Exciton spin relaxation in InAs/InGaAlAs/InP(001) quantum dashes emitting near 1.55 μm

M. Syperek,[1] Ł. Dusanowski,[1] M. Gawełczyk,[1,2] G. Sęk,[1] A. Somers,[3] J.P. Reithmaier,[3,4] S. Höfling,[3,5] and J. Misiewicz[1]

[1] *Laboratory for Optical Spectroscopy of Nanostructures, Division of Experimental Physics, Faculty of Fundamental Problems of Technology, Wrocław University of Science and Technology, Wybrzeże Wyspiańskiego 27, 50-370 Wrocław, Poland*

[2] *Division of Theoretical Physics, Faculty of Fundamental Problems of Technology, Wrocław University of Science and Technology, Wybrzeże Wyspiańskiego 27, 50-370 Wrocław, Poland*

[3] *Technische Physik, University of Würzburg and Wilhelm-Conrad-Röntgen-Research Center for Complex Material Systems, Am Hubland, D-97074 Würzburg, Germany*

[4] *Institute of Nanostructure Technologies and Analytics (INA), CINSaT, University of Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany*

[5] *SUPA, School of Physics and Astronomy, University of St. Andrews, North Haugh, KY16 9SS St. Andrews, United Kingdom*

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Exciton spin and related optical polarization in self-assembled InAs/In$_{0.53}$Ga$_{0.23}$Al$_{0.24}$As/InP(001) quantum dashes emitting at 1.55 μm are investigated by means of polarization- and time-resolved photoluminescence as well as photolumienescence excitation spectroscopy in cryogenic temperatures. We investigate the influence of highly non-resonant and quasi-resonant optical spin pumping conditions on the spin polarization and memory of the quantum dash ground state. We show the spin pumping scheme, utilizing the longitudinal optical phonon-mediated coherent scattering process, can lead to the polarization degree of nearly 50%. We discuss the role of intrinsic asymmetries in the dash that influence values of the degree of polarization and its time evolution.

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Self-assembled InAs quantum dashes (QDashes) epitaxially grown on InP(001) substrate resemble quantum dots (QDs), however, strongly elongated in one of the inplane dimensions.[1–4] So far, such QDashes have been exploited mostly as a gain medium in semiconductor lasers, amplifiers, or superluminescent diodes suited for telecom technology operating at 1.3 and 1.55 μm low-loss spectral windows of silica fibers.[5–7] Recent research promises new possible applications of QDashes in long-haul secure quantum data transmission lines.[8,9] An InAs/InP(001) QDash-based non-classical single photon emitter operating at 1.55 μm has been demonstrated[8] along with a possibility to tune the exciton fine structure splitting down to zero by applying external magnetic field[9]. While the former demonstrates a capability of QDashes to generate a single photon at a time, the latter can lead to the generation of polarization-entangled photon pairs at telecom wavelengths essential for, e.g., quantum repeater technology. Since semiconductor QDashes can be considered as a bridge platform between the solid-state quantum information storage/operation and the quantum state of light, it is therefore crucial to investigate properties of confined spin states that can mediate the exchange process of quantum information.

The effects concerning spin excitation and spin-related phenomena in self-assembled quasi-0D quantum systems capable of generating photons at 1.55 μm wavelength have not been investigated very extensively so far. Existing reports address only the problem of either exciton or electron/hole g-factors in InAs/InP QDs.[10–13] However, issues such as the longitudinal or transverse spin relaxation or the role of a spin pumping scheme on the spin memory effect in this particular quantum system have not been explored up to date. In this letter, we investigate properties of polarized emission and spin states of excitons confined in an ensemble of InAs/In$_{0.53}$Ga$_{0.23}$Al$_{0.24}$As/InP(001) QDashes by means of polarization- and time-resolved photoluminescence (TRPL), and photoluminescence excitation spectroscopy (PLE). We demonstrate various schemes of spin injection and their impact on the spin memory effect in QDashes emitting near 1.55 μm. The investigated structure was grown in an EIKO gas source molecular-beam epitaxy system on a sulfur-doped InP(001) substrate. The structure consists of QDashes formed in the Stranski-Krastanow growth process by a deposition of nominally 1.3 nm-thick InAs layer at 470°C onto a 200 nm-thick In$_{0.53}$Ga$_{0.23}$Al$_{0.24}$As barrier. QDashes were covered by 100 nm of In$_{0.53}$Ga$_{0.23}$Al$_{0.24}$As and the layer sequence was finalized by a 10 nm-thick InP cap layer. Both In$_{0.53}$Ga$_{0.23}$Al$_{0.24}$As barriers are lattice matched to InP and were grown at 500°C. Structural data reveal that QDashes are triangular in a cross-section with 20 nm in base width and 3.5 nm in height. The length varies between 50 and hundreds of nanometers. The areal density of QDashes is $\sim 5\times 10^{10}$cm^{-2}. QDashes are nominally undoped, however, a small residual electron doping may be present. For time-integrated (PL) and time-resolved

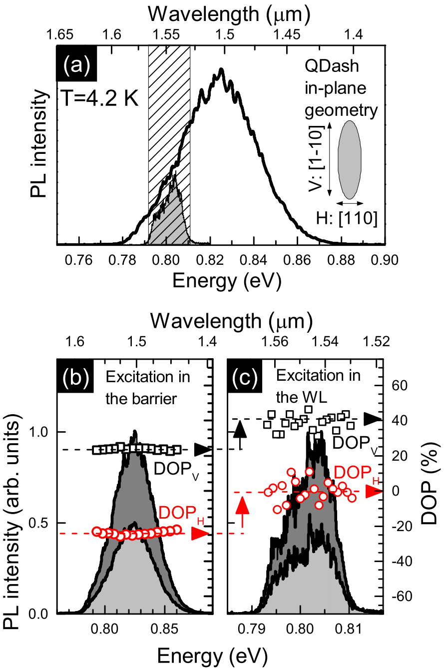

FIG. 1. (a) Low temperature (T=4.2 K) PL spectrum of InAs/In$_{0.53}$Ga$_{0.23}$Al$_{0.24}$As/InP(001) QDashes (E$_{\rm exc}$=1.49 eV, $P_{\rm exc}$=0.4 μW/μm^{-2}). Shaded area shows the spectral range defined by the bandpass filter. (b), (c) The degree of linear polarization (DOP) for the two in-plane directions (H, V) obtained under the pulse excitation in the barrier (E$_{\rm exc}$=1.49 eV), and in the WL (E$_{\rm exc}$=1.1 eV), respectively. (Color online)

photoluminescence (TRPL) experiments the structure was held in a continuous flow liquid helium cryostat at T=4.2 K and was illuminated through a microscope objective (NA=0.4) by a train of laser pulses with a pulse duration of ~2 ps and 76 MHz repetition frequency. In the case of resonant excitation conditions the pulse train was generated by an optical parametric oscillator synchronously pumped by a mode-locked Ti:Sapphire laser. This system provides a tuneability of the photon energy in the range of 0.82-1.24 eV (1.51-1.00 μm). In highly non-resonant excitation conditions only the Ti:Sapphire laser operating at 1.49 eV (0.83 μm) photon energy was used. Photons emitted from the QDash structure were collected by a microscope objective and directed to a spectral analyzer consisting of a 0.3 m-focal length monochromator and an InGaAs-based multichannel detector or a state-of-the-art nitrogen-cooled streak camera system from Hamamatsu operating in a photon counting mode. The streak camera system covers a spectral range of 1.0-1.7 μm and provides a temporal resolution on the level of ~20 ps. The linear polarization of excitation was controlled by a calcite polarizer with the extinction ratio of 10^5:1 and a multi-order half-waveplate. Polarization of emission was analyzed in front of the monochromator where the incident light passes through a multi-order half-waveplate first, and then through a calcite polarizer set in a fixed position. It let to eliminate possible impact of the internal elements of monochromator on the light analysis process. A low temperature PL spectrum obtained from the ensemble of studied InAs/In$_{0.53}$Ga$_{0.23}$Al$_{0.24}$As/InP(001) QDashes is shown in Fig.1(a). It was collected under the non-resonant excitation, where e-h pairs are photogenerated mainly in the In$_{0.53}$Ga$_{0.23}$Al$_{0.24}$As barrier and in the InP capping layer and subsequently populate the ensemble of QDash states after the energy dissipation and total angular momentum relaxation. Since the excitation was rather weak (~1 e-h pair/QDash), the observed PL spectrum is produced mainly by the recombination of confined neutral excitons, possibly partially affected by the presence of negatively charged excitons (trions) and biexcitons. The observed ~35 meV broadening of the PL band reflects the ensemble non-uniformity caused by fluctuations in, e.g., QDash size, strain and chemical composition. Most of QDashes are preferentially aligned and elongated along the [1$\bar{1}$0] crystallographic direction, with the in-plane aspect ratio exceeding 2.5. Despite of other effects, such a geometrical property of a QDash, especially the lack of in-plane rotational symmetry, can directly suggest existence of polarization anisotropy in the light emission process from the ground state (GS)[14], and hence it must be addressed before the analysis of any spin properties of the system. In order to examine the optical anisotropy, the structure was excited in the barrier by linearly polarized pulses with two directions of polarization axis: V ([1$\bar{1}$0])- along the QDash , and H ([110]) - in a perpendicular direction. Two linearly polarized components of emission, labeled as I_V and I_H, were measured with respect to these directions. Fig.1(b) collects the results, which clearly confirm a strong polarization anisotropy of the emission process as the two cross-polarized PL spectra exhibit significant difference in their peak intensities. For a quantitative discussion one can introduce the degree of linear polarization (DOP), defined by

$$DOP_{V(H)} = \frac{I_{V(H)} - I_{H(V)}}{I_{V(H)} + I_{H(V)}}. \qquad (1)$$

In the above-mentioned case, the $\left|DOP_{V(H)}\right|$ reaches 25±5% that defines the so-called intrinsic DOP. It is important to note that the intrinsic DOP is not affected by the process of building-up a certain exciton spin population at the GS of the QDash ensemble. The non-resonant excitation results in energy dissipation accompanied by efficient spin relaxation that in turn erases the memory of an initial exciton spin state acquired from the polarization of excitation. In these conditions, a substantial DOP appears due to intrinsic properties of the e-h confinement that pins the polarization state of emission. This can be explained in terms of a QDash shape, anisotropic confinement for carriers, a non-uniform strain field and piezoelectricity induced by it, atomistic disorder at interfaces, and finally the local asymmetry of the InAs zinc-blend crystal lattice.[10,14–18] From a theoretical

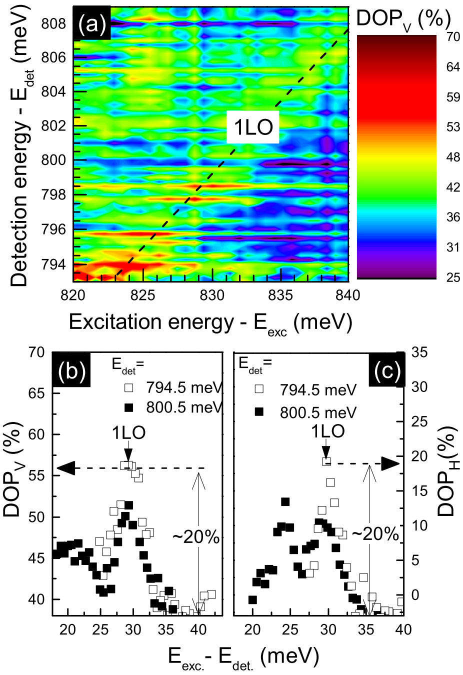

FIG. 2. (a) A 2D intensity map of the DOP_V measured in the exciton-LO phonon excitation scheme in T=4.2 K. Dashed line indicates position of the LO resonance across the QDash emission. (b), (c) vertical profiles of the 2D map at a given detection energy indicating enhancement of the DOP at the LO phonon resonance for the $|V\rangle$ and $|H\rangle$ spin state pumping, respectively. (Color online)

point of view these asymmetries with respect to V and H axes lead to a light-hole ($|\downarrow/\uparrow\rangle$) admixture to a nominally purely heavy-hole ($|\Uparrow/\Downarrow\rangle$) state, producing new hole eigenstates[19], i.e. $|\Uparrow'/\Downarrow'\rangle \propto |\Uparrow/\Downarrow\rangle \pm i\varepsilon\,|\downarrow/\uparrow\rangle$. It converts excitons' polarizations from circular to elliptical with major axes tilted towards the V axis in the case of both states. The same holds for trions preventing them to be efficiently addressed by the linearly polarized light due to the lack of an e-h exchange interaction. In case of neutral excitons, the presence of an e-h exchange interaction lifts the degeneracy between excitonic states. For an in-plane anisotropy introduced between V and H axes it produces two bright, $|H/V\rangle \propto |\downarrow\Uparrow'\rangle \pm i\,|\uparrow\Downarrow'\rangle$, and two dark eigenstates, $|D1/2\rangle \propto |\uparrow\Uparrow'\rangle \pm |\downarrow\Downarrow'\rangle$. Calculation of interband dipole moments[20], $\boldsymbol{d}_{\text{inter}}$, for the bright states indicate that they should couple to the light field linearly polarized along H and V axes, respectively, with unequal oscillator strengths ($f_V/f_H \propto \varepsilon$ for small light-hole admixtures). It is considered to be a reason for a substantial value of the intrinsic DOP at the QDash GS. Since the intrinsic DOP is known, one can measure an excess DOP that may result from a certain optical spin pumping scheme. In the following case, the excitation is energetically tuned to the wetting layer (WL) of the QDash structure in order to decrease the role of spin scattering events leading to a collapse of the desired spin state. Excitons are expected to be created in one of the earlier defined states ($|H\rangle$ or $|V\rangle$) by setting up the certain polarization of excitation. Partially, trion states are also addressed. However, as trion emission is polarized independently of the excitation, it should not contribute to the excess DOP, apart from acting as a background which weakens the observed effect. The emission was analyzed in a similar way as in the highly non-resonant excitation case, however, the PL spectrum was filtered out around 0.80 eV which allows for elimination of the unwanted scattered laser light. The $DOP_{V(H)}$ obtained according to Eq.1 is plotted with open points in Fig.1(c). One may notice that under such an optical pumping scheme the DOP increased up to ~40% and ~0% in the case of DOP_V and DOP_H, respectively. This leads us to a conclusion that the injected spin state is partially preserved during carriers' relaxation to the QDash GS, so the memory of the excitation process is present to some extent. Further, it was verified an even more reliable spin pumping procedure in which the excitation is quasi-resonant with the QDash GS, namely the pumping via a longitudinal optical (LO) phonon was used to assure minimal excess energy and nearly immediate injection of a spin state to the QDash GS. Fig. 2(a) presents a 2D map of the DOP_V obtained by scanning the laser photon energy ($\mathrm{E_{exc}}$) towards the QDash emission band defined by the spectral filter. One can notice a well resolved intensity feature across the detection energy ($\mathrm{E_{det}}$) that shifts parallel to the $\mathrm{E_{exc}}$ while keeping the constant distance of ~30 meV between excitation and the characteristic energy of the feature. The energetic distance is close to the LO-phonon energy in InAs. The upper-left corner of the 2D map reveal also another intensity feature that may be related to a transverse-optical-phonon excitation in InAs, however, we skip it in a further discussion. For the concerning experiment the sample was patterned by a 5×5 μm mesa structure. The discrete character of the map along its vertical axis corresponds to the fact that with the small ensemble size within the mesa and under quasi-resonant excitation single emission lines from individual QDashes can be seen. Figures 2(b), and (c) present examples of horizontal profiles, registered for the case of V and H pumping, respectively, which were cut-out from the 2D map at two different $\mathrm{E_{det}}$. As may be easily noticed, at the 1LO-phonon feature the DOP is strongly enhanced by ~20% with respect to the DOP measured for the WL excitation and ~35% as compared to the intrinsic DOP. It leads us to an initial conclusion that the creation of exciton accompanied by emission of a LO phonon can significantly preserve coherence within the injected spin state as it is realized likely by a coherent inelastic Stokes Raman scattering process. Although the spin memory effect is clearly present, the DOP value is expected to be much higher, up to the limit of 100%[21]. The lack of full polarization of emission in the V-V configuration could be partially caused by (i) random alignment of a QDash along the V axis in an ensemble of Dashes, (ii) exciton localization in a

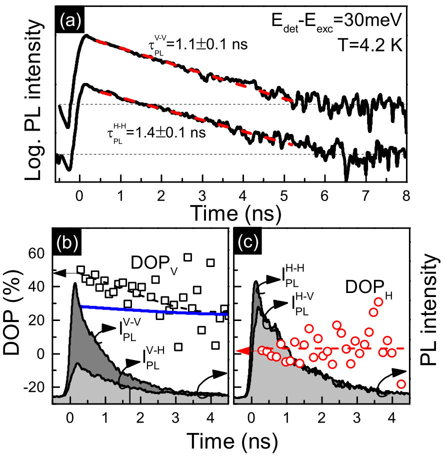

FIG. 3. (a) Low temperature TRPL traces measured within the exciton-1LO phonon excitation scheme for two configurations of the linear polarization of excitation and detection: V-V (upper trace) and H-H (lower-trace). Dashed red line shows the fit to a decay function. (b), and (c) TRPL traces taken for various excitation-detection schemes: V-V, V-H, H-H, H-V and resultant temporal evolution of the DOP: DOP_V (black open squares in (b)), DOP_H (red open circles in (c)). The blue solid line in (b) shows a temporal evolution of the intrinsic DOP. E$_{\mathrm{det}}$=799.4 meV, T=4.K, P$_{\mathrm{exc}}$=30 μW. (Color online)

QDash[18], (iii) elliptically polarized emission from trion states[8] independent of polarization of excitation, acting as a background for excitonic emission. A more unexpected issue, which needs to be addressed here is the asymmetry between V-V and H-H configurations manifested in a significantly lower values of DOP_H. This phenomenon, not present in the literature, has not been fully understood yet, however, we propose an initial explanation. Based on the preliminary discussion of excitonic states, polarization injection might be expected to be equally effective for H and V cases as interband dipole moments of the two bright states are collinear with respective axes. The interband contribution to the dipole moment is commonly regarded as dominant[22], however, the usually neglected intraband term may become substantial for structures of a large volume. The macroscopic character of this contribution ($\boldsymbol{d}_{\mathrm{intra}} \propto \langle\psi_{\mathrm{e}}|\,\boldsymbol{r}\,|\psi_{\mathrm{h}}\rangle$, where $|\psi_{\mathrm{e/h}}\rangle$ are the e/h envelope functions) in combination with a significant elongation of QDashes promote the V component of $\boldsymbol{d}_{\mathrm{intra}}$ approximately to the same extent for both bright states. Such contribution strongly affects polarization properties of the $|H\rangle$ state as it is perpendicular to its $\boldsymbol{d}_{\mathrm{inter}}$, which is not the case for the $|V\rangle$ state. For $|\boldsymbol{d}_{\mathrm{intra}}|/|\boldsymbol{d}_{\mathrm{inter}}| \sim 1/2$ we were able to approximately reproduce DOP values both cases of excitation: the quasi-resonant one ($DOP_V \approx 48\%$, $DOP_H \approx 12\%$) and non-resonant ($DOP_{V/H} \approx \pm 21\%$).

Let us shift the discussion towards dynamical properties of the spin excitation in QDashes. In Figure 3(a) we present TRPL traces registered at E$_{\mathrm{det}}$=799.4 meV under the exciton-1LO phonon spin pumping scheme. As predicted from the light-hole admixture considerations, the obtained PL lifetimes ($\tau \propto f^{-1}$) for both states slightly differ: τ^{V-V}=1.1±0.1 ns *vs.* τ^{H-H}=1.4±0.1 ns. The average exciton lifetime is surprisingly low as compared to a theoretically predicted one that is expected to be approximately 2.0 ns[23] for the strong confinement limit. In this case one has to take into account a dense structure of hole states induced by the QDash size, as indicated by calculations of the QDash band structure[18,19,23], that influences the exciton radiative lifetime. In order to study a temporal evolution of the DOP, additional two TRPL traces were measured in the following excitation-detection configurations: V-H, and H-V. Subsequently, the DOP was calculated according to Eq.1 and the results are presented in Fig. 3(b) (open black squares) - for the DOP_V, and Fig. 3(c) - for the DOP_H. The asymmetry between DOP_H and DOP_V discussed for time-integrated spectra is naturally present also here. Unfortunately, the amplitude of the DOP_H is so small that it is registered with a rather large uncertainty in the present experiment. The DOP_V decays from ∼50% to ∼27% within a time interval of ∼4.5 ns from the excitation, which gives ∼1.7 ns of the decay time constant. This time is not related to the spin polarization lifetime since it results from the difference in oscillator strengths along H and V directions. However, the observation of purely monoexponential decays (up to 6 ns) allows us to estimate, basing on a rate equation model, that any spin relaxation process present in the system has to be slower than 15 ns.

In conclusion, we investigated the impact of various schemes of the optical spin pumping in the self-assembled InAs/In$_{0.53}$Ga$_{0.23}$Al$_{0.24}$As/InP(001) quantum dash structure emitting at 1.55 μm on spin memory at the ground state. The highly non-resonant spin pumping did not lead to a preservation of the spin memory of an excitation, however, the registered polarization degree of ∼25% pointed to an important intrinsic property of a QDash confinement caused by the valence band mixing and anisotropic exchange interaction. In the case of spin injection into the WL of QDashes, the DOP increased considerably by more than 15% with respect to the intrinsic DOP, which means the presence of spin memory effect. However, the best results were achieved in the case of exciton-1LO pumping scheme, for which the resultant DOP reached ∼50%.

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