Room temperature Optical Orientation of Exciton Spin in cubic GaN/AlN quantum dots

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The optical orientation of the exciton spin in an ensemble of self-organized cubic GaN/AlN quantum dots is studied by time-resolved photoluminescence. Under a polarized quasi-resonant excitation, the luminescence linear polarization exhibits no temporal decay, even at room temperature. This demonstrates the robustness of the exciton spin polarization in these cubic nitride nanostructures, with characteristic decay times longer than 10 ns.

Wide bandgap GaN-based semiconductors and related heterostructures represent promising candidates for spintronic applications. The weak spin-orbit coupling in these materials should yield long electron spin relaxation times, and their large exciton binding energy (~26 meV in bulk GaN) should allow the control and manipulation of the exciton spin, even at high temperature.

Compared to GaAs-based structures, very few measurements of the carrier spin properties have been performed on these wide bandgap nitrides which can crystallize either in the wurtzite (Wz) or zinc-blende cubic (ZB) structure. Most of these studies deal with the measurement of the electron or hole spin dynamics in Wz GaN or InGaN structures. Beschoten et al. measured an electron spin lifetime of ~35 ps at room temperature in n-doped Wz epilayers. A hole spin coherence time of ~120 ps was found in p-type bulk Wz GaN at low temperature. However, a very fast exciton spin relaxation time, of the order of 1 ps, was deduced from transient reflectivity or spin gratings experiments in unintentionally doped Wz GaN epilayers. In Wz nitride nanostructures (quantum wells or quantum dots), exciton spin relaxation times of the order of 200 ps were also reported at $T = 300$ K with a rather weak temperature dependence.

In all these wurtzite nitride structures, the electronic and spin properties are highly affected by the strong built-in electric field due to the spontaneous and piezoelectric polarizations. In contrast to the usual behavior in other III-V or II-VI nanostructures, the exciton spin dynamics in cubic nitride nanostructures is usually weaker thanks to the carrier confinement.

We present in this letter a detailed time-resolved investigation of the exciton spin dynamics in self-organized ZB GaN/AlN quantum dots (QDs). These optical orientation experiments clearly evidence an exciton linear polarization which persists at room temperature, in contrast to the usual behavior in other III-V or II-VI nanostructures.

The QD sample investigated here consists in 18 self-assembled ZB GaN quantum dot planes in AlN barriers, grown on 3C-SiC(001)/Si pseudosubstrate by molecular-beam epitaxy. The dots have a typical diameter of 12 nm and a height around 1.5 nm. The sample is nominally undoped and the areal dot density is of the order of $10^{11}$ cm$^{-2}$. In time-resolved photoluminescence (PL) experiments, the excitation source is provided by a mode-locked frequency tripled Ti:Sa laser, with a 1.5 ps pulse width and a tunable wavelength in the range 260-310 nm. The laser beam is focused onto the sample to a 100 µm diameter spot with an average power $P_{exc} = 1$ mW. The PL signal is dispersed by an imaging spectrometer and then temporally resolved by a S20 photocathode streak camera with an overall time resolution of 8 ps. The linear polarization degree of the luminescence is defined as $P_{lin} = (I^X - I^Y)/(I^X + I^Y)$. Here $I^X$ ($I^Y$) denotes the $X$($Y$) linearly polarized PL components ($X$ and $Y$ are chosen parallel to the [110] and [110] crystallographic directions).

The inset of Fig. 1 presents the time-integrated PL spectrum measured at $T = 20$ K for a non-resonant excitation energy $E_{exc} = 4.77$ eV. The ground state emission is centered around 4.15 eV with a full width at half maximum of 250 meV which results from the inhomogeneous broadening of the ground state exciton energies due to the size and strain distributions among the QD ensem-
as it was observed in Wz InGaN QDs induced by the strain or by the QD shape anisotropy, amonds) does not arise from the valence state mixing angles) proves that the measured polarization (solid di-polarized excitation (σ+)) of the time-integrated PL emission spectrum is measured in this case due to the spectral selectivity of the excitation. The absence of linear polarization following a circularly-polarized (σ−) QR excitation is also plotted. Inset: Time-integrated PL spectra of the QD for quasi-resonant excitation (E_{exc} = 4.11 eV) and non-resonant excitation (E_{exc} = 4.77 eV). The excitonic properties of nitride-based ZB QDs are still poorly understood. However, from similar results on InAs, CdTe or CdSe self-organized QDs, the experimental data presented above strongly suggest that the linear polarization in Fig. 1 is indeed due to the light-induced optical alignment of excitons in the QDs. Let us furthermore recall that the observation of optical pumping in linear configuration is a clear signature of intrinsic exciton emission: the loss of coherence between electron and hole spins, inherent in the impurity bound excitons or charged excitons, inhibits the optical alignment of these pseudo-particles.

FIG. 1: Time-integrated PL spectrum under a quasi-resonant excitation at T = 20 K. The linear polarization degree following a linearly-polarized (σ+) or circularly-polarized (△) excitation is also plotted. Inset: Time-integrated PL spectra of the QD for quasi-resonant excitation (E_{exc} = 4.11 eV) and non-resonant excitation (E_{exc} = 4.77 eV).

Fig. 2 displays the time evolution of the co-polarized I^X (○) and counter-polarized I^Y (+) to the σ^X polarized excitation laser at T = 20 K and the corresponding linear polarization degree P_{Lin} (●) . E_{exc} = 4.11 eV, E_{det} = 4.02 eV.

FIG. 2: Temporal dependence of the PL components copolarized I^X (○) and counter-polarized I^Y (+) to the σ^X polarized excitation laser at T = 20 K and the corresponding linear polarization degree P_{Lin} (●). E_{exc} = 4.11 eV, E_{det} = 4.02 eV.
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PL linear polarization detected at a fixed energy

FIG. 3: PL linear polarization detected at a fixed energy $E_{\text{det}} = 4.02$ eV as a function of excitation energy at $T = 20$

K.Inset: Spectral dependence of the co-$(I^X)$ and counter-
polarized $(I^Y)$ PL components and the associated linear po-
larization degree for $E_{\text{exc}} = 4.11$ eV.

FIG. 4: (a) Linear polarization dynamics for $T = 20$, 150 and
300 K. (b) Temperature dependence of the linear polarization
degree (the full line is a guide to the eyes). For $T = 20$ K,
the excitation and detection energies are $E_{\text{exc}} = 4.11$ eV and
$E_{\text{det}} = 4.02$ eV, respectively. For higher temperature, the
excitation and detection conditions are detailed in the text.

directions. This exciton fine structure may originate from
QD elongation and/or interface anisotropy which yield a
significant anisotropic exchange interaction between the
electron and the hole forming the exciton. No circular
polarization of the PL emission was observed following
a circularly-polarized excitation. Similarly, no PL linear
polarization was detected after a linearly-polarized excita-
tion along $[100]$ or $[010]$ (not shown). This is consistent
with $|X\rangle$ and $|Y\rangle$ linearly-polarized exciton eigenstates
in an inhomogeneous QD ensemble. When the polarized
excitation creates a coherent superposition of these exciton
eigenstates, the anisotropic exchange energy statistical
fluctuations among the detected QD ensemble prevent
the observation of spin quantum beats.$^{13,15}$

The relative low value of the linear polarization

$(P_{\text{Lin}} \sim 20 \%)$ observed in Fig.2 is mainly due to the fact
that the experiments are not performed under strictly
resonant conditions (which could not be performed with
our present experimental set-up because of the large laser
scattering on the sample surface). In these QR excita-
tion conditions, both the energy relaxation processes and
the excitation of exciton states with different symmetries
contribute to decrease the measured $P_{\text{Lin}}$. Nevertheless,
the exciton spin relaxation quenching, evidenced by the
absence of decay of the linear polarization, is a striking
feature here. Already observed in other self-organized
QD structures at low temperature, it proves that neither
the electron nor the hole spin relax on the exciton lifetime
scale and that once the exciton occupies the ground state
of the QD, no transient change of the polarization occurs
within the time window under investigation.$^{13,15,27}$

Fig. 5 displays the degree of the PL linear polarization
detected at a fixed energy $(E_{\text{det}} = 4.01$ eV) as a function
of the excitation energy. The average $P_{\text{Lin}}$ decreases from
20 to 0 % when the excess energy $E_{\text{exc}} - E_{\text{det}}$ increases
from $\sim 90$ to 400 meV. This behavior is consistent with
the variation of the linear polarization in the PL spec-
trum measured in Fig. 1. The decrease of $P_{\text{Lin}}$ appears as
a monotonic function of the excess energy, in agreement
with the pioneering work in bulk CdS or recent results
in CdSe/ZnSe self-organized QDs.$^{20,28}$ Nevertheless, no
distinct peaks due to LO-phonon cascade processes are
evidenced. The absence of such peaks in Fig. 3 is prob-
ably due to the large inhomogeneous broadening in ZB
GaN QDs. We emphasize that for $E_{\text{exc}} - E_{\text{det}} > 400$
meV, no linear polarization is measured for any detec-
tion energy in the PL spectrum.

FIG. 5: Linear polarization degree as a function of excitation and
detection energies for $T = 20$ K. (a) $T$ = 20, 150 and
300 K. (b) Temperature dependence of the linear polarization
degree (the full line is a guide to the eyes). For $T = 20$ K,
the excitation and detection energies are $E_{\text{exc}} = 4.11$ eV and
$E_{\text{det}} = 4.02$ eV, respectively. For higher temperature, the
excitation and detection conditions are detailed in the text.

Fig. 4(a) presents the dependence of the exciton PL
linear polarization dynamics upon the lattice tempera-
ture. As the amplitude of the linear polarization de-
deps on the detection energy in the spectrum (Fig. 1),
we varied the excitation and detection energies for the
different temperatures following a simple Varshni law$^{29}$
keeping constant $E_{\text{exc}} - E_{\text{det}} = 90$ meV. This makes pos-
sible the measurement of the temperature dependence
of the exciton spin properties for the same QDs. It
is worth mentioning that we obtained qualitatively the
same results for fixed excitation and detection energies
(not shown). Contrary to the reported results in ZB
bulk GaN,$^{22}$ optical orientation of exciton spin is clearly
observed in Fig. 4 above 75 K. Moreover, no temporal
decay of $P_{\text{Lin}}$ is observed, even for high temperatures.
This result is to be compared with those obtained on
QDs made of other semiconductor materials (InAs, CdSe,
CdTe)$^{13,14,15}$. In these QDs, the linear polarization decay
time, usually longer than the exciton lifetime at $T=10$ K,
rapidly drops when the temperature increases a few
tens of K$^{13,14}$. Thus, the robustness of the exciton linear
polarization up to room temperature appears here as a
unique property of the ZB GaN QDs. Though we have
never observed any temporal decay of $P_{\text{lin}}$ on the exciton lifetime scale, a clear decrease of the amplitude of the linear polarization is evidenced. The uncertainty on the measurements in Fig. 1(b) does not allow us to extract an accurate thermal activation energy (it lies within the 50-100 meV range which is a plausible value for the exciton binding energy in these structures).\(^{30,31}\)

Finally, we have investigated the possible conversion from optical alignment to optical orientation of excitons when a magnetic field is applied along the growth direction (Faraday geometry).\(^{11,12}\) If the exciton Zeeman splitting $\Delta E_z = g\mu_B B$ is much larger than the anisotropic exchange energy, the QD exciton eigenstates are no longer the exciton $|X\rangle$ and $|Y\rangle$ linearly-polarized states but the $|+1\rangle$ and $|-1\rangle$ circular ones. However, this conversion has not been observed for any magnetic field up to 4 T (the maximum field available on our experimental set-up), probably because of the very large value of the exchange energy compared to the exciton Zeeman term. Much stronger magnetic fields would be required to evidence these effects.

In conclusion, we have studied the optical orientation of exciton spin in self-organized cubic GaN/AlN quantum dots. We observe no measurable temporal decay of the exciton linear luminescence polarization at any investigated temperature. Even at room temperature, the exciton spin decay time is larger than 10 ns, i.e. 2 or 3 orders of magnitude longer than in wurtzite-type structures. Moreover, these results contrast with the fast exciton spin relaxation previously observed in other III-V or II-VI QD systems at high temperature.

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