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博　士　学　位　论　文

III 族氮化物材料弱极性晶面量子阱的生长及其光电特性的研究

Research on Growth and Optical Properties of Semipolar III-Nitrides Nanostructures

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摘 要

III 族氮化物半导体材料具有许多优良的性质，其纤锌矿结构晶体具有宽禁带，直接带隙，化学特性稳定，耐高温，质地坚硬等特点。近十几年对其的研究取得了瞩目的成果，商业价值巨大。具体应用包括显示，照明，存储，探测器，通讯，农作物生长照明，医疗健康，以及电力电子器件等领域。尽管如此，III 族氮化物材料在发光器件应用领域仍然面临许多需要解决的问题，例如：电极化效应，高缺陷密度，光提取效率低，droop 效应，Green Gap 等。

为了改善上述问题，科研人员提出了多种研究方案，其中采用弱极性量子阱结构是比较有前景的研究方向之一。弱极性量子阱具有阱内极化电场较小，发光效率高，适合制作长波段器件，droop 效应低等优点。因此，弱极性晶面量子阱的相关研究逐渐受到重视，尤其是在光电特性以及阱内动态复合机制方面的研究。在此背景下，本文围绕如何改善 III 族氮化物量子阱结构发光效率这一主题，首先研究了采用选区外延技术生长的$\{11\bar{2}2\}$和$\{1\bar{1}01\}$两种弱极性晶面量子阱的光电特性和阱内动态复合机制的特点，其次提出在弱极性量子阱结构中引入阱内 In 组分渐变技术来进一步优化其发光效率，并通过理论计算对阱内 In 组分渐变的形式进行了优化研究。本文主要研究内容和结论摘要如下：

1. 利用 MOCVD 外延设备并采取选区外延技术来生长立体形貌的$\{11\bar{2}2\}$和$\{1\bar{1}01\}$弱极性量子阱。根据变温光致荧光（PL）实验的测试结果得出：$\{1\bar{1}01\}$弱极性量子阱 LED 结构的室温 PL 归一化强度约为 6.2%，比极性阱提高一倍多，也就是说其相对内量子效率比极性阱提高一倍多。此外，$\{11\bar{2}2\}$弱极性阱结构的室温 PL 归一化强度约为 12%，表现出比$\{1\bar{1}01\}$阱还要高的效率。变温时间解析光致荧光测试和理论计算验证了弱极性量子阱具有较高效率的结论。

2. 根据对样品变温光致荧光测试和变温时间解析光致荧光测试的结果，本文讨论了弱极性量子阱的阱内载流子的动态复合过程，解释了弱极性量子阱光荧光光谱峰值随着温度增加并没有出现明显的类“S”型变化的原因，提出由于弱极性量子阱阱内极化电场的减弱，导致载流子与声子耦合强度减弱，使得载流子 hopping/tunneling 等运动相对极性量子阱要弱。这表明弱极性量子阱的阱内载流子有更大机率进行原位辐射复合，而载流子的再分布过程相对较弱。这一特性有助于降低载流子被缺陷捕获的机率，提高弱极性量子阱的辐射复合效率。

3. 本文提出了将 In 组分渐变技术引入到弱极性量子阱结构中，来优化弱极性量子阱的辐射复合效率。并采用$\boldsymbol{k}\cdot\boldsymbol{p}$能带理论，研究了不同的阱内 In 组分渐变形式对弱极性量子阱光电特性的影响。通过计算，获得了导带和价带能带结构，阱内波函数分布，并进而获得波函数交叠积分，跃迁矩阵元，自发辐射谱，偏振率等参数。根据计算结果讨论得出：电子-空穴的波函数交叠积分和自发辐射谱（y'偏振）随着阱内 In 组分最大值和最小值差值的增加而增加，随着阱内 In 组分最大值沿着恒定组分弱极性量子阱阱内极化电场相反的方向移动而增加。其中，在类抛物线渐变 3nm 量子阱中，当 In 组分最大值的相对位置为 3/4，且 In 组分最小值为 0%的结构电子-空穴波函数交叠积分达到了 94.15%，这相对于组分恒定极性量子阱的 30.37%和组分恒定的弱极性量子阱的 83.74%都有了很大的提升。

4. 根据$\boldsymbol{k}\cdot\boldsymbol{p}$能带理论计算结果，本文还进一步讨论了 In 组分渐变弱极性量子阱的光偏振特性。得出 In 组分渐变弱极性量子阱的偏振比率 $\rho_{y'x'}$随着阱内 In 组分最大值和最小值差值的增加而降低，随着阱内 In 组分最大值沿着恒定组分弱极性量子阱阱内极化电场相反的方向移动而增加。其中，阱内 In 组分最大值的相对位置为 3/4 且 In 组分百分比最大值与最小值差值为 5%的结构偏振比率 $\rho_{y'x'}$最大，达到了 44.6%；而阱内 In 组分最大值的相对位置为 1/4 且 In 组分最小值为 0%的结构偏振比率 $\rho_{y'x'}$最小，数值为 36%。

5. 此外，对不同阱宽结构的理论计算结果表明，随着弱极性量子阱阱宽的增加，阱内 In 组分渐变形式经过优化的弱极性量子阱的波函数交叠积分尽管会随着阱宽的增加而降低，但是其降低的速率比阱内 In 组分恒定的弱极性量子阱要慢。阱内 In 组分渐变弱极性量子阱的偏振比率 $\rho_{y'x'}$随着阱宽的增加而增加，但是增加速率相对于阱内 In 组分恒定的弱极性量子阱要慢。

关键词： III 族氮化物半导体，弱极性晶面，量子阱结构，SAE 外延技术，组分渐变。

Abstract

III-nitrides and their alloys are highly attractive materials due to their superior properties of wurtzite crystalline structure, including direct and wide bandgap, high thermostability, chemically stable, hard texture, etc. Remarkable progresses on scientific research have been achieved in past decades, and commercial devices have been overwhelmingly accepted by the markets, revealing of their highly commercial potentials. The applications of III-nitride semiconductors include displays, illuminations, detectors, digital storage, communication, crops lighting, healthcare, etc. However, challenges still remain for further improving, such as spontaneous and the piezoelectric polarizations, high density of threading dislocations, low light extraction efficiency, droop effect, Green Gap, etc.

Remedies have been proposed to mitigate the complications mentioned above. Adopting semipolar quantum wells (QWs) is one of the promising solutions. Semipolar QWs possess low internal field, high radiative efficiency, applicable to long wavelength LEDs devices, low droop effect and so on. Researches on semipolar QWs are attracting much attention, such as electronic properties, optical properties, carrier recombination dynamics. In this thesis, under the above circumstance, we focusing on improving the radiative-recombination efficiency of III-Nitrides QWs, discussed the optimized-growth conditions of selective area epitaxy (SAE) to grow semipolar QWs structures, and studied the optical properties of these 3D $\{11\bar{2}2\}$ and $\{1\bar{1}01\}$ semipolar QWs structures and the carrier recombination dynamics. Moreover, electronic and optical properties of indium-graded semipolar QW structures were studied with different indium variation schemes. Details of the content are summarized as below:

1. The 3D $\{11\bar{2}2\}$ and $\{1\bar{1}01\}$ semipolar QWs were grown by MOCVD using SAE technique on *basal* sapphire substrates with optimized epitaxy-parameters. Temperature-dependent photoluminescence (PL) measurement results showed that the $\{1\bar{1}01\}$ semipolar QWs has a normalized PL-integrated intensity of 6.2%, about 1 time higher than the polar QWs, this means that relative internal quantum efficiency (IQE) of $\{1\bar{1}01\}$ semipolar QWs is also about 1 time higher than the polar one. Moreover, the relative IQE of $\{11\bar{2}2\}$ semipolar QWs is about 12%, which is even higher than the $\{1\bar{1}01\}$ semipolar QWs. The superior radiative-recombination efficiencies of semipolar QWs were also proved by both time-resolved photoluminescence (TRPL) measurements and theoretical calculation results.

2. Based on temperature-dependent PL and TRPL measurements, carrier recombination dynamics were discussed. The weakened S-shaped temperature-dependent PL peak-energy of semipolar samples were explained by the weak phonon-assisted carrier in-plane hopping/tunneling processes, which originated from the weakened internal field and piezoelectric effect. This feature was further verified by PL spectra-dependent decay times and temperature dependent TRPL. The weakened carrier hopping/tunneling effects are beneficial to carrier in-suit radiative recombination processes rather than carrier redistribution among inhomogeneous potential fluctuation. This could help carriers from being captured by defects, which may lead to the participating of nonradiative processes.

3. The indium-graded technique was introduced in the semipolar well layers to improve the radiative-recombination efficiencies. And then the $\boldsymbol{k}\cdot\boldsymbol{p}$ theory was employed to investigate electronic and optical properties of indium-graded $\{11\bar{2}2\}$ semipolar QW structures with different indium variation schemes. The conduction and valence band structures, the electron and hole wave functions have been solved for all QW structures, and then the overlap of electron-hole wave functions, the transition matrix elements between the first conduction and the topmost valence bands, the spontaneous emission spectra, and the optical polarization ratio were studied. According to the calculation results, both increasing the indium composition difference (Δ_D) between the maximum and the minimum points in the well layer and moving the location of the maximum indium composition (MIC) in the opposite direction of the built-in field existing in the well layer of indium constant semipolar QW can improve the overlap of electron and hole wave functions, as well as the intensity of spontaneous emission rate spectra for y'-polarization of the indium-graded semipolar QW. For the 3 nm semipolar QW structure, the maximum overlap of 94.15% is achieved by the QW with MIC=3/4 and minimum indium composition of 0%, this result is much higher than the 30.37% of indium constant polar QW and 83.74% of indium constant semipolar QW.

4. The optical polarization ratio $\rho_{y'x'}$ decreases by larger Δ_D and increased by moving MIC to the opposite direction of the built-in field existing in the well layer of indium constant semipolar QW. The maximum $\rho_{y'x'}$ of 44.6% for the 3nm semipolar QW is achieved by MIC=3/4 and Δ_D=5%, and the minimum $\rho_{y'x'}$ of 36% is achieved by MIC=1/4 and minimum indium composition of 0%.

5. The overlaps of optimized indium-graded semipolar QWs are decreased with the increasing of well width, but with slower decreasing rates than the indium constant semipolar QW. The $\rho_{y'x'}$ of indium-graded semipolar QWs is increased with the well width but also in a slow increasing rate compared with the indium constant semipolar QW.

Keywords: III-nitrides, semipolar, quantum wells, SAE, indium-graded technique.

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