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Multiexciton complex from extrinsic centers in AlGaAs epilayers on Ge and Si substrates

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The multiexciton properties of extrinsic centers from AlGaAs layers on Ge and Si substrates are addressed. The two photon cascade is found both in steady state and in time resolved experiments. Polarization analysis of the photoluminescence provides clearcut attribution to neutral biexciton complexes. Our findings demonstrate the prospect of exploiting extrinsic centers for generating entangled photon pairs on a Si based device. © *2013 AIP Publishing LLC*.
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Quantum states of light are building blocks for most quantum-communication approaches,[1] including quantum teleportation.[2] Solid state source of quantum states of light can be realized by quantum dots (QDs) that are semiconductor nanocrystals being alike artificial atoms.[3] The excitonic complexes in QDs show antibunched luminescence leading to high fidelity single photon emitters.[4,5] At the same time, the two photon emission of a neutral biexciton can be exploited to generate polarization entangled photon pairs.[4,5] Recently, the monolithic integration on Si of a quantum emitter based on GaAs quantum dots has been achieved, opening the possibility to integrate "classical" electronics and "quantum" photonics on a single platform.[6,7]

The QD route to solid state quantum emitters requires the control of the 3D nucleation in the epitaxial growth of semiconductors. A different approach has recently emerged by using photons antibunched from individual extrinsic centers in different semiconductor materials.[8,9] In particular, single-photon emission from neutral excitons has been demonstrated from a single nitrogen-vacancy center in diamond,[8,9] nitrogen bound-excitons in ZnSe,[10] Te pairs in ZnSe,[11] F impurity in ZnMgSe/ZnSe quantum-well nanostructures,[12] single N centers in GaAs,[13] and extrinsic centers in AlGaAs.[14] An important difference with QDs is that single impurity centers are usually not able to confine two excitons: as an example we are not aware of biexcitons in nitrogen-vacancy centers in diamond. Only recently it has been proved that dyads of isoelectronics centers can be exploited to obtain biexciton emission from extrinsic centers in ZnSe, GaAs, and AlAs,[15,16] opening the route to exploit extrinsic centers also for generating entangled photon pairs.

Here, we address the biexciton properties of the unintentional contamination with defects of $Al_{0,3}Ga_{0,7}As$ layers grown on Si and Ge substrates. The extrinsic bands of the alloy $Al_{0,3}Ga_{0,7}As$ break into bright and sharp emission lines in the visible spectral region around 670 nm. The excitation power dependence of the photoluminescence (PL) shows the two photon radiative cascade both in steady state and time resolved (TR) measurements. By using polarization resolved measurements, a clear attribution of the observed photon cascade to neutral biexcitons is done.

We studied the emission properties of two different hybrid samples grown on a Ge and Si substrate, respectively, by using (001) substrate miscut 6° towards the [110] direction to allow growth of GaAs layers free of antiphase domains. In the case of Si substrate, a 2.9 μm of a fully relaxed Ge layer was first grown by low energy plasma enhanced chemical deposition. For the hybridization with AlGaAs layers, we used a conventional solid-source molecular beam epitaxy (MBE) system. The GaAs/AlGaAs nominally undoped epilayer is made by an initial GaAs buffer (580 °C) layer, 650 nm thick. An $Al_{0,3}Ga_{0,7}As$ layer, 200 nm thick is then deposited at the same temperature and As pressure of the GaAs buffer, finally 10 nm thick GaAs caps the structure. More details on the growth can be found in Ref. 14.

PL measurements were accomplished at low temperature ($T = 10$ K) with the sample placed in a cold-finger helium flow cryostat. A frequency-doubled Nd-yttrium aluminium garnet laser with $\lambda_{exc} = 532$ nm was used for continuous wave measurements, while the second harmonic $\lambda_{exc} = 400$ nm of a fs Ti-sapphire laser was used for TR experiments. Micro-PL spectra were performed by a confocal setup (main objective 100×, NA = 0.7) with a lateral resolution of about 0.7 μm. The emission was dispersed either by a 30 cm single monochromator or by a 100 cm focal length double monochromator (depending on the required spectral resolution) and detected by a silicon charge coupled device camera for cw experiments and by an avalanche Si photodiode for TR measurements. In the following, we will

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concentrate only on the data on the sample grown on the Ge substrate; similar results are found in the sample with Si substrate, even if the density of extrinsic centers is larger with respect to the sample on Ge substrate.

Typical micro-PL map, at 50 μW excitation power, of the sample grown on the Ge substrate is reported in the bottom panel of Fig. 1 and compared with the semilogarithmic representation of the macro-PL spectrum obtained with a 100 μm laser spot (yellow line in the top panel). The four main contributions of the extrinsic AlGaAs recombination (bound exciton (BX) band at 1.93 eV, donor-acceptor (DA) bands at 1.895 eV and at 1.88 eV, deep centers (DC) tail extending from 1.87 to 1.7 eV) break into spatially localized sharp lines when using micro-PL. In particular, in the range 1.80–1.87 eV, we found very well isolated bright emissions which show, at this excitation power, a multiplet nature. The map shows that each multiplet arises, within our instrumental resolution, from the same spatial point.

The detail of the power dependence of one of these extrinsic multiplets is reported in Figure 2(a). At low power, a single line (hereafter denominated with X) is observed; the minimum full width at half maximum linewidth δE is of the order of 30 μeV, which is comparable to our instrumental resolution. Increasing the excitation power P_{exc}, a second line (hereafter denominated with XX) comes out and it becomes the dominant contribution at large P_{exc}. Both X and XX lines quench at the highest P_{exc}. Summary of the power dependence of X and XX for three different centers are shown in Figs. 2(b)–2(d); the general trend is qualitatively very similar to the exciton and biexciton recombination from QDs.[7,17] For a quantitative analysis, the data are fitted with Poissonian distributions $P(m) = \langle n \rangle^m \exp\{-\langle n \rangle\}/m!$ with $m = 1$ for the X line, $m = 2$ for the XX line, and $\langle n \rangle = \alpha P_{exc}{}^{\beta}$ in both cases.[7,17] The experimental data in Fig. 2 agree very well with the Poissonian distribution and the values of the parameters α and β (given in the figures, with P_{exc} in mW), which may slightly change from center to center, are consistently equal for the X and XX lines of the same center. These findings suggest the attribution of the X and XX lines to neutral exciton and biexciton from the same center, respectively.

Time resolved measurements of the X and XX lines of a single center can be used to prove the exciton-biexciton

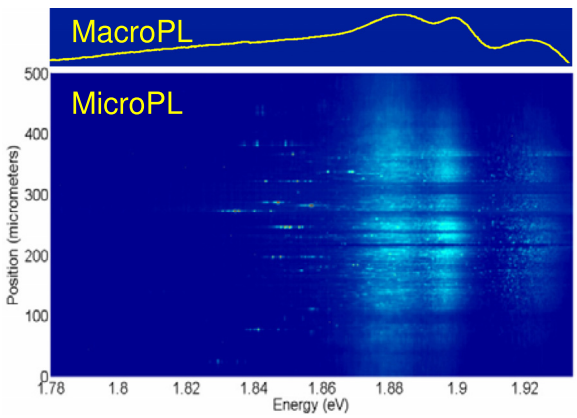

FIG. 1. Micro-PL map of the AlGaAs emission. In the bottom of the figure, the macro-PL spectrum is reported for comparison in a semilogarithmic scale.

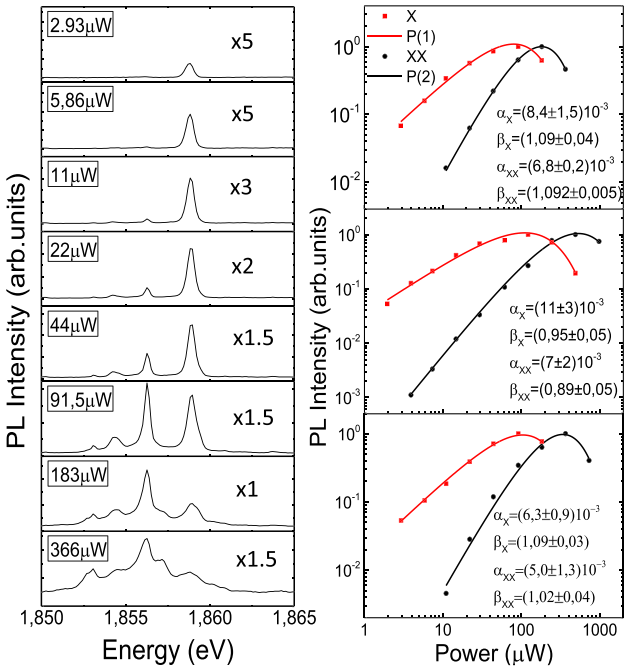

FIG. 2. (a) Power dependence analysis of the PL spectra where two main PL lines of the multiplet band are labelled X and XX. (b)–(d) PL intensity of the X (orange dots) and XX (purple dots) recombination intensity as a function of the excitation power for three different centers. The data are compared with the Poissonian fits and the fitting parameters are reported (P in mW).

cascade: in the presence of two electron-hole pairs inside the same confining potential, the biexciton emission (recombination of the first pair) always occurs before the exciton emission (recombination of the second pair).[18] Data for two different centers are reported in Figure 3, comparing the X

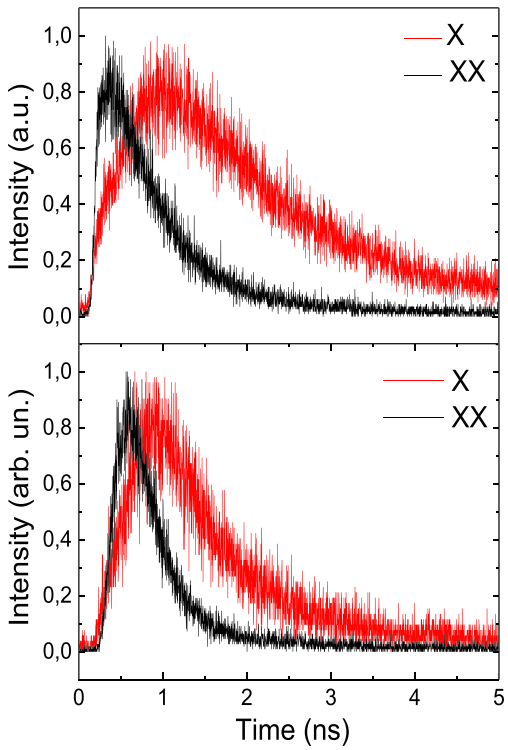

FIG. 3. Time resolved PL spectra at the emission energy of the X (purple line) and the XX (orange line) emission for two different centers.

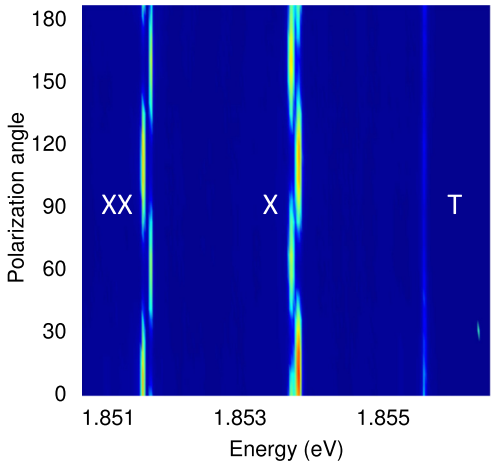

FIG. 4. PL maps of the dependence of the PL spectrum on the polarization of the emission.

and XX time dependence. The lifetime of the X emission is in the range of 700 ps and 1.4 ns for the two centers and in both cases the lifetime of the XX emission is roughly half of the X lifetime, as expected for a biexciton, in case of strong confinement.[7] Even more striking is the fact that, at all the excitation powers, the X line intensity reaches the maximum at a delay time of the order of the decay time of the XX line, demonstrating the emission cascade in the recombination.

The final and definite proof of the neutral biexciton-exciton cascade can be obtained by polarization resolved PL measurements. It is well known that any asymmetry in the shape or in the carrier confining potential determines a fine structure splitting (FSS) of the excitonic states. The finger print of a zero spin value of the biexciton state is then the symmetric splitting of the X and XX lines, because the sum of their emission energies is polarization independent.[19,20] On the contrary, the charged exciton state does not split, i.e., the FSS is zero.[19,20] The map of a typical multiplet PL spectrum as a function of the polarization angle of the PL emission is reported in Figure 4. We clearly see that the X and XX lines have an opposite shift when rotating the polarization angle, so demonstrating the attribution to neutral exciton and biexciton. In this peculiar center, we were also able to detect a third line in the high energy side of the X line which does not shift when rotating the polarization angle, allowing us to attribute it to a charged exciton complex.

The attribution of the nature of the extrinsic centers in AlGaAs involved in this multiexciton recombination is not clear, and two scenarios seem to be more likely. On one side, since biexciton is not expected from single impurities, there is the possibility of dyads of impurities[15,16] related to the Ge contamination of the AlGaAs alloy. On the other side, it is also possible, following the recent findings of Ref. 21, to have nanoclusters of Al poor material in Al rich barriers due to Al segregation in the AlGaAs alloy. In this latter case, the extrinsic centers will be a sort of natural QD in the AlGaAs barrier.

In conclusion, we have addressed the possibility of using peculiar extrinsic centers in AlGaAs monolithically integrated on Si and Ge substrates to achieve neutral biexciton states in the visible spectral range around 670 nm. Clear signatures of the biexciton cascade are observed in power dependence and time resolved measurements. Eventually, the neutral nature of the excitonic complexes is demonstrated by polarization resolved measurements. Our results highlight the possibility of obtaining entangled photon pairs emission on a Si based device by exploiting unintentional extrinsic centers. We expect also that the spectral range of the biexciton emission could be largely tuned by changing the Al content in the AlGaAs alloy.

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