Spatial localization of quantized states responsible for sharp optical transition in AlGaN/GaN superlattice

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We present atomic level studies of the quantized electronic states in AlGaN/GaN superlattice by employing first-principles calculations. Based on the complete band profile, distribution of the discrete energy levels inside the GaN quantum well is obtained and the well-in-well confinement is observed. This second-level well introduces the spatial localization of quantized states. The calculated envelopes of the quantized state densities reveal the spatial overlap between certain electron and hole states. This, together with theoretical absorption spectra, enables us to assign the origin of the band-edge peak to the $e_0$-$h_2$ intersubband transition, which governs the optical band gap in the superlattice structure. © 2010 American Institute of Physics. [doi:10.1063/1.3410675]

III-nitrides have been the important and promising materials for the fabrication of short wavelength optical and high power electronic devices.1,2 Due to its wide band gap and strong polarizability, AlGaN system has recently attracted broad concerns in both advanced optoelectronic devices and novel nanostructured materials.3–5 In particular, AlGaN/GaN superlattice structure plays a key role in the functionalization by active layer design for various applications such as the ultraviolet light emitting devices.6 However, the special features of superlattice systems, e.g., nanoscale, heterogeneity, interface effect, and mismatch strain, lead to the difficulty and exclusivity in the study of important optical and electrical issues. Because of the nanoscale structure of AlGaN superlattice layers, in-depth investigations at atomic scale are necessary and crucial for clarifying the optical process and improving the advanced optoelectronic devices.

Despite efforts made in the past, characterization experiments are still somewhat limited by their spatial resolution and/or lack of methodology. In order to study the superlattice properties at atomic scale, the most successful tool at present would be the first-principles calculation. Based on this method, recently, progress in the clarification of the interfacial behavior of dopants in AlN/GaN superlattice was achieved and active sites responsible for the light emission in AlGaN/GaN superlattice was observed.7,8 On the other hand, with respect to the definition of a superlattice system, the feature of quantized states plays an extremely important role in interpreting the optical properties. As this point, the behavior of subband and interband transitions in the strongly polarized AlGaN/GaN quantum wells has been considered by theoretical modeling and experimental spectroscopic investigations.9–11 However, microscopic understanding of those quantized states is still scarce and the underlying mechanism of the involvement of the quantized levels in light emissions remains unclear.

In this paper, ab initio calculations were performed on the AlGaN/GaN superlattice system to study the properties of quantized states and optical spectra. In the band profile, the location and distribution of quantized levels inside the GaN quantum well is determined. According to these quantized levels, corresponding quantized states confined in the conduction band (CB) and the valence band (VB) are calculated, and the envelope and number of the spatial nodes of quantum states are revealed. Together with the absorption spectra, the optical transitions responsible for the sharp band edge peak are identified. These results clarify the mechanism of light emission and explain the difference between the real optical band edge and the fundamental band gap of nitride-based superlattice.

First-principles calculations were performed within the framework of density functional theory,12 by using the VENNA AB INITIO SIMULATION PACKAGE (Ref. 13) and also homemade codes for calculating optical properties.14 The projector augmented wave method15 was used and the planewave cutoff energy was set to 500 eV. The supercell of Al0.5Ga0.5N/GaN superlattice along the [0001] direction was constructed with barrier and well width of 2.2 nm each, comparable with the size of experimental quantum well structure in optoelectronic devices. The AlN fraction in Al0.5Ga0.5N barrier was evenly distributed for the correct and homogeneous chemical composition. Full geometric optimization was carried out and the distribution of the inhomogeneous local stress field due to the misfit strain release was determined. For the calibration of optical spectra, the dielectric function of bulk GaN was calculated with band gap correction by the scissors operation.16 The results showed that the absorption spectrum agrees well with the experimental result by spectroscopic ellipsometry.17

The bilayer [Ga(Al) and N] decomposed density of states (DOS) were calculated, and then, by joining the edges of the bilayer local DOS of CB and VB along the [0001] direction, the profile of band configuration in the Al0.5Ga0.5N/GaN superlattice was obtained, as shown in Fig.
The band profile demonstrates the inhomogeneous bending of potential well in the GaN layer due to the local polarization effect and the nonuniform stress field. In this quantum well configuration, the confinement on carriers (electrons and holes) leads to the interesting behavior of the quantized subbands. Based on the band profile, the quantized energy levels inside the GaN well are obtained by calculating the subband energies in different well layers. Energy levels of each atomic layer inside the well were calculated and meanwhile, the decomposed electron density at those levels was taken into account to determine the presence of subband levels at different sites. One can see that three discrete electron sublevels form in the CB potential well and the distribution and energy variation in them are highly position-dependent. The lowest electron subband $e_0$ above the CB minimum is located in the left hand side of the GaN well near the (0001) interface. This can be explained by the shape of the CB potential well that actually forms an energetically lowest well-in-well in the region of 2.6–3.6 nm. This local well strongly confines the $e_0$ energy level uniquely in the left hand side region. Consequently, the electrons at $e_0$ level are polarized to the [0001] direction. In contrast, the $h_0$ energy level in VB is confined to the right hand side of the well and the holes at this level are shifted to the [0001] direction. This provides the explanation of the charge separation in nitride quantum well from the viewpoint of band configuration, instead of the polarization field driving.

On the other hand, it can also be observed that the higher level the subband is, the weaker the polarization and confinement effect will be. From $h_0$ to $h_2$, the localization of quantized levels tends to shift from the right side toward the center of the quantum well, and even into the left part. Similarly, because the energy of $e_2$ level is at the topmost of the well, the confinement by the AlGaN barrier becomes weak. As a result, the $e_2$ level expands widely in the whole well and particularly, the level energy increases gradually toward the (0001) interface. This trend follows the bending curve of CB and shows a systematic shift due to the variation in the local stress field and piezoelectric polarization. What is interesting is that the $e_1$ subband appears only in the right part of the quantum well, which indicates a motion against the polarization field. To explain the formation of these electron subbands, the details of the local electron DOS are discussed below.

Figure 2 shows the local DOS of CB for the sequence of the atomic nitrogen layers (L0–L9) inside the GaN well. At the band edge, distinct peaks of quantized states can be seen and the correspondence of these states with the sublevels is assigned with respect to the level energy. The quantized states are discrete due to the quantum well confinement. The quantization of electron density in narrow energy levels can effectively give rise to the high efficiency of light emissions. In particular, in the highlight layers, L2 to L4, the $e_0$ and $e_2$ densities are strongly quantized and entirely separated, as indicated by arrows. This is because (i) the strong well-in-well confinement, as mentioned previously, gives rise to the significant quantization (nearly one-dimensional) of the particles in this region and (ii) the $e_1$ density has been kept outside this region (0.4–1.2 nm in the well). In principle, the higher the discreteness and sharpness of quantized electron states in the quantum well is, the sharper and brighter the light emission generated from this quantum system can be. This is manifested by the strong intensity of the band edge $z_1$ peak of the absorptive spectrum, as shown in Fig. 4. The $e_1$ states residing in the right part of the well are in comparatively weak density and particularly, the DOS shape of L9 close to the (0001) interface becomes steplike. These step-like states indicate a two-dimensional quantum confinement phenomenon. Similarly, the quantized states in VB also show this kind of steplike configuration.

Figure 3 shows the envelopes of the electron and hole densities of three lowest quantized energy levels in the
Al$_{0.5}$Ga$_{0.5}$N/GaN superlattice. It is known that the density also represents the possibility density of finding a particle at a given position inside the quantum well. From Fig. 3, one can see that the possibility strongly depends on the number of energy levels and the shape of the spatial nodes of densities. In principle, the optical transition is a process of recombination between CB electrons and VB holes. Therefore, the possibility of finding both the electrons and holes at the same position, namely, e-h spatial overlap, is of importance in determining the optical transition and spectral properties. One can find that the e$_0$-h$_0$ and e$_0$-h$_1$ overlap rate actually is negligibly low. Whereas, the higher overlap exists between the e$_0$ and h$_2$ levels in the left hand side of the well. To study the selection rule of interband transitions, an alternative scheme was used to determine the contribution of different quantized states to the band-edge peak. As shown in Fig. 4(a), the complete absorption spectrum of the Al$_{0.5}$Ga$_{0.5}$N/GaN superlattice is calculated. Then, by switching off the quantized levels one by one, the origin of z$_1$ peak can be determined. One can see in Figs. 4(b) and 4(c) that the lowest two hole levels h$_0$ and h$_1$ have rather small contributions. Together with Fig. 4(d), conclusion can be drawn that the z$_1$ band edge absorption derives mainly from the e$_0$-h$_2$ intersubband transition. Clearly, the z$_1$ energy, which represents the optical band gap of GaN/GaN superlattice, is higher than the electronic band gap. Therefore, the e$_0$-h$_2$ intersubband transition eventually governs the actual optical gap of this superlattice system. Furthermore, this transition occurs locally in the narrow region from 2.1 to 2.6 nm inside the GaN well, as shown by the arrow in Fig. 3. This strong spatial localization of the occurrence of transitions between quantized states shows the crucial mechanism leading to the sharp z$_1$ intensity.

In summary, our ab initio calculations performed on the Al$_{0.5}$Ga$_{0.5}$N/GaN superlattice have revealed the quantized electronic properties and the underlying mechanism of optical transitions. Distribution of the discrete energy levels inside the GaN quantum well was obtained with respect to the complete band profile. The shapes and variations in the corresponding quantized states were detailed, which demonstrated the strong well-in-well confinement, forming the active region in the GaN well. Based on the envelopes of the discrete energy levels and absorption spectra, the origin of band-edge peak was assigned to the e$_0$-h$_2$ intersubband transition. Our findings will be valuable for the future design of functional optoelectronic nanostructures.

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