

传输矩阵所描述的界面是光滑的理想介质界面。而实际界面粗糙程度的增加造成光线入射角度的改变以及散射现象，从而使反射率出现起伏。带宽变窄的可能原因是相邻界面处一定程度的 Al、Ga 组分扩散和混合生长。计算表明，两种材料的折射率差减小会导致带宽变窄。由于 MBE 生长过程中，在 GaAs 和 AlAs 界面处 Al 炉和 Ga 炉切换开闭，其束流变化均经过一渐变过程才达到稳定，因而在小区域内形成 AlGaAs 层，导致有效折射率发生变化。

第三，测试谱在波长超过 950nm 后反射率即出现明显的下降趋势，这是由于 GaAs 层厚度略小于设计值造成的。根据我们对改变层厚的模拟计算显示，不同折射率材料的厚度差 (Δd) 增大，会使高反带出现向右侧倾斜的趋势。

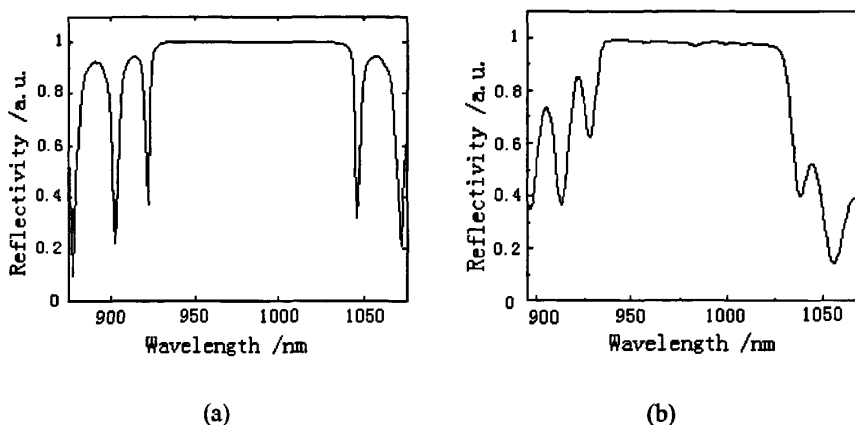


图 6.5 反射率谱 (30 对 DBR, 980nm) (a)计算结果 (b)实验测得结果

根据上述分析，计算了 980nm AlAs/GaAs DBR 的反射谱，并对生长工艺参数进行了优化。图 6.5 为中心波长 980nm，30 对 AlAs/GaAs DBR 样品的反射率谱计算结果（图 6.5 a）和实验测得结果（图 6.5 b）。与 920nm 样品相比，DBR 的最大反射率达到 99.5%，虽然仍略小于计算值，但高反射率区域相对平坦，并未出现明显的起伏现象，这表明材料的界面状况有了较大改善。样品的中心波长和带宽非常接近预期值，结果达到了优化设计的要求。

6.1.4 结论

利用传输矩阵的方法计算了不同对数 GaAs/AlAs 多层分布反馈布拉格反射镜的反射率曲线。设计生长了中心波长为 920nm 和 980nm、高反射率的半导体

多层膜 DBR 反射镜。反射率谱拟合与实验结果的比较显示,界面平整度不理想会引起高反射率区域谱线的起伏,Al、Ga 的组分混合会导致高反射率带宽改变,而 GaAs/AlAs 层周期厚度偏差则是导致中心波长漂移的主要原因。在上述研究的基础上,通过对生长工艺的优化,实现了多层厚度的精确控制,获得了中心波长为 980nm,最大反射率达 99.5%的高质量 DBR 材料。

第二节 GaAs/AlGaAs 多量子阱材料的非线性吸收效应研究^[14]

6.2.1 引言

近几十年来,因为在制作光学双稳开关、半导体激光器、光信息处理和别的非线性光学器件方面具有潜在的优势,半导体多量子阱非线性光学效应引起了人们相当大的研究兴趣^[15~18]。在设计这些器件时,非线性参数是非常重要的。材料的非线性性质可以通过许多方法来测量,比如非线性干涉测量计^[19]、简并四波混频^[20]、近简并三波混频^[21]、椭偏仪^[22]和光束扭曲测量^[23-24]等。与这些实验方法相比,反射 Z 扫描技术被认为是最简单和有效的单光束测量方法之一,可以用来测量小厚度样品(比如衬底上的薄膜)的非线性吸收系数。采用这种方法,被非线性材料所反射的入射光束的空间界面修正通过一个放置在远场区的小孔所监控,这被称做“闭孔”测量,用于测量轴上的反射光强。这样,反射光束的相位扭曲被转换为可以被光电探测器探测的强度变化。通过分析实验数据,可以得到非线性吸收系数。如果小孔被移走,则为“开孔”测量,可以探测到所有的反射光强,通过分析实验数据,可以得到非线性折射率。

在本节中,通过采用反射 Z 扫描方法,测量了 GaAs/AlGaAs 多量子阱的室温三阶非线性系数。并且一个基于速率方程和带填充效应的模型被展示来计算非线性吸收系数。

6.2.2 反射 Z 扫描实验及结果拟合

GaAs/AlGaAs 多量子阱样品采用 MBE 设备在半绝缘的 GaAs 衬底上生长。每个样品的结构为:首先是一个 GaAs 缓冲层,接着是 30 周期的由 5 nm GaAs 和 7 nm AlGaAs 组成的多量子阱层,最后是一个 100 Å GaAs 帽层来阻止氧化。

在样品 TD001, TD002 和 TD003 中, 垒层的 Al 组分分别是 0.22, 0.19 和 0.34。所有各层的生长温度都是 610℃。

反射 Z 扫描的实验布局如图 6.6 所示。激发光源为 532nm 的 YAG 激光器, 脉冲持续时间 30ps, 重复频率为 10Hz, 束腰半径 18μm。激光脉冲的能量约几百纳焦耳。

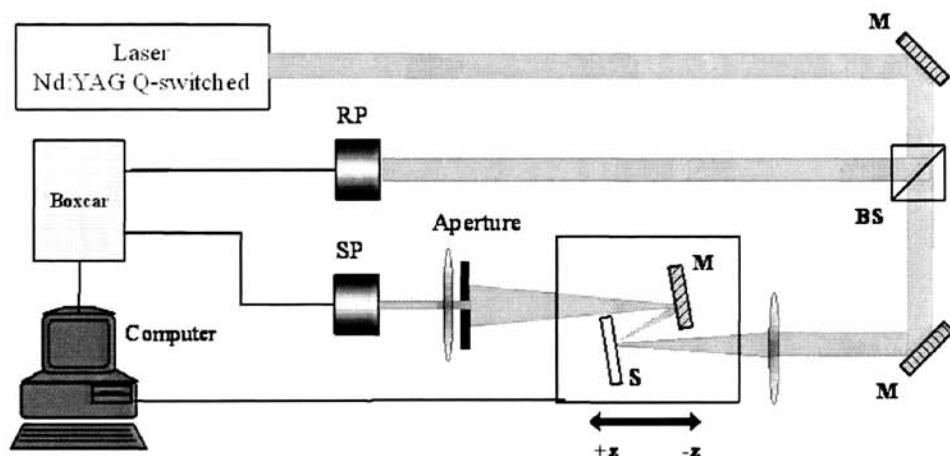
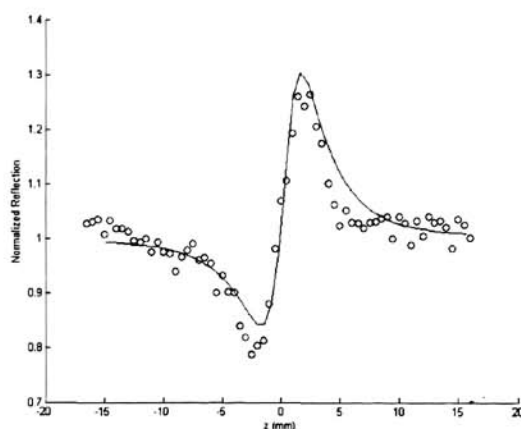
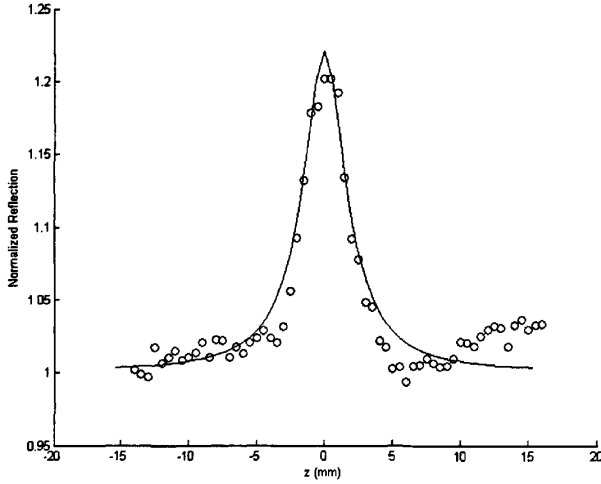


图 6.6 反射 Z 扫描实验布局

在上述实验条件下, 我们测量了对应于开孔和闭孔条件下归一化反射光强与样品位置的关系, 如图 6.7 中分立点所示。



(a) 闭孔测量条件下的归一化轴上光强



(b) 开孔测量条件下的归一化总反射光强

图 6.7 TD003 样品的 RZ-scan 测量结果与拟合曲线

以下对实验数据进行数据拟合^[25~27]。考虑一束沿 Z 轴传播的高斯入射光束，它的束腰处在 $z=0$ 。束腰和样品表面的距离是 z ，小孔放在距束腰为 d 的地方。反射光束的菲涅尔系数包含了由于非线性效应导致的折射率变化和消光系数变化以及可能存在的表面热扩散导致的反射波的相位变化。因此，有效反射系数为

$$R(z, r) = \left\{ \frac{\hat{n}(z, r) - 1}{\hat{n}(z, r) + 1} \right\} \exp \left[i 2kU |E(z, r)|^2 \right] \quad (6.9)$$

这里

$$\hat{n}(z, r) = n_0 + n_2 |E(z, r)|^2 + i[\kappa_0 + \kappa_2 |E(z, r)|^2] \quad (6.10)$$

是复折射率， n_0 和 κ_0 是线性折射率和消光系数， n_2 和 κ_2 是非线性折射率和非线性消光系数， $E(z, r)$ 是界面处的发射波， $U|E(z, r)|^2 = H$ 是表面扩张幅度， k 是波数。因此，方程(4.19)显示了反射波是由表面非线性吸收和表面扩张引起的。另一方面，表面非线性折射率则对反射波起振幅调制的作用。考虑光致反射率改变，样品界面空气处的镜面反射波场为：

$$\begin{aligned}
E_R(r, z, t) = & E_0(t) \frac{w_0}{w(z)} \exp \left[-i\Phi(z) - \frac{ik}{2R(z)} \right] \exp(ikz) (R_0 \exp[-r^2/w^2(z)] + \\
& R_1(n_2 + i\kappa_2) |E_0(t)|^2 [1 + (z/z_0)^2]^{-1} \times \exp[-3r^2/w^2(z)] + \\
& R_0 \sum_{m=1}^{\infty} \frac{\{2ikU|E_0(t)|^2 [1 + (z/z_0)^2]^{-1}\}^m}{m!} \exp[-(2m+1)r^2/w^2(z)]
\end{aligned} \quad (6.11)$$

式中 $E_0(t)$ 是 $r=0$ 处的电场振幅, w_0 是束腰半径, $w(z) = w_0[1 + (z/z_0)^2]^{1/2}$ 是光斑半径, $z_0 = kw_0^2/2$ 是光束衍射长度, $R(z) = z(1 + z_0^2/z^2)$ 是在 z 处的弯曲波前半径, $R_0 = (n_0 - 1 + i\kappa_0)/(n_0 + 1 + i\kappa_0)$ 是线性反射系数, $R_1 = 2(n_0 + 1 - 2i\kappa_0)/(n+1)^3$, $\Phi(z) = \tan^{-1}(z/z_0)$ 。方程 (4.21) 中第一项描述光的线性反射, 第二项包括了非线性极化率的实部和虚部, 最后一项是表面扩张的贡献。

为了计算孔径平面处光斑的远场模式, 我们采用“高斯分解方法”。利用高斯光束的性质, 孔径平面处轴上 ($r=0$) 光强分布是表面位置 z 的函数, 它的形式为:

$$\begin{aligned}
I_R(z) = & I_0 | (R_0 G_0^{-1}(z) + R_1(n_2 + i\kappa_2) I_0 [1 + (z/z_0)^2]^{-1} G_1^{-1}(z) + \\
& R_0 \sum_{m=1}^{\infty} \frac{\{2ikU I_0 [1 + (z/z_0)^2]^{-1}\}^m}{m!}) G_m^{-1}(z) \times (1 - iz/z_0)^{-1} |^2
\end{aligned} \quad (6.12)$$

其中 $g(z) = 1 + (z+d)z/(z_0^2 + z^2)$, $G_m(z) = \{g(z) - i \times (2m+1)(z+d)/[kw^2(z)/2]\}$, $I_0 \propto |E_0(t)|^2$ 。

通过上式, 可得到拟合曲线如图 6.7 中实线所示, 并可计算出相应不同样品的非线性消光系数 κ_2 和非线性折射率 n_2 。根据非线性吸收系数 α_2 和非线性消光系数 κ_2 之间的关系,

$$\alpha_2 = \frac{4\pi}{\lambda} \kappa_2 \quad (6.13)$$

还可计算得到各样品的非线性吸收系数, 具体计算结果如表 6.1 所示。非线性吸收系数前的负号表示该吸收属于饱和吸收。从表中可以看出, 虽然有 Al 组分和 $z=0$ 处入射光强 (即中心光强) 两个影响因素, 但是非线性吸收系数只是主要取决于后者; 换句话说, 中心光强越弱, 则非线性吸收系数越大。这个现象可以被后面的模型所解释。

表 6.1 $z=0$ 处的入射光强和非线性系数的拟合数据

sample	Al fraction	I_0 (W/cm ²)	α_2 (cm/W)	n_2 (cm ² /W)
TD001	0.22	1.76E+9	-4.30 E-4	2.20E-11
TD002	0.19	1.48E+9	-6.50E-4	6.50E-11
TD003	0.34	1.39E+9	-1.80 E-3	5.55E-11

6.2.3. 讨论

半导体多量子阱材料的非线性吸收，与载流子的密度和载流子在能带中的填充状态都有密切关系。当入射光场的光子能量 $\hbar\omega$ 等于或大于材料能隙 E_g 时，在半导体中会产生电子从价带到导带的激发，从而得到大量的电子和空穴。在热平衡情况下，粒子态的分布由费米-狄拉克函数来描述。这个函数包括两个参数：温度 T 和费米能级 E_F ，它们对所有的能级都适用。在光泵浦的情况下，粒子态的可能分布会被快速的谷间散射过程（通常是亚皮秒量级）所热弛豫。粒子态的分布仍然保持费米-狄拉克函数的形式，但是费米能级 E_F 在不同的带中是不同的，我们采用准费米能级 E_{FC} 和 E_{FV} 来分别描述导带和价带的情况^[28~29]。

在严格的动量守恒假设下，量子阱的吸收系数可以写为

$$\alpha = \frac{\pi e^2}{\epsilon_0 N_r c a m_0^2} \int dE \sum_n \sum_{j=l,h} \rho_{nj}(E) M_j^2(E) \cdot [f_{cn}(E) - f_{jn}(E)] L(E) \quad (6.14)$$

这里 $\rho_{nj}(E)$ 是折合态密度函数， $\hbar$ 是普朗克常数， e 是电子电荷， c 是光速， ϵ_0 是自由空间介电常数， m_0 是电子质量， N_r 是介质有效折射率， $M_j^2(E)$ 是偶极矩阵元的平方， $f_{cn}(E)$ 和 $f_{jn}(E)$ (脚标 c 代表电子， $j = h$ 指重空穴， $j = l$ 指轻空穴) 分别是与第 n 个量子化能级对应的导带和价带的费米电子占据几率。由于导带量子能级和价带量子能级的数目不等，所以 (6.14) 式的第一项求和的上限由这两个数中较小的一个给出。

$L(E)$ 唯象地表示了带内弛豫过程的洛伦兹函数，

$$L(E) = (1/\pi) [\Lambda(E_j)/2] / [(E_i - E)^2 + (\Lambda(E_j)/2)^2] \quad (6.15)$$

这里 $\Lambda(E_j)$ 是能级 E_j 的碰撞展宽的宽度。使用折合有效质量 m_{ij}^* 和严格的 k 选择定则，吸收表达式的各个量可以被写为：

$$m_{ij}^* = (m_c^* m_{jh}^*) / (m_c^* + m_{jh}^*) \quad (6.16)$$

$$\rho_{ij}^*(E) = [(4\pi m_{ij}^*) / (\hbar^2 L_z)] H(E - E_{gw} - E_{cn} - E_{jn}) \quad (6.17)$$

$$E_j = E_{cn} + (m_{ij}^* / m_c^*)(E - E_{gw} - E_{cn} - E_{jn}) \quad (6.18)$$

$$f_{cn}(E) = \{1 + \exp[(E_{cn} + \{m_{ij}^* / m_c^*\} \cdot \{E - E_{gw} - E_{cn} - E_{jn}\} - E_{FC}) / (kT)]\}^{-1} \quad (6.19)$$

$$f_{jn}(E) = \{1 + \exp[(-E_{jn} - E_{gw} - \{m_{ij}^* / m_{jh}^*\} \cdot \{E - E_{gw} - E_{cn} - E_{jn}\} - E_{FV}) / (kT)]\}^{-1} \quad (6.20)$$

式中 $H(E - E_{gw} - E_{cn} - E_{jn})$ 是一元 Heaviside 函数， E_{gw} 是量子阱层的体材料带隙， k 是波尔兹曼常数， T 为温度。

偶极矩阵元的平方的表达式为

$$M_h^2 = (3/4)M_0^2(1 + E_{cn} / E_j) \quad (6.21)$$

$$M_l^2 = 2M_0^2 - M_h^2 \quad (6.22)$$

这里 M_0^2 是体材料的偶极矩阵元 $M_0^2 = (4/3)m_0 E_{gw}$ 。半导体量子阱中的电子和空穴的密度为：

$$N_e = \frac{4\pi m_c^*}{h^2} \sum_n \int_{E_{cn}}^{\infty} dE \frac{H(E - E_{gw} - E_{cn} - E_{jn})}{1 + \exp[(E - E_{FC}) / (kT)]} \quad (6.23)$$

$$N_v = \frac{4\pi m_{jh}^*}{h^2} \sum_n \int_{E_{cn}}^{\infty} dE \frac{H(E - E_{gw} - E_{cn} - E_{jn})}{1 + \exp[(E - E_{FV}) / (kT)]} \quad (6.24)$$

令入射光场的光强为 I ，则可得到如下的激发电子密度的速率方程为

$$\frac{dN_e}{dt} = \frac{\alpha I}{\hbar \omega} - \frac{N_e - N_{e0}}{\tau_R} \quad (6.25)$$

在稳态情况下则有激发电子密度为

$$N_e = N_{e0} + \tau_R \frac{\alpha I}{\hbar \omega} \quad (6.26)$$

其中 τ_R 本为载流子复合寿命（纳秒量级），由于实验所用激光为皮秒脉冲，脉冲持续时间短于载流子复合寿命，所以我们这里采用皮秒脉冲的持续时间。

考虑到材料中电子和空穴的个数相等，则有

$$N_e = N_v \quad (6.27)$$

材料的吸收由线性部分 α 和非线性部分 $\Delta\alpha$ 组成，即

$$\alpha = \alpha_0 + \Delta\alpha \quad (6.28)$$

由于入射波长远远小于材料的带边吸收波长，我们采用 GaAs 体材料的线性吸收系数作为量子阱的线性吸收系数，在 532nm 处，这个值是 $7 \times 10^4 \text{ cm}^{-1}$ [30]。

在上述理论中，我们忽略了 AlGaAs 层的吸收。这是因为，量子阱中的能级位置远低于垒层的导带底，而载流子倾向于占据较低的能级，所以量子阱中由载流子分布导致的带填充效应要比垒层中更明显。

我们采用自洽方法来解上述的方程组，可以获得材料中的电子和空穴的准费米能级，以及获得材料中的非线性吸收系数。在我们的模型中，吸收变化对 $z=0$ 处光强的比值可以反映非线性吸收系数，即

$$\alpha_2 \propto \frac{\Delta\alpha}{I_0} \quad (6.29)$$

当 $z=0$ 处光强变化时， $\Delta\alpha$ 和 I_0 的比值和电子空穴准费米能级之差的关系如图 6.8 所示。

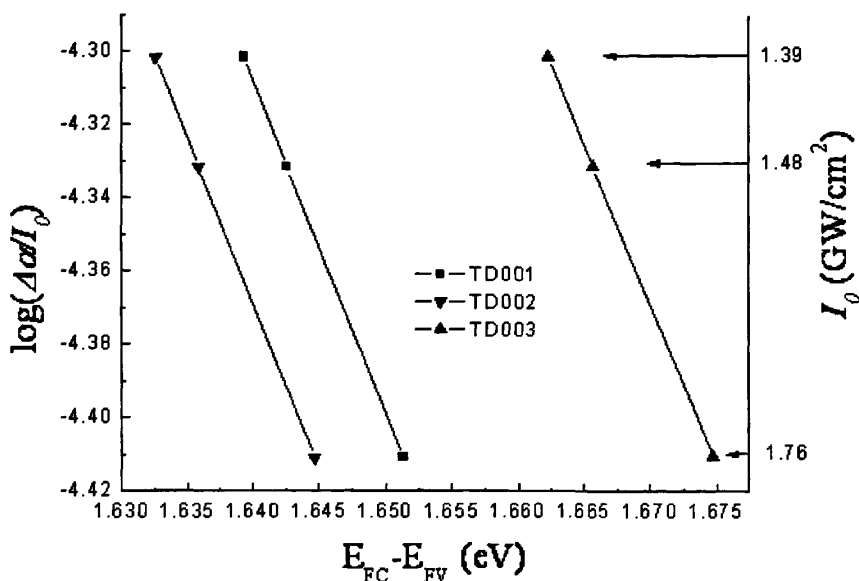


图 6.8 三个样品的 $\Delta\alpha$ 和 I_0 的比值和电子空穴准费米能级之差的关系

从图中可以看到，对同一个样品，非线性吸收系数 $\Delta\alpha/I_0$ 的对数与准费米能级之差有一个线性关系。这是由于，准费米能级之差正比于导带和价带的态密度，而非线性吸收系数也与导带和价带的态密度成正比。从图中还可以发现，当入射光强相同时，三个样品的非线性吸收系数几乎相同，即使它们有不同的 Al 组分和不同的准费米能级之差 $E_{FC} - E_{FV}$ ，后者是由量子阱中的能级位置决定的。这说明在目前的光强范围，Al 组分对非线性吸收系数的影响是不重要的。最后也是最重要的是，对同一个样品来说，非线性吸收系数随光强的增大而减小。

我们的实验数据可以在模型模拟的结果上很好地被解释。因为在目前的光强范围内 Al 组分对非线性吸收系数的影响是不重要的，并且对同一个样品来说，非线性吸收系数随着光强增大而减小，具有不同 Al 组分的三个样品才显示出它们的非线性吸收系数主要取决于光强。

6.2.4. 结论

通过采用反射 Z 扫描技术,研究了 GaAs/AlGaAs 多量子阱的室温非线性吸收效应。量子阱中的非线性效应可以被带填充效应所解释。一个可以计算量子阱非线性吸收系数的模型被提出,并很好地解释了实验现象。

第三节 本章结论

作为 VECSEL 的一个重要组成部分,半导体分布布拉格反射镜的制备和性质对于激光器的性能有重要的影响,我们采用传输矩阵模型,计算了 GaAs/AlAs DBR 的反射率,与实验结果能很好吻合;并在此基础上进一步指导实验,优化生长工艺,成功制备出反射率达 99% 以上、中心波长 980nm 的 GaAs/AlAs DBR。

成功地将反射 Z 扫描技术应用到 GaAs/Al_xGa_{1-x}As 多周期结构的非线性吸收研究中,并提出一个可以半定量计算非线性吸收系数的模型。实验和理论结果表明,在一定范围内, $z=0$ 时的入射光强是决定非线性吸收系数大小的主要因素。此外,该模型也展示了不同 Al 组分 x 对此多周期结构非线性吸收系数的影响,这在之前的研究中还未见报道过。

第七章 总结与展望

第一节 总结

InGaAs/GaAs 应变量子阱在现代光电材料和器件的应用中具有非常重要的地位。本论文“GaAs 基低维应变异质结和多周期结构的 MBE 制备及特性研究”从多个方面对 InGaAs/GaAs 应变量子阱的生长和性质进行了讨论和研究, 主要内容包括: MBE 系统原理与外延材料的表征方法、InGaAs/GaAs 多量子阱的生长与光学性质研究、快速热退火对 InGaAs/GaAs 应变量子阱光学性质的影响、应变补偿技术在 InGaAs/GaAs 应变量子阱中的应用以及多周期结构的性质研究等共五个主要部分。各部分的主要研究内容及研究结果如下:

1. 阐述了分子束外延设备系统原理、对真空系统的和 MBE 原位检测手段进行了介绍。讨论了外延层生长动力学和热力学原理, 研究了源炉温度和束流的精确控制。介绍了外延材料的几种常用表征手段, 对透射电子显微镜、X 射线双晶衍射、光致发光谱和拉曼光谱的原理及其特点进行了分析。

2. 制备出了具有优良室温发光特性的 InGaAs/GaAs 应变量子阱, 探讨了应变、临界厚度以及温度对应变量子阱带隙的影响, 研究了不同生长条件如生长温度、生长速度、界面生长停顿等对应变量子阱光学性质的影响。首次采用亚单层交替生长方式来生长 InGaAs 量子阱层, 增进了量子阱中 In 组分的均匀分布, 提高了 PL 谱的峰值波长发光强度; 并对亚单层生长方式对 InGaAs/GaAs 应变量子阱光学性质的提升进行了研究和讨论。

3. 从实验和理论上研究了快速热退火对 InGaAs/GaAs 应变量子阱光学性质的影响, 在退火时间 30 秒条件下, 退火温度为 650°C、700°C 和 750°C 的样品的 PL 峰值波长都发生了红移, 只在 800°C 退火时 PL 峰值波长蓝移; 在退火温度 750°C 条件下, 除了退火时间 60 秒的样品相对于退火时间 45 秒的样品的发光峰有一个蓝移之外, 发光峰波长整体向长波长方向移动。通过比较可以发现, 发光峰值波长红移量越多, 发光峰强度越小。这可以由应变弛豫造成的位错对 PL 发光峰强度的影响来解释。提出了新的拟合扩散激活能的方法, 得到了组分为 0.24 的样品的扩散激活能为 1.2eV, 该结果与文献的报道能够很好的吻合。与传统的传输矩阵方法处理相比, 减少了数据运算量。在此基础上, 我

们研究和讨论了扩散和应变弛豫在快速热退火过程中各自所起的作用：在退火时间 30 秒、不同退火温度条件下，应变弛豫提供了 PL 峰值能量移动的初始值，随后 PL 峰值能量移动随温度的变化主要受扩散影响；在退火 750°C、不同退火时间条件下，PL 峰值能量移动明显偏离扩散理论的结果，主要受应变弛豫的影响；而当样品先后经历 750°C、30 秒退火和 800°C、30 秒退火之后，PL 峰值能量的移动可以由扩散造成的影响来解释。

4. 进行了应变补偿技术在 InGaAs/GaAs 应变量子阱中的应用研究，得到了具有优良室温发光特性的多层 InGaAs/GaAs 应变量子阱；讨论了应变补偿的设计；通过对室温 PL 谱的计算，确认了未采用应变补偿的样品中，应变弛豫的发生及弛豫程度；通过结合 DCXRD 和 PL 谱的结果，详细讨论了应变弛豫样品中双峰的几种可能来源；在此基础上，讨论了应变补偿对量子阱光学性质的改进，以及不同补偿方式对量子阱光学性质的影响。进行了应变补偿对高分 InGaAs/GaAs 应变量子阱发光波长的拓展实验，得到了发光波长大于 1.1 μm 的多层应变量子阱，In 组分为 40%，室温发光特性良好。

5. 成功地将反射 Z 扫描技术应用到 GaAs/Al_xGa_{1-x}As 多周期结构的非线性吸收研究中，并提出一个可以半定量计算非线性吸收系数的模型。实验和理论结果同时表明，在一定范围内， $z=0$ 时的入射光强是决定非线性吸收系数大小的主要因素。此外，该模型也展示了不同 Al 组分 x 对此多周期结构非线性吸收系数的影响，这在之前的研究中还未见报道过。采用传输矩阵模型，计算了 GaAs/AlAs DBR 的反射率，与实验结果能很好吻合；并在此基础上进一步指导实验，优化生长工艺，成功制备出反射率达 99% 以上、中心波长 980nm 的 GaAs/AlAs DBR。

第二节 展望

以 GaAs 基为代表的低维应变半导体异质结具有丰富的物理内涵与广泛的应用前景，在未来一段时间内该领域的研究依然是半导体材料与器件的前沿。分子束外延技术是一门系统而工艺复杂的技术，如何利用 MBE 方法，实现高性能材料的精确可控生长，是获得高质量光电器件的核心。本论文通过对 MBE 制备材料的研究，为今后的工作打下了良好的基础。同时，也还有大量的工作值得进一步的分析和研究：

1. 应变补偿技术已经证明其对大应变、高组分 InGaAs/GaAs 应变量子阱生长和应用的重要性。同时,使用应变补偿技术可以使作为有源区的 InGaAs/GaAs 应变量子阱的层数增加很多,可以极大地提升半导体激光器的输出功率。如何更加深入地认识应变补偿的本质,如何更加有效地把应变补偿结合到器件的实际应用中,还需要相当多的研究。

2. 本论文所讨论的 InGaAs/GaAs 应变量子阱主要针对在半导体激光器,尤其是在垂直外腔表面发射激光器中的应用。作为一个完整的器件,其对作为有源区的量子阱势必会有更多的要求,这还需要进一步的认识和研究。

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攻读博士期间发表论文情况

发表的论文:

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