PHYSICAL REVIEW B **81**, 241309(R) (2010)

Exciton fine-structure splitting in GaN/AlN quantum dots

C. Kindel,[1,2,*] S. Kako,[3] T. Kawano,[1] H. Oishi,[1] Y. Arakawa,[1,3,4] G. Hönig,[2,†] M. Winkelnkemper,[2] A. Schliwa,[2] A. Hoffmann,[2] and D. Bimberg[2]

[1]*Research Center for Advanced Science and Technology, University of Tokyo, 4-6-1 Komaba, Meguro, Tokyo 153-8904, Japan*

[2]*Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstraße 36, D-10623 Berlin, Germany*

[3]*Institute for Nano Quantum Information Electronics, University of Tokyo, 4-6-1 Komaba, Meguro, Tokyo 153-8505, Japan*

[4]*Institute of Industrial Science, University of Tokyo, 4-6-1 Komaba, Meguro, Tokyo 153-8505, Japan*

(Received 19 April 2010; published 24 June 2010)

Exciton bright-state fine-structure splitting (FSS) in single GaN/AlN quantum dots (QDs) is reported, presenting an important step toward the realization of room temperature single-qubit emitters for quantum cryptography and communication. The FSS in nitride QDs is up to 7 meV and thus much larger than for other QD systems. We find also a surprising dependence of FSS on the QD size, inverse to that of arsenide QDs. Now we are able to explain why FSS can only be observed in small QDs of high-emission energies. Our calculations reveal a shape/strain anisotropy as origin of the large FSS allowing different approaches to control FSS in nitrides.

DOI: 10.1103/PhysRevB.81.241309 PACS number(s): 73.21.La, 71.35.−y, 78.20.Bh, 78.67.Hc

Zero-dimensional semiconductor nanostructures, such as self-assembled quantum dots (QDs), are unique to study and exploit features of single, discrete quantum systems for real-world applications, such as devices generating single-polarized photons (qubits) or entangled photon pairs on demand.[1–5] Such devices might present the physical basis for future practical quantum cryptography and communication systems.[6–8] Until now electrically driven qubit emitters have been demonstrated at cryogenic temperatures only based on Group III-As QDs.[1,3,9] Ubiquitous room-temperature devices for quantum-information systems on demand must be based on QDs with a much deeper confinement potential such as GaN/AlN QDs. No exciton bright-state fine-structure splitting (FSS) for such QDs has been reported yet and detailed theoretical understanding of exchange interaction in wurtzites is missing. We present in this Rapid Communication the first comprehensive experimental and theoretical study of FSS in GaN/AlN QDs. Values up to 7 meV are reported, much larger than in other semiconductor QDs.[10–13] Surprisingly, the dependence of FSS on emission energy reported here is inverse to that of arsenide-based QDs. These observations are well explained by our theoretical modeling based on realistic $\mathbf{k}\cdot\mathbf{p}$ wave functions. We are able to identify the anisotropic strain distribution in the QD as origin of the FSS. Our discoveries open new ways to effectively control the FSS, toward the realization of qubit emitters on demand operating at room temperature.

The self-assembled hexagonal GaN/AlN QDs investigated in this work were grown by low-pressure metal-organic vapor deposition on 100 nm AlN on *n*-type 6H-SiC [0001] substrate and capped with another 100 nm AlN layer. The samples were processed to mesa structures to facilitate single-dot microphotoluminescence measurements.[2] A frequency-doubled solid-state laser with $\lambda=266$ nm was used to excite the QDs with varying excitation power at 4 K. Polarization-dependent optical spectra were taken using a $\lambda/2$ waveplate and a Glan-Taylor polarizer. For an accurate assignment of linear-polarization angle φ and degree of polarization P we took 36 spectra per QD while rotating the waveplate by $\Delta\alpha=5°$ per spectrum. The integrated peak intensities I were then fitted by $I(\alpha)=I_0/2[1+P\cos(4\alpha-2\varphi)]$. φ has an accuracy of $\approx 5°$. The error of P is estimated as 20% due to the difficulty of correct background subtraction. The spectral resolution of our setup is ≈ 1 meV.

Large GaN/AlN QDs emitting around 3.4 eV exhibit strong, yet incomplete linear polarization of about 80%, [Fig. 1(a)].[14,15] Biexcitonic emission is observed with negative (i.e., antibinding) biexciton binding energy in agreement with previous studies.[2,11,16,17] The polarization has been interpreted to stem from valence-band mixing effects induced by shape/strain anisotropy of the QD.[14,15,18,19] Presence of

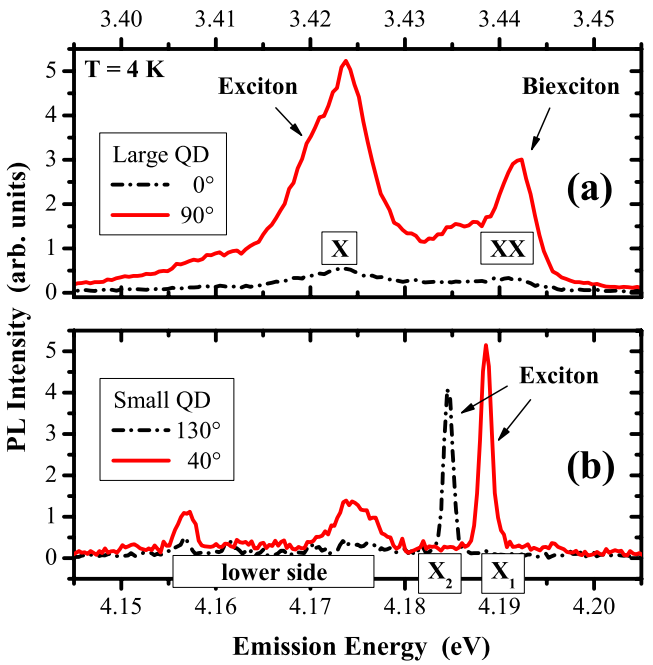

FIG. 1. (Color online) Linear polarization of luminescence from a large QD (a) showing identical polarization of exciton and biexciton, Ref. 14, and a small QD (b) with clear orthogonal polarization of excitonic peaks X_1 and X_2 ($\Delta E_{X_{1,2}}=3.9$ meV, $P_{X_1}=0.95$, $P_{X_2}=0.96$, and $\Delta\varphi_{X_{1,2}}=98°$).

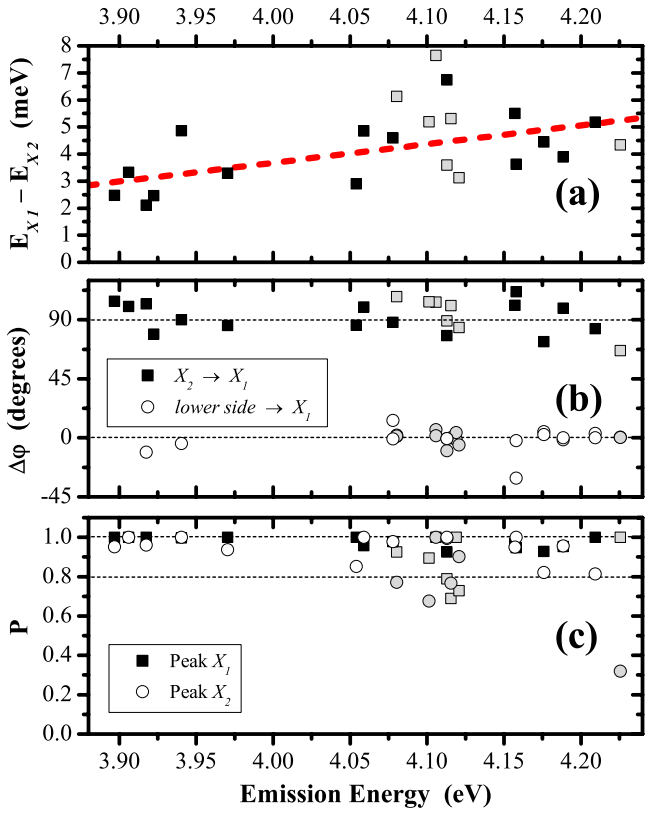

FIG. 2. (Color online) Overview of luminescence properties from different QDs. (a) Energy splitting between peaks X_1 and X_2; linear regression (dashed line) shows clear tendency to larger splitting for higher emission energies. (b) Difference of linear-polarization angles between peaks X_2 and X_1 (cubes) and *lower side* peaks and X_1 (circles). (c) Degree of polarization of peaks X_1 and X_2. Data points from QDs with $P<0.8$ for peak X_1 or X_2 are shaded in gray.

QD shape anisotropy in our samples was confirmed by atomic force microscopy.[14] In this Rapid Communication, we concentrate on much smaller QDs with emission energies above ≈3.9 eV. This energy range is particularly interesting, because the QDs show significantly smaller line widths, thereby enabling us to resolve more details of the emission. The dependency of line width on emission energy is attributed to the stronger impact of the quantum-confined stark effect in large QDs. A detailed discussion is to be published soon elsewhere. We find a surprising qualitative difference in the polarization of emission: in the high energy range, pairs of lines with orthogonal linear polarization are observed [Fig. 1(b)]. The energy splitting of these pairs is in the range of 2–7 meV [Fig. 2(a)]. The difference between the linear polarization angles is around 90° [Fig. 2(b)]. The distribution of polarization angles is seemingly random. A correlation with the crystal axes could not be observed. The degree of polarization is close to one [Fig. 2(c)]. Excitation power-dependent measurements show almost linear dependences, indicating excitonic origin of the doublets. Generally, we do not observe emission on the higher-energy side of the doublet. However, we regularly see a pattern of strong peaks on the lower-energy side [Fig. 2(b)]. The excitation-power dependence of these peaks is super linear with exponents between 1.3 and 2. Furthermore, the linear-polarization angle of these peaks seems to always follow the angle of the energetically higher peak of the orthogonally polarized line pair. The superlinear excitation power dependence indicates biexcitonic emission in agreement with the observation of Simeonov *et al.*[16] of positive biexciton binding energies for QDs emitting at energies ≳4 eV. The assignment is, however, still subject to further research.

To investigate the origin of the different spectra, we have calculated the fine structure and absorption spectra of excitons confined in GaN/AlN QDs. Our model is based on single-particle electron and hole wave functions calculated by means of eight-band $\mathbf{k}\cdot\mathbf{p}$ theory. The $\mathbf{k}\cdot\mathbf{p}$ model is implemented for arbitrarily shaped QDs and includes the effects of strain, piezoelectricity and pyroelectricity, and spin-orbit and crystal-field splitting.[20] To calculate the excitonic (two-particle) states, first the Hartree-Fock equations for the four Hartree-Fock states $|\downarrow\downarrow\rangle$, $|\downarrow\uparrow\rangle$, $|\uparrow\downarrow\rangle$, and $|\uparrow\uparrow\rangle$ of both excitons (A and B excitons) have been solved self-consistently. Excitonic bright states and their splitting are not sufficiently described by these Hartree-Fock states. Their linear combinations (e.g., $c_1|\downarrow\downarrow\rangle \pm c_2|\uparrow\uparrow\rangle$, $c_i \in \mathbb{C}$) are additionally needed. Thus, the eight Hartree-Fock states are subsequently used as basis for the matrix expansion of the exchange operator and the true excitonic states and energies are obtained by diagonalizing this matrix. This way the advantages of a self-consistent Hartree-Fock calculation are combined with an accurate description of the excitonic bright-state splitting.

In agreement with experimental observations[2,21,22] and previous theoretical considerations[23–27] the QDs have been modeled as truncated hexagonal pyramids[19] of pure GaN with a height h from 0.8 to 2.0 nm and vertical aspect ratio ar_{vert} (height h to diameter d) between 0.14 and 0.22. The QDs are placed on a 0.2-nm-thick GaN wetting layer (included in the total height h) and are embedded in a matrix of pure AlN. For the calculation of excitonic FSS, a lateral anisotropy of the QDs is assumed, induced by either elongated QD structures or compressive or tensile strain, applied uniaxially with relaxation in the other directions. The absorption spectrum of the A- and B-band exciton states in a QD with $h=1.2$ nm, $d=7.5$ nm, and an in-plane structural elongation of 20% along the y ($\equiv[11\bar{2}0]$) axis is shown in Fig. 3(a). The bright states of the A exciton (formed by an electron in the ground state and a hole in the ground state derived from the A valence band) and the B exciton (hole in the first excited state derived from the B valence band) show a FSS of 3.1 meV and 3.2 meV, respectively. All transitions are fully linearly polarized either parallel or perpendicular to the long axis of the QD. The energetically lower (higher) transition of the A exciton is polarized perpendicular (parallel) to the elongation, vice versa for the B exciton. The differences of the oscillator strengths are owed to the character of the single-particle hole states in anisotropic nitride QDs:[18,19] for A (B) excitons transitions polarized parallel (perpendicular) to the elongation are favored.

Figures 3(b) and 3(c) show the FSS of the A exciton as function of in-plane elongation [Fig. 3(b)] and uniaxial stress [Fig. 3(c)], respectively, with constant h and ar_{vert}. Negative (positive) elongation corresponds to elongation along the y

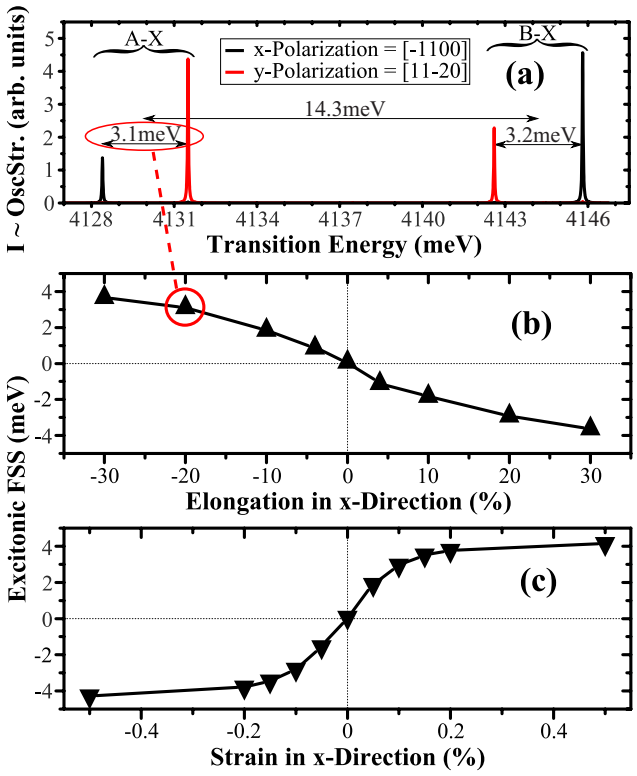

FIG. 3. (Color online) Calculated FSS of QDs with a height of $h=1.2$ nm and diameter of $d=7.5$ nm. (a) Absorption spectrum of the A band (A-X) and B band (B-X) excitonic states in a QD with an in-plane elongation of 20% along the y axis. [(b) and (c)] Fine-structure splitting of the A exciton as a function of (b) in-plane elongation or (c) uniaxial stress.

(x) direction; negative (positive) strain represents compressive (tensile) uniaxial strain in x direction. For small elongations or strains the FSS increases rapidly, tending to a maximum for largely elongated (strained) structures. This limit is affected by structural properties of the QDs (for instance, their volume) and equals twice the excitonic dark-bright splitting. No dependence of FSS on the direction of elongation is found. Note that only 10% of the lattice mismatch between GaN and AlN (=2.6%) is enough to maximize the FSS. This is easily compensable by anisotropic strain relaxation, e.g., through sample processing, or externally applied stress. The FSS of the B exciton (not shown here) differs only slightly from the FSS of the A exciton for all QDs.

One might argue that the orthogonally polarized line pairs originate from A and B excitons[18,19] or trions.[15] We believe this interpretation is unlikely: an exciton bright-state FSS doublet should always be 100% polarized, with orthogonal linear polarization. If the FSS doublet cannot be resolved spectroscopically and only the sum of the two lines combined is detected, the effective degree of polarization is much lower (e.g., it is zero, if both lines have equal intensity.) This effective degree of polarization for the A/B exciton lines is strongly dependent on the asymmetry of the QD. The largest degree of polarization is reached for the largest QD elongations and largest A/B splittings. In our calculations, the largest degree of polarization of 80% for the A-exciton line and 50% for the B-exciton line was obtained for a strongly elongated QD with an A-B splitting of 20 meV. For rather symmetric QDs with the smallest A/B splittings of 9 meV, the degree of polarization is reduced to only 3%. Furthermore, the B hole band should not be populated at liquid-helium temperature. In conclusion, line pairs with a splitting of less than 10 meV and a degree of polarization close to 100% cannot originate from A/B exciton or trion lines. The only other possible interpretation is that these lines originate from exchange-interaction induced exciton bright-state FSS. Some doublets with incomplete linear polarization were observed (shaded data points in Fig. 2) but the similar spectroscopic fingerprints still support the assignment as FSS.

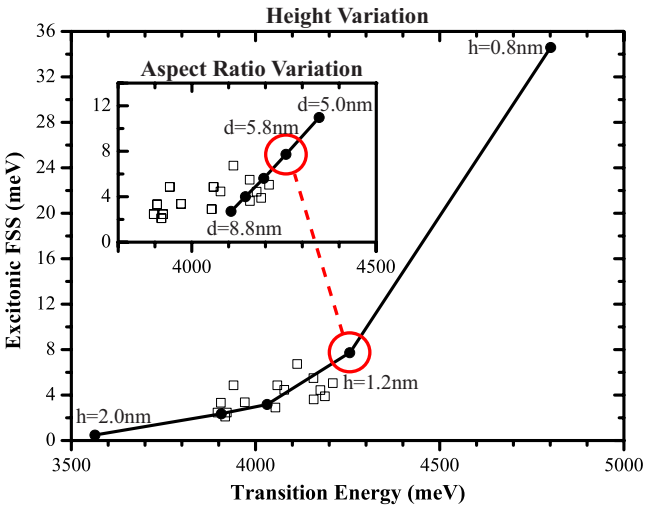

FIG. 4. (Color online) FSS of A-exciton bright states as function of QD size. Solid circles are calculated, empty squares are experimental values. The model QDs have a fixed aspect ratio $ar_{vert}=h/d=0.2$ but varying height h (0.8–2.0 nm). Inset shows FSS for QDs with constant height $h=1.2$ nm but varying aspect ratio ar_{vert} (0.14–0.22).

In contrast to the calculated oscillator strengths, the FSS doublets show comparable intensities in experiment. The calculated spectra are, however, absorption spectra. In emission the state occupancy must be taken into account. Carriers created by the excitation laser are considered to have a random-spin orientation in the QD.[15] Thus, initially, upper and lower exciton bright states are equally populated. For relaxation from upper to lower state, a spin flip is required (in contrast to B- to A-exciton relaxation). Assuming reasonably a spin-flip time to be comparable or longer than the radiative recombination time, a FSS line pair with close to equal intensity is expected. In QDs based on different materials, exciton bright-state FSS doublets with comparable intensity at liquid-helium temperature for a FSS of ≈ 1 meV were observed as well.[28]

We investigated the dependence of excitonic FSS on QD size using QD structures with identical elongation (20% in y direction) and aspect ratio ($ar_{vert}=0.20$) but varying QD height (Fig. 4). The FSS depends inversely on QD size, i.e., increases superlinearly with increasing transition energy, consistent with the absence of FSS in low-energy experimental spectra (below ≈ 3.9 eV), where the FSS is smaller than the experimental linewidth. Thus, the two exciton lines

merge to one peak with partial linear polarization. For higher transition energies the FSS increases rapidly and consequently excitonic FSS is observable. A trend to larger FSS for increasing transition energies is also seen in the experimental data [Fig. 2(a)]. Note that the trend of increasing excitonic FSS for increasing transition energy is opposite to what has been observed for InAs/GaAs QDs.[11] Also the absolute values of the FSS in GaN/AlN QDs are significantly larger. The FSS depends also on the vertical aspect ratio ar_{vert} of the QD, increasing with decreasing diameter d [Fig. 4 (inset)]. Thus, assuming at least some variation in the QD shape, the strong variation of several millielectron volt of the FSS for different QDs with the same emission energy can be well explained. For two-photon entanglement based on GaN/AlN QDs the ability to controllably reduce the FSS to zero is essential. Exploiting the strain dependence offers new approaches to obtain this goal, e.g., by creating a strain gradient through careful structuring of the sample, or by micromechanic methods.

In conclusion, we have shown that single GaN/AlN QDs exhibit a huge exciton bright-state FSS of up to 7 meV, larger than in any other QD system. Surprisingly, the FSS increases drastically with increasing emission energy, which is directly opposed to recent observations on InAs/GaAs QDs. Employing realistic simulations we have identified the origin of the huge FSS as an anisotropic strain distribution within the QD. Thus, the results of this work open the possibility to tailor the FSS in GaN/AlN QDs in order to develop high-temperature emitters of polarized single photons or entangled photon pairs on demand as needed for future quantum cryptography and communication systems.

We are indebted to Till Warming and Sven Rodt for fruitful discussions. Parts of this work have been supported by the Special Coordination Fund for Promoting Science and Technology of the Japanese Ministry of Education, Culture, Sports, Science and Technology and by Sonderforschungsbereich 787 of Deutsche Forschungsgemeinschaft. Parts of the calculations were performed on the IBM pSeries 690 supercomputer at HLRN within Project No. bep00014.

*ckindel@iis.u-tokyo.ac.jp

†gerald.hoenig@physik.tu-berlin.de

[1] A. Lochmann, E. Stock, O. Schulz, F. Hopfer, D. Bimberg, V. A. Haisler, A. I. Toropov, A. K. Bakarov, and A. K. Kalagin, Electron. Lett. **42**, 774 (2006).

[2] S. Kako, C. Santori, K. Hoshino, S. Götzinger, Y. Yamamoto, and Y. Arakawa, Nature Mater. **5**, 887 (2006).

[3] T. Miyazawa, S. Okumura, S. Hirose, K. Takemoto, M. Takatsu, T. Usuki, N. Yokoyama, and Y. Arakawa, Jpn. J. Appl. Phys. **47**, 2880 (2008).

[4] A. J. Shields, Nat. Photonics **1**, 215 (2007).

[5] D. Bimberg *et al.*, IEEE Photonics J. **1**, 58 (2009).

[6] R. Stevenson, R. Young, P. Atkinson, K. Cooper, D. Ritchie, and A. Shields, Nature (London) **439**, 179 (2006).

[7] N. Akopian, N. H. Lindner, E. Poem, Y. Berlatzky, J. Avron, D. Gershoni, B. D. Gerardot, and P. M. Petroff, Phys. Rev. Lett. **96**, 130501 (2006).

[8] J. E. Avron, G. Bisker, D. Gershoni, N. H. Lindner, E. A. Meirom, and R. J. Warburton, Phys. Rev. Lett. **100**, 120501 (2008).

[9] A. Lochmann, E. Stock, J. Töfflinger, W. Unrau, A. Toropov, A. Bakarov, V. Haisler, and D. Bimberg, Electron. Lett. **45**, 566 (2009).

[10] G. Bester, S. Nair, and A. Zunger, Phys. Rev. B **67**, 161306(R) (2003).

[11] R. Seguin, S. Rodt, A. Strittmatter, L. Reißmann, T. Bartel, A. Hoffmann, D. Bimberg, E. Hahn, and D. Gerthsen, Appl. Phys. Lett. **84**, 4023 (2004).

[12] H. Htoon, M. Furis, S. A. Crooker, S. Jeong, and V. I. Klimov, Phys. Rev. B **77**, 035328 (2008).

[13] M. J. Fernée, B. N. Littleton, and H. Rubinsztein-Dunlop, ACS Nano **3**, 3762 (2009).

[14] C. Kindel, S. Kako, T. Kawano, H. Oishi, and Y. Arakawa, Jpn. J. Appl. Phys. **48**, 04C116 (2009).

[15] R. Bardoux, T. Guillet, B. Gil, P. Lefebvre, T. Bretagnon, T. Taliercio, S. Rousset, and F. Semond, Phys. Rev. B **77**, 235315 (2008).

[16] D. Simeonov, A. Dussaigne, R. Butte, and N. Grandjean, Phys. Rev. B **77**, 075306 (2008).

[17] S. Kako, K. Hoshino, S. Iwamoto, S. Ishida, and Y. Arakawa, Appl. Phys. Lett. **85**, 64 (2004).

[18] M. Winkelnkemper, R. Seguin, S. Rodt, A. Schliwa, L. Reissmann, A. Strittmatter, A. Hoffmann, and D. Bimberg, J. Appl. Phys. **101**, 113708 (2007).

[19] M. Winkelnkemper, R. Seguin, S. Rodt, A. Hoffmann, and D. Bimberg, J. Phys.: Condens. Matter **20**, 454211 (2008).

[20] The entire method is described in Ref. 29. The material parameters from Refs. 30 and 31 have been included as described in Ref. 19.

[21] F. Widmann, J. Simon, B. Daudin, G. Feuillet, J. L. Rouviere, N. T. Pelekanos, and G. Fishman, Phys. Rev. B **58**, R15989 (1998).

[22] K. Hoshino, S. Kako, and Y. Arakawa, Appl. Phys. Lett. **85**, 1262 (2004).

[23] A. D. Andreev and E. P. O'Reilly, Phys. Rev. B **62**, 15851 (2000).

[24] V. A. Fonoberov and A. A. Balandin, J. Appl. Phys. **94**, 7178 (2003).

[25] S. Tomić and N. Vukmirović, Phys. Rev. B **79**, 245330 (2009).

[26] O. Marquardt, T. Hickel, and J. Neugebauer, J. Appl. Phys. **106**, 083707 (2009).

[27] D. P. Williams, S. Schulz, A. Andreev, and E. O'Reilly, IEEE J. Sel. Top. Quantum Electron. **15**, 1092 (2009).

[28] S. Rodt, A. Schliwa, R. Heitz, V. Turck, O. Stier, R. Sellin, M. Strassburg, U. Pohl, and D. Bimberg, Phys. Status Solidi B **234**, 354 (2002).

[29] M. Winkelnkemper, A. Schliwa, and D. Bimberg, Phys. Rev. B **74**, 155322 (2006).

[30] P. Rinke, M. Winkelnkemper, A. Qteish, D. Bimberg, J. Neugebauer, and M. Scheffler, Phys. Rev. B **77**, 075202 (2008).

[31] I. Vurgaftman and J. R. Meyer, J. Appl. Phys. **94**, 3675 (2003).