nuclear spin polarization.[9,10] In the cw excitation measurement, the effective excitation rate of the electron is roughly limited by the exciton lifetime in the ground state, ~1.5 ns. On the other hand, a pulse occurs only once in ~13 ns because of a repetition rate of 76 MHz. Therefore, the effective excitation rate in cross-linear polarized excitation is about one-ninth that in cw excitation. In fact, the calculation curve with $f \sim 0.9$ is fitted to the experimental data in the cw excitation. Furthermore, this compatibility suggests that the observed DCP of the optically created exciton via the half-localized states in a QD is attributed not to hole spin polarization but $2\langle S_z\rangle$, because the OHS is not observed when electron spin polarization is created in the continuum state with instantaneous depolarization. Therefore, the optically created exciton in the half-localized state proved to consist of an electron with a long coherence in a discrete QD state and hole in the continuum state.

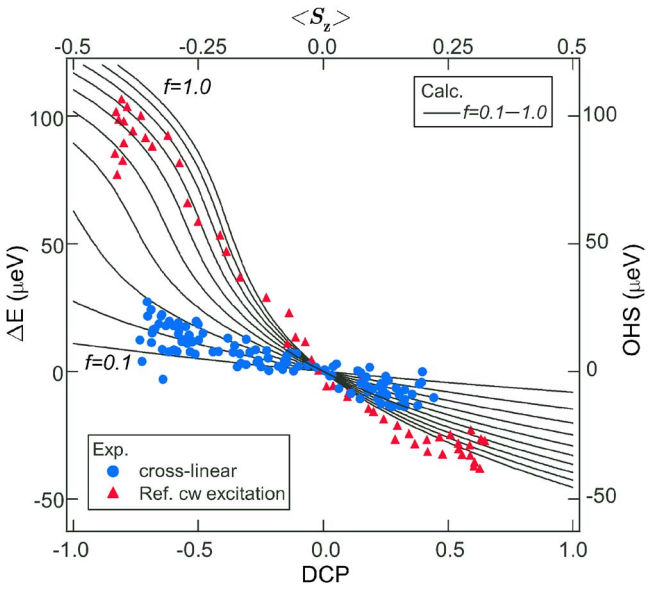

FIG. 5. (Color online) $\langle S_z\rangle$ dependence of OHS, which is calculated by Eqs. (1) and (2) with $f = 0.1-1.0$ (solid curve). DCP dependence of ΔE in cross-linearly polarized excitation [lower panel of Fig. 4(b)] is replotted (solid circle). In addition, we show the reference date (solid triangle) obtained by a different method using cw-TIS laser with an excitation energy ~1.7 eV.

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IV. SUMMARY

The antiphase polarization oscillations of exciton spins in a single QD are clearly observed by changing the interpulse delay time, which suggests that the half-localized state has a large oscillator strength for the optical transition between the Zeeman-split exciton states with opposite spins, in which the direct transition between the Zeeman-split exciton states is optically inactive. The optically created Overhauser field in a QD was monitored through the modulated OHS by changing the delay time between the orthogonally polarized optical pulses, which signifies that the confined electron and hole created in continuum state constitute the half-localized state in a QD.

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