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Citation	Journal of Applied Physics, 116(9), 94309 https://doi.org/10.1063/1.4894712
Issue Date	2014-09-04
Doc URL	http://hdl.handle.net/2115/57544
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Type	article
File Information	1.4894712.pdf



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Citation: *Journal of Applied Physics* **116**, 094309 (2014); doi: 10.1063/1.4894712

View online: <http://dx.doi.org/10.1063/1.4894712>

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Growth-temperature dependence of optical spin-injection dynamics in self-assembled InGaAs quantum dots

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(Received 3 July 2014; accepted 25 August 2014; published online 4 September 2014)

The growth-temperature dependence of the optical spin-injection dynamics in self-assembled quantum dots (QDs) of In_{0.5}Ga_{0.5}As was studied by increasing the sheet density of the dots from 2×10^{10} to 7×10^{10} cm⁻² and reducing their size through a decrease in growth temperature from 500 to 470 °C. The circularly polarized transient photoluminescence (PL) of the resulting QD ensembles was analyzed after optical excitation of spin-polarized carriers in GaAs barriers by using rate equations that take into account spin-injection dynamics such as spin-injection time, spin relaxation during injection, spin-dependent state-filling, and subsequent spin relaxation. The excitation-power dependence of the transient circular polarization of PL in the QDs, which is sensitive to the state-filling effect, was also examined. It was found that a systematic increase occurs in the degree of circular polarization of PL with decreasing growth temperature, which reflects the transient polarization of exciton spin after spin injection. This is attributed to strong suppression of the filling effect for the majority-spin states as the dot-density of the QDs increases.

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I. INTRODUCTION

The spin states and dynamics present in self-assembled quantum dots (QDs) of III-V compound semiconductors have come to receive significant attention as a result of the long spin lifetime of their carriers or excitons.¹⁻⁴ This suppression of spin relaxation in QDs is understood to be essentially a process of motional freezing of carriers combined with weak hole-state mixing, which originates from the strong quantum confinement of the carriers.^{2,3} However, there is still a need to take a closer look at the spin states during the injection of spin-polarized carriers or excitons, i.e., spin injection from a layered semiconductor barrier into a QD.⁵⁻¹⁰ Such spin injection has the potential to provide the key to applying the spin states of QDs to spin-functional optical devices, as one would need to inject spin-polarized carriers from electrodes into QDs to achieve a practical device structure. Indeed, spin-polarized light emitting diodes and lasers based on QDs have been discussed;¹¹⁻¹⁴ however, spin injection is recognized as being much more difficult than spin-independent carrier injection due to the relative instability of the spin states in layered semiconductors, which allows them to easily relax during the injection process.

Recently, we reported that the continuous injection of majority spins into QDs can be blocked by a state-filling effect by the limited density of excited spin-polarized states in the QD side immediately after optical-spin injection in the case of high-density spin excitation.⁵ This results in a decrease in the total spin polarization at excited states in the QDs due to the subsequent accumulation of minority spins,

during the injection of majority spins is blocked. The problem which one has to consider next, however, is precisely what kind of QD system is the most appropriate for achieving efficient spin injection while also maintaining high spin polarization, even in the case of strong excitation or pumping. With this in mind, the growth-temperature dependence of the spin-injection dynamics in QDs was systematically studied, as the growth temperature of InAs and InGaAs QDs is known to significantly affect their size, shape, density, compositional distribution and resultant strain, and also optical quality.¹⁵⁻¹⁷ The transient behavior of the circular polarization property of photoluminescence (PL) in InGaAs QDs after optical spin generation in GaAs barriers, which directly reflects the spin-injection dynamics, is herein discussed in relation to growth temperature through the development of rate equations that incorporate all aspects of spin-injection dynamics for spin-polarized states in QDs, and their subsequent spin relaxation. These results are also compared with the dot structure, with a view to determining the optimal growth conditions for spin-functional applications.

II. EXPERIMENTAL

Optically active layers of In_{0.5}Ga_{0.5}As QDs were epitaxially grown on GaAs (100) substrates by molecular beam epitaxy under growth conditions similar to those used in a previous work.⁵ The exception was the substrate temperature during QD growth (T_s), which for the purposes of this study was varied from 470 to 520 °C. A GaAs barrier was then applied to the QD layer, onto which an additional layer of QDs was grown to allow observation of the dot structure by atomic force microscopy (AFM) and high-resolution scanning electron microscopy (SEM). Cross-sectional

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transmission electron microscopy (TEM) was also used for observation of the layered sample structure.

Three-dimensional calculations of the electronic band structure and resultant wavefunction of carriers in the dots were also made, taking into account the observed size and shape of the dots, as well as their composition and strain. Circularly polarized time-resolved PL spectra were obtained using a previously described method,⁵ wherein optical spin injection was achieved by resonantly exciting the bottom of the energy-band gap of the GaAs barrier using circularly polarized light pulses. The spin-polarized electron-hole pairs were subsequently generated in the barrier in accordance with the optical spin orientation rule,¹⁸ which takes into account the spin states of the carriers and the circular polarization of the excitation light. The circularly polarized transient PL from the excited state of the QDs was observed to be energetically slightly below the barrier level, allowing the spin states after spin injection from the barrier to QDs to be probed before additional spin relaxation could occur inside the QDs. This detection of the PL spectrum was performed using a streak camera with discriminating the circular polarization and a time resolution of 5 ps after convolution analysis.

III. RESULTS AND DISCUSSION

From the growth-temperature dependence of the QD structure shown in Figure 1, we can see that there is a systematic increase in dot diameter with temperature; the peak of the distribution from 15 to 30 nm quite clearly shifting as T_s is increased from 470 to 500 °C. Furthermore, the plot of average dot diameter and sheet density as a function of T_s shown in Figure 1(c) indicates that the dot density systematically

increases from 2×10^{10} to $7 \times 10^{10} \text{ cm}^{-2}$ with decreasing T_s . Both these trends are quite typical of this InGaAs QD system, and originate from the thermal migration of In and Ga on the GaAs surface. It should be noted here that the surface morphology with $T_s = 520 \text{ °C}$ was rather rough, with irregular bumps and valleys on a scale of several tens nm, and thus this sample was omitted from the subsequent optical study. Cross-sectional TEM images of the samples show layered structures of the dots with wetting layers (WLs), as shown in Figs. 2(a)–2(c). Figure 2(d) shows a typical three-dimensional calculation of eigenvalues of the electron and hole wavefunctions along the thickness direction in the QD grown at 480 °C, where the structural properties, such as, the dot shape including the WL, average size, composition, and resultant strain, were taken into account. As can be seen, sufficiently confined electron and hole states are situated in the QD with lower potentials than those of the GaAs barrier. The spin injection can be induced by these potential differences with phonon emissions. The most probable path of the injection is tunneling from the barrier to WL through gentle potential barriers due to strain. The direct tunneling from the barrier to the QD is also possible; however, this probability seems to be lower because of the difference of dimensionality of the wavefunction between the barrier and QD. After the tunneling to the WL, the spin-polarized carriers can rapidly migrate and be injected into the QD from the WL before spin relaxation via energy relaxation with efficient phonon emissions.

Figure 3 shows the circularly polarized PL spectra obtained at 20 K and with an excitation power of 20 mW from the QD samples grown with various T_s . In this, the degree of circular polarization (CPD) is defined as $(I_{\sigma_+} - I_{\sigma_-}) / (I_{\sigma_+} + I_{\sigma_-})$, with the circularly polarized PL intensities given

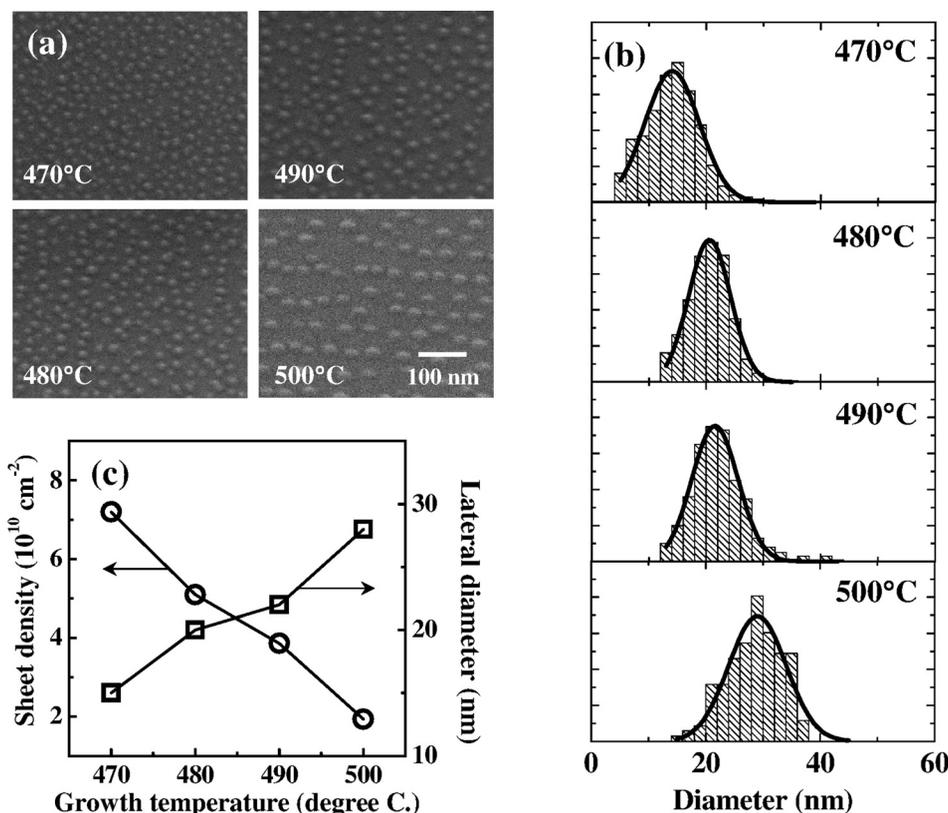


FIG. 1. (a) SEM images showing the surface of In_{0.5}Ga_{0.5}As-QD layers formed using different substrate temperatures (T_s). (b) Distribution of lateral diameter for the QDs observed in the SEM images as a function of T_s . (c) Average diameter (open squares) and sheet density (open circles) as a function of T_s . The solid lines are intended only as a guide for the eyes.

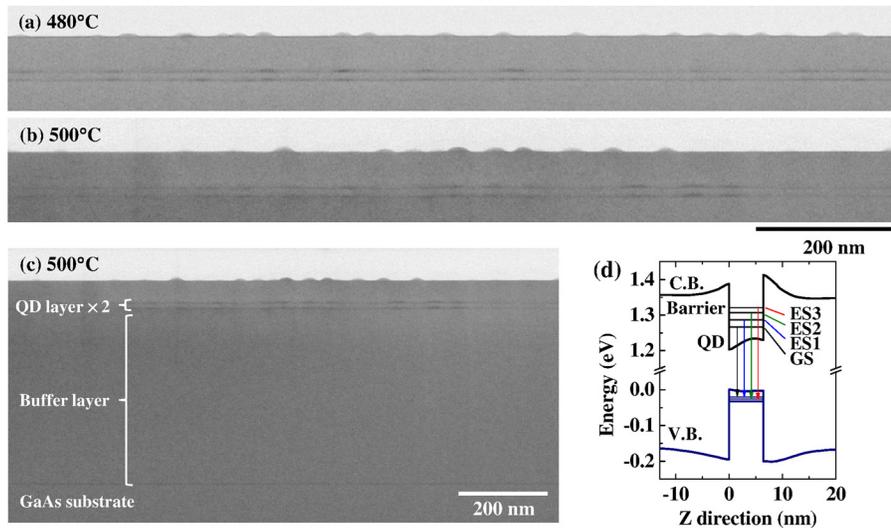


FIG. 2. Cross-sectional TEM images of the QD samples grown at (a) $T_s = 480$ and (b) 500°C . (c) The whole layered structure of the latter QD sample on a GaAs substrate is also shown. (d) Three-dimensional calculation results of the potential profile and eigenvalues of the electron and hole wavefunctions for the case of QD grown at $T_s = 480^\circ\text{C}$.

as I_{σ^\pm} . The excitation of the GaAs barrier with the energy of 1.506 eV was achieved using these circularly polarized light pulses (σ^+ in this case) for the purposes of optical spin generation. Each PL spectrum is composed of emissions from excited states in the QD ensemble, with the intensity contributions dependent on the dot structure and therefore T_s . Calculated results for the energies of these excited levels are indicated by arrows, though it should be noted that the PL intensity appears significantly lower at energies less than 1.3 eV due to this being the detection limit of the streak camera used. Consequently, only the higher-region of the emission bands was used to determine the circularly polarized PL intensity and corresponding CPD as a function of time. This emission region corresponds to the exciton energies of excited states energetically slightly below the GaAs barrier, making the time dependences of the circularly polarized PL a reflection of the spin states of excitons immediately after spin injection from the

barrier into the QDs. The positive CPD values are caused by a co-circular PL property for the excitation polarization, which is indicative of spin conservation after spin injection. These CPD values remain high (~ 0.55) at the higher-energy tail of the PL spectrum, the notable exception being the 500°C sample, in which the CPD decreases monotonically with decreasing exciton energy toward the spectral peak. It can be seen from this that CPD values around the PL-spectral peak depend on T_s , and increase systematically with decreasing T_s .

To understand the dependence of the CPD value on T_s , and its relationship to spin-injection dynamics, the transient circularly polarized PL was analyzed using rate equations that took into account all parameters responsible for the spin injection dynamics. Figure 4 shows the circularly polarized PL intensity and corresponding CPD value for the PL spectral peak associated with an energy window of 25 meV as a function of time following the pulsed excitation of spin-

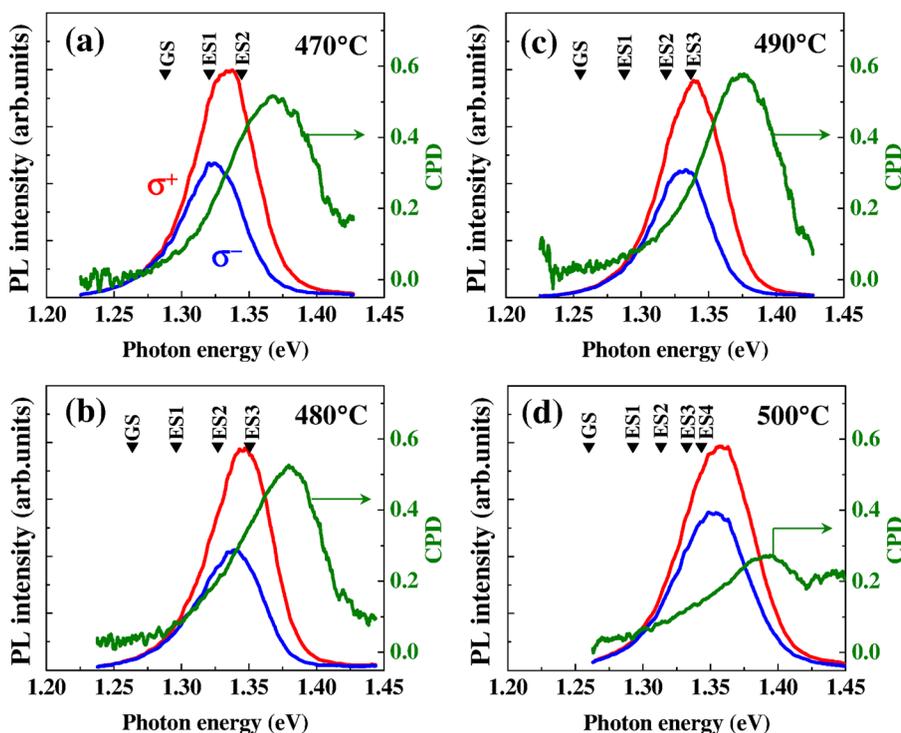


FIG. 3. Circularly polarized PL spectra (red solid lines: σ^+ and blue solid lines: σ^-) and corresponding CPD values (green solid lines) at 20 K for QD samples grown at (a) 470 , (b) 480 , (c) 490 , and (d) 500°C under conditions of σ^+ -polarized excitation with a 20 mW for the GaAs barrier. The energies of the ground state (GS), and the 1st (ES1), 2nd (ES2), 3rd (ES3), or 4th (ES4) excited state, are indicated by arrows.

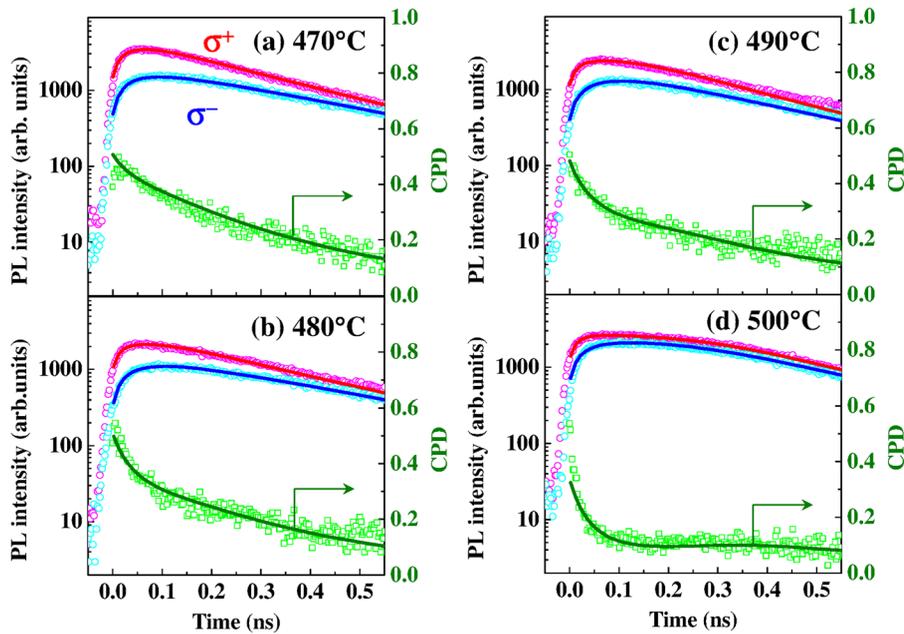


FIG. 4. Circularly polarized PL time-profiles (red open circles: σ^+ and blue open circles: σ^-) and corresponding CPD values (green open squares) at 20 K for QD samples grown at (a) 470, (b) 480, (c) 490, and (d) 500 °C under conditions of σ^+ -polarized excitation with a 20 mW. The solid lines represent best-fit calculations of the rate equations.

polarized carriers at 20 K and 20 mW. The best-fitted rate-equation calculations are shown by solid lines, with specific details of the rate-equation analysis described in a previous paper.⁵ The rate-equation model is schematically illustrated in Fig. 5(a), where parameters responsible for the injection of spin-polarized excitons and subsequent relaxation are included. The spin-polarized excitons generated in the barrier are injected into excited states in the QD with a time constant of τ_{inj} and relax via the spin-relaxation ($\tau_{QD}^{spin-relax}$), non-radiative decaying process to lower-lying states ($\tau_{non-rad}^{relax}$), and radiative recombination (τ_{rad}^{relax}). A state-filling effect is also included in this model, where the initial spin polarization during the injection is expressed by a parameter of η , where $\eta = 1$ corresponds to the case of full polarization while $\eta = 0.5$ corresponds to the random polarization. If the excited state in the QD is fully occupied, the population of the state ($N_{QD}^{\sigma_{\pm}}$) is equal to the density of the state ($D_{QD}^{\sigma_{\pm}}$) and thus the injection from the barrier stops. This situation can be expressed by introducing parameters of $1 - N_{QD}^{\sigma_{\pm}}/D_{QD}^{\sigma_{\pm}}$. These results demonstrate that CPD decays monotonically with increasing time in samples with a relatively low T_s of 470 or 480 °C, which is attributed to spin relaxation after spin injection. In contrast, those samples with a higher T_s of 490 or 500 °C show a steep decrease in the CPD value immediately after pulsed excitation due to a filling effect related to the limited density of states in QDs.^{5,19} The mechanism of the spin filling blocking is schematically illustrated in Fig. 5(b). This filling effect can have a significant effect on the spin polarization in the spin injection process, as it allows the majority-spin states of the excited states in the QDs to be rapidly occupied by spin-polarized excitons immediately after injection of spin-polarized excitons from the barrier. However, once these majority-spin states are filled with spin-polarized carriers or excitons, any additional spin injection is effectively blocked by the state filling (spin-dependent Pauli blocking).^{5,20,21} When this state filling is active minority spins can still continue to be injected, despite the

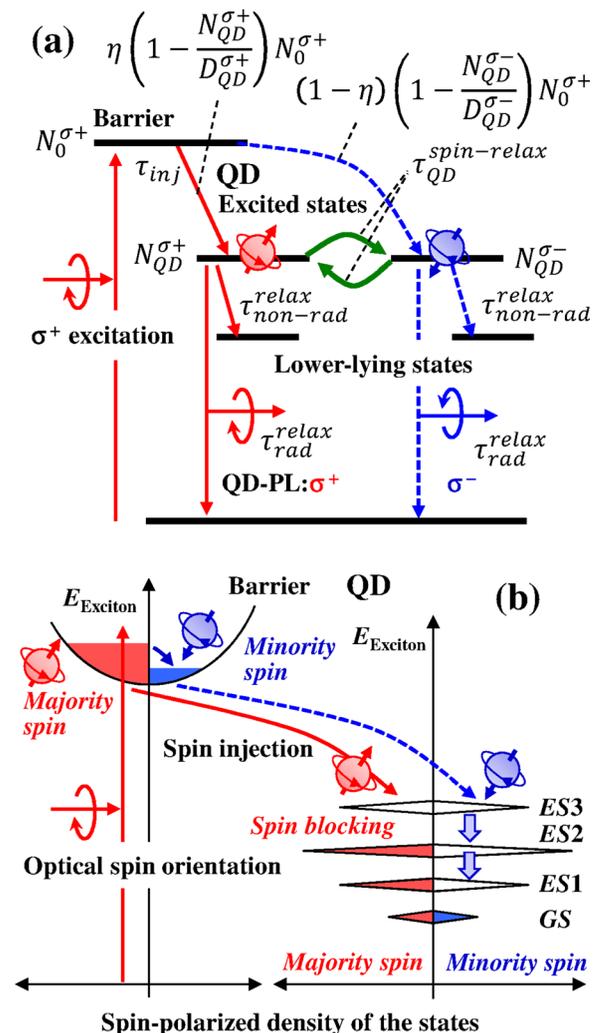


FIG. 5. (a) A rate-equation model for the spin injection. (b) A schematic illustration indicating the spin-injection dynamics from the barrier into spin-polarized excited states in the QD, where the spin blocking effect due to the state filling is included.

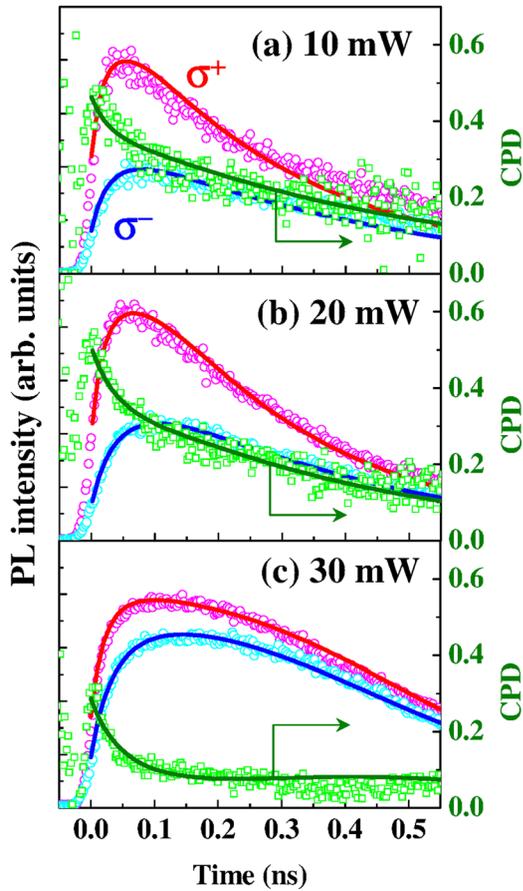


FIG. 6. Circularly polarized PL time-profiles (red open circles: σ^+ and blue open circles: σ^-) and corresponding CPD values (green open squares) at 20 K for QD samples grown at 480 °C with excitation powers of (a) 10, (b) 20, and (c) 30 mW under σ^+ -polarized excitation conditions. The solid lines represent best-fit calculations of the rate equations.

blocking of majority spins, resulting in a decrease in the total spin polarization. The plateau-like behavior of the time-dependent CPD that is observed after the steep decrease from 0.1 to 0.4 ns can therefore be attributed to a suppression of the spin-dependent filling effect caused by a decay of the majority-spins due to spin relaxation, energy relaxation to lower lying states, and radiative recombination.

Furthermore, the fact that the characteristic time-dependent CPD profile is dependent on the excitation power (Fig. 6) can also be easily explained by spin-dependent Pauli blocking.⁵ At a high power of 30 mW, there is a steep decrease in CPD to a much smaller value than is seen at 10 or 20 mW, with a plateau-like behavior observed with both the PL intensity and CPD value as a function of time beyond this point.

Figure 7 shows the excitation-power dependencies of the spin-injection parameters obtained from the fitted rate equations for a T_s of 480 and 500 °C. The marked difference between the two is attributed to the aforementioned filling effect, which is expressed as N_0/D_s , where N_0 is the initial population of the majority spin in the barrier and D_s is the density of the majority spin states in the QDs. This parameter can also be deduced from the rate-equation fitting, wherein a higher value of N_0/D_s means a stronger filling effect. Thus, the filling effect is weak and nearly constant at a $T_s = 480$ °C, except in the case of a 30 mW excitation power. However, at 500 °C, N_0/D_s is significantly increased from an excitation power as low as 10 mW. The CPD decreases monotonically from 0.21 to 0.03 with this increase in power from 5 to 30 mW. It is known that the CPD value can be affected by the relationship between the spin-relaxation time τ_s and PL decay time τ_r ,²² however, this seems unlikely in this instance due to the fact that τ_s and τ_r are almost identical for the excitation power, as shown in the bottom panel. The power dependence of the CPD value at $T_s = 500$ °C can therefore be attributed to the spin-state filling effect indicated by N_0/D_s , which increases monotonically with power. The fact that the time-integrated CPD value is inversely proportional to the filling parameter clearly indicates that the filling effect can degrade spin polarization during spin injection. On the other hand, the power dependence of the CPD at $T_s = 480$ °C shows the maximum at 15 mW. This trend will be discussed later. The spin-injection time τ_{inj} shows a systematic increase with increasing power. We observe that the values of τ_{inj} are almost identical among all samples with different T_s and this excitation-power dependence is also common. Therefore, one reasonable explanation of this excitation-power dependence of τ_{inj} is a state filling effect in a wetting layer where the density of the states is limited

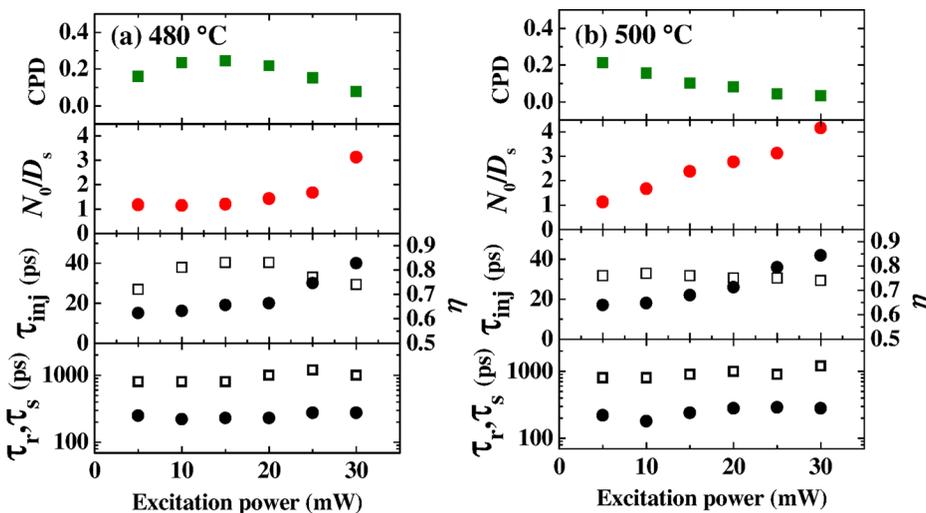


FIG. 7. Spin-injection parameters as a function of excitation power. Top panel: time-integrated CPD values. Second panel: N_0/D_s parameter, which indicates the spin-state filling effect. Third panel: (solid circles) spin injection time, τ_{inj} ; and (open squares) the η parameter, which indicates the initial spin relaxation in the spin-injection process. Fourth panel: (open squares) spin relaxation time, τ_s ; and (closed circles) the decay time of PL τ_r , as determined from the excited state of the QDs with a $T_s =$ (a) 480 or (b) 500 °C.

originating from being very thin layered structure. Carriers or excitons generated in the barrier can rapidly flow into the wetting layer. In the case of high excitation, this rapid flow can be blocked by the limited density of the states, and the subsequent migration from the wetting layer into the QDs slows down and can continue. However, note that the change of τ_{inj} cannot significantly affect the spin polarization after the spin injection into the QDs. The spin relaxation during injection can be expressed by the parameter of η , where $\eta = 1$ corresponds to 100% conservation of the spin state during injection and $\eta = 0.5$ corresponds to 100% relaxation (i.e., the number of majority and minority spins becomes identical), as introduced in above description about the rate-equation model. The decreases in η at high excitation powers are seen above 15 mW ($T_s = 480^\circ\text{C}$) and 10 mW (500°C , slightly in this case), which can be explained by the Bir-Aranov-Pikus (BAP) mechanism;²³ that is, the number of electron-hole pairs in the barrier increases at higher powers, thereby increasing the effective concentration of the hole-spin relative to the electron-spin. The resulting higher effective field of the hole-spin can then enhance the extent of electron-spin relaxation prior to the excitons reaching the emissive excited states in the QDs.^{23,24}

The relation between growth-temperature and the most important spin-injection parameters responsible for spin polarization in QDs is summarized in Fig. 8. As can be seen from this, the CPD value depends greatly on the T_s and excitation power; with the decrease in value seen in the high-power region of >15 mW for $T_s \leq 490^\circ\text{C}$, or 5 mW for $T_s = 500^\circ\text{C}$, readily explained by the aforementioned spin-state filling effect. There is also a systematic decrease evident in the N_0/D_s value with decreasing T_s (except at 5 mW), which indicates a dissipation of the spin-dependent filling effect and subsequent enhancement of the CPD. A weaker filling effect in those samples with a lower T_s is reasonable to expect, as the filling effect is a function of the total number of states, i.e., the number of dots. Since all measurements in this study were performed under identical excitation conditions in terms of excitation power and the size of the light spot, the number of dots involved in spin-injection is directly proportional to the dot density. On the other hand, the CPD values of the samples with a $T_s \leq 490^\circ\text{C}$ exhibit a maximum of around 0.25 at 15 mW, and then decrease toward 5 mW. This can be interpreted by considering the spin-injection dynamics, in particular, the initial spin relaxation η which decreases with decreasing power below 15 mW. The sample produced with a $T_s = 470^\circ\text{C}$ shows slightly different properties, in that its η value is generally lower (except at 30 mW), which results in the CPD value being degraded at powers of less than 15 mW. Given that the initial spin polarization can be affected by spin relaxation during the migration of spin-polarized carriers in the barriers prior to the spin injection, these lower values of η are attributed to enhanced spin relaxation in the barrier. This is most likely caused by the fact that the initial barrier with the thickness of 10 nm was grown at the substrate temperature used for QD growth, with 470°C seemingly insufficient to ensure high-quality GaAs despite clear streak patterns being observed immediately after capping the ultrathin GaAs barriers to a thickness of more

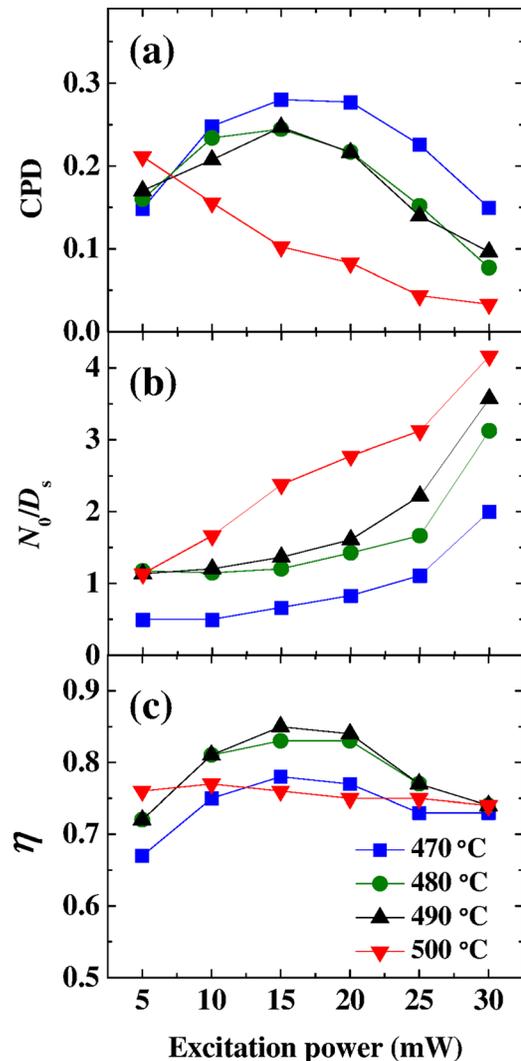


FIG. 8. (a) PL-CPD values, (b) saturation factor indicating the filling strength, N_0/D_s , and (c) the initial spin relaxation, η as a function of excitation power for samples with a T_s of 470 (blue solid squares), 480 (green solid circles), 490 (black solid triangles), or 500°C (red solid inverted triangles).

than 2 nm. Furthermore, spin relaxation is known to be extremely sensitive to the presence of defects in 2- or 3-dimensional GaAs.²⁵ An excitation-power dependence of η is also seen in the samples with a $T_s \leq 490^\circ\text{C}$, with the decrease in η at high powers again originating from the BAP mechanism described earlier.

In terms of practical application, it is evident from the efficient spin injection with highly conserved spin states demonstrated in this study that higher-density QDs of InGaAs can offer a good platform for spin-functional photonic devices. In fact, a higher spin polarization after spin injection is considered essential in spin-polarized light emitting diode and laser devices that are based on the injection of spin-polarized electrons from metallic ferromagnetic electrodes. High-density dots can be obtained by optimizing growth conditions such as temperature, while still maintaining a high optical quality. This is evidenced by the fact that the time constants responsible for the spin-injection dynamics (e.g., τ_{inj} , τ_s , and τ_r) are sensitive to the optical quality of QDs, yet remained almost identical among samples with

various T_s during this study. It should be noted, however, that the PL intensity of the sample produced at the lowest temperature of 470 °C was similar to the other samples, which given its higher dot density suggests that its optical performance was lower to some extent. That is, if we assume that PL intensity is proportional to dot density, then it is clear that a proportion of the dots are not optically active. This means that any optimization of the growth conditions needs to not only take into account the spin-injection efficiency but also the optical quality. Finally, it should be emphasized that a higher-density dot system is essential for QD-based optical devices to offer high performance in terms of bright PL and high-gain lasing with lower threshold currents. The future direction of high-density QD-based spintronic device development should therefore be focused on at least matching the performance of conventional QD lasers, which will require a lateral dot-density greater than $5 \times 10^{10} \text{ cm}^{-2}$ to ensure excellent performance with high optical or modal gains.^{26,27}

IV. CONCLUSIONS

By studying the growth-temperature dependence of the optical spin-injection dynamics in $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$ QDs, it has been found that the filling effect associated with the excited majority-spin states in QDs can be suppressed by increasing the dot density through a reduction in growth temperature. This suppression of spin blocking is highly effective in achieving spin conservation during the spin-injection process into QDs, which suggests that a high-density QD system would be highly beneficial to spin-polarized photonic applications by allowing efficient spin injection without significant loss of spin information. This direction of the high-density QD-based spintronic device development will closely match the requirement for conventional QD lasers with high optical gains.

ACKNOWLEDGMENTS

This work was supported by the Japan Society for the Promotion of Science (JSPS), under a Grant-in-Aid for Scientific Research (S) No. 22221007. One of the authors (T.K.) also gratefully acknowledges the support received from the Japan Science and Technology Agency (CREST).

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