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InAs 自组织量子点的光学性质研究

**The Study of Optical Properties on InAs
Self-organized Quantum Dots**

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摘要

由于半导体量子点具有零维电子特性，它不仅成为基本物理研究的重要对象，也成为研制新一代量子器件的基础。正因如此，量子点材料及器件成为目前国际上最前沿的研究课题之一。GaAs 基 InAs 自组织量子点因其成本低廉、器件工艺成熟，成为替代 InP 基材料、制备光纤通信用 1.3—1.55 μm 发光激光器的热门材料之一。本文采用分子束外延技术制备了高质量的 GaAs 基 InAs 自组织量子点材料。利用原子力显微镜 (AFM)、扫描电子显微镜 (SEM)、变温及时间分辨的光致发光谱 (PL) 等手段，分别研究了 InGaAs 应变层 (指在 InGaAs 层上生长量子点，下同)、InGaAs 盖层、InGaAs/InAlAs 联合盖层、InGaAs/GaAs 量子阱和多层生长等多种结构对量子点材料光学性质的影响，取得了以下主要结果：

1. 原子力显微镜 (AFM) 测量结果表明，与在 GaAs 层上直接生长 InAs 量子点相比，引入 $\text{In}_{0.1}\text{Ga}_{0.9}\text{As}$ 应变层，可以有效地释放量子点中的应变，使量子点的密度显著增大。当 InAs 淀积厚度较小时，量子点连在一起，形成了具有强烈耦合效应的量子点串结构，这种结构的变化导致量子点 PL 峰强烈红移，室温发光波长达到 1.3 μm 以上。

2. 首次系统研究了量子点发光寿命与量子点的密度、尺寸以及温度变化的关系。发现在温度低于 50 K 时，量子点的发光寿命基本不随温度变化；高于 50 K，发光寿命随着温度升高首先增加，温度升高到某一特定温度 T_c 后，开始减小。我们认为，量子点发光寿命同时由内在因素和外在因素决定，其中内因主要是指辐射复合，而外因包括载流子热发射、迁移、非辐射复合等。决定量子点辐射复合寿命的根本因素是量子点内电子—空穴跃迁振子强度：即跃迁振子强度越大，其辐射复合寿命越小，反之亦然。不同量子点之间载流子波函数的交叠，减弱了激子的相干性使量子点发光寿命增大，同时也增强了载流子从尺寸较小量子点到尺寸较大量子点的迁移

过程。这也是导致量子点发光寿命具有不同的尺寸效应的根本原因：当量子点密度较小时，发光寿命随量子点尺寸增大而减小；而当量子点密度较大时，发光寿命随量子点尺寸增大而增大。随着温度进一步升高，从势垒层及浸润层弛豫到量子点中的载流子数目增加及载流子在不同量子点中的迁移与扩展，导致量子点发光寿命增大。当温度高于 T_c 时，热发射、非辐射复合导致量子点发光寿命随着温度升高而减小。

3. 采用变温光致发光谱和时间分辨谱等手段，系统地研究了 InGaAs/InAlAs 盖帽层对 GaAs 基多层 InAs 量子阱的稳态及动态发光特性的影响。发现多层生长的方法可以改善量子点尺寸的均匀性，提高 InAs 量子点材料发光的温度稳定性。InGaAs 盖帽层使多层生长量子点具有更长的发光波长和更窄的谱线，是由于载流子在不同量子点间的迁移效应减弱。利用 InGaAs/InAlAs 联合盖帽层可以更有效地缓冲量子点中的应力和抑制 In 组分的偏析，使 InAs 量子点的发光进一步向长波移动(室温 1337nm)。同时，联合盖帽层可以提高量子点对载流子的限制势垒，提高量子点的发光效率。

4. 研究发现，在 InGaAs/GaAs 量子阱中生长 InAs 量子点样品，可以同时具有应变层和盖帽层的优点，更有效地缓冲 InAs 量子点中的应力，提高量子点的生长质量，从而可以在室温下探测到较强的发光信号。与直接在 GaAs 衬底或者在 InGaAs 应变层上生长相比，阱中生长模式可以使发光向长波长移动（室温达到 1318nm），半高宽减小（最大的半高宽为 38meV，最小的半高宽只有 25meV）。

5. 通过在 InAs/GaAs 量子点上覆盖 GaAs/InAs 短周期超晶格，获得了室温发光波长在 1.48 μm 的发光材料，该发光峰的积分强度从 15K 到室温仅仅减小一半，而另外一个发光峰（SLs）减小到 $1/10^5$ 。

关键词：InAs 自组织量子点；载流子；光致发光谱；寿命

Abstract

Quantum dots (QDs), with zero-dimensional electronic properties, have stimulated great interest due to their important roles in fundamental physical research and for developing novel devices. In recent years, it has been one of frontier topics of materials science to study the characterization of self-organized quantum dots and device applications. GaAs-based In(Ga)As self-organized QDs have become another promising gain material for optical fibre communication other than InP-based semiconductor lasers due to relatively inexpensive cost and maturity of fabrication on devices. In this thesis, high quality GaAs-based InAs self-organized QDs were grown by molecular-beam epitaxy (MBE). The QDs were systematically researched by atomic force microscopy (AFM), scanning electric microscopy (SEM), temperature-dependent and time resolved-photoluminescence (TDPL and TRPL). Results are summarized as follows:

1. The influences of a thin $\text{In}_{0.1}\text{Ga}_{0.9}\text{As}$ layer(5 nm) grown on GaAs(100) substrate before deposited InAs QDs were experimentally investigated. It is shown that $\text{In}_{0.1}\text{Ga}_{0.9}\text{As}$ strained layer could release the strain between wetting layer (WL) and QDs, and then significantly increased the density of QDs. When the InAs layer was thinner, the QDs are chained and strong coupled with adjacent dots. The photoluminescence peaks of InAs QDs with $\text{In}_{0.1}\text{Ga}_{0.9}\text{As}$ (emitting at above $1.3\mu\text{m}$.) show much red shift compared with the QDs directly deposited on GaAs matrix.

2. Firstly, the relations between the density and size of QDs and their PL lifetimes have been studied. The results showed that the PL life was insensitive to the temperature below 50 K, and then increases with the temperature until a

critical temperature T_C , after the T_C , finally decreases with the temperature. We consider the radiative recombination lifetime at low temperature is originated from the intrinsic factor that the oscillator strength of electron and hole in the QDs and the larger oscillator strength results in the smaller lifetime. But the wave function overlap of inter-dots may reduce the oscillator strength of intra-dots, which lengthen the lifetime. It is also the ultimate reason that PL lifetimes increase with the size of QDs for the low QDs density, while the PL lifetimes decrease with the size for high QDs density. The main reason for the PL lifetime increasing with temperature is that more carriers migrate to the QDs from barriers and WL (Wetting layer). Thermal emission and the non-radiative recombination reduce the PL lifetime above T_C .

3. The effects of InGaAs/InAlAs cap layers to the stable and dynamic optical characteristics of GaAs-based InAs QDs with multi-layer structures were systematically studied by the temperature-dependent and the time-resolved PL spectra. The uniformity of the dots may be improved for self-align effects after multi-growth, which resulted in the diminution of PL emission full width at half maximum and benefited to the thermal stabilization of the PL properties. Longer and narrower emission were obtained from multiplayer InAs QDs with InGaAs cap-layer which was probably due to reducing carriers migration among different QDs. We found that InGaAs/InAlAs combined cap layer could effectively release the strain in the QDs and suppress the segregation of In component, which made the QDs emitting at even longer wavelength(1337 nm at room temperature). At the same time, PL efficiency was enhanced for the higher potential barrier of InAlAs layer than that of InGaAs layer. The results are useful for application of QDs devices.

4. The InAs QDs grown in an InGaAs/GaAs quantum well(DWELL) could combined both the effects of InGaAs buffer layer and cap layer and even effectively release the stress between the buffer layer and the QDs, which may greatly improve the QDs quality. The DWELL samples could be detected strong signal at room temperature. Comparing with QDs grown on InGaAs or GaAs matrix, the PL of DWELL which was emitted at 1318 nm at room temperature, was much longer than the other two types of structures.

5. By depositing GaAs/InAs short period superlattices (SLs), 1.48 μ m emission is obtained at room temperature. Temperature dependent PL measurements show that the PL intensity of the emission is very steady. It decreases only to half as temperature increasing from 15K to room temperature, while at the same time, the intensity of the other emission decreases by a factor of 5 orders of magnitude. We attribute these two emissions to large-size QDs and short period superlattices (SLs) respectively. Large-size QDs are easier to capture and confine carriers, which benefits the lifetime of PL, and therefore makes the emission intensity insensitive to temperature.

Keywords: InAs self-organized quantum dots; carriers; photoluminescence spectra; lifetime

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第一章 绪论

§ 1.1 低维半导体材料概述

自从1969年L.Esaki和R.Tsu提出超晶格概念以来^[1-2],以半导体超晶格、量子阱、量子线和量子点为代表的低维半导体结构成为国际上研究的热点之一^[3-5]。这种完全由人工合成的新结构显示出天然晶体所不具有的许多新现象,从而予以半导体物理的基础研究新的刺激。从此人们所研究的材料对象便超出了自然界所赋予我们的那种现成式样^[6]。在低维半导体材料中,由于载流子在运动方向上受到限制,电子的能量因而发生量子化。随着量子尺寸的减小,会表现出明显的量子效应,如:量子限制、量子约束斯塔克效应^[7-8]、共振隧穿、库仑阻塞以及激子的吸收饱和非线性效应^[9]等。目前基于超晶格量子阱材料的量子器件,如高电子迁移率晶体管(HEMT)、异质结双极晶体管(HBT)、半导体量子阱激光器、自光电效应器件(SEED)、高频振荡器件,量子阱红外探测器等器件已经得到了广泛的应用。

理论上讲,一维量子线和零维量子点材料比量子阱具有更优越的性能^[10],以量子点为有源区的激光器将具有更低的阈值电流、更高的特征温度和微分增益^[11-12]。考虑到Pauli不相容原理,量子点尺寸的减小会使库仑阻塞效应更加显著,这可以用于单电子晶体管、存储器、探测器等的制作和研究。并且,利用量子点电偶极矩辐射耦合效应可以制备数字信息传输、计算单元等^[13]。总之,低维半导体结构的光电子、微电子器件将具有更高速、更低功耗、更多新功能等独特的优越性。

分子束外延(MBE)及金属有机化学气相沉积(MOCVD)等晶体生长技术的发展,使在原子级上精确制备二维层状半导体结构成为可能。而随着纳

米结构工艺和方法的进步，人们采用自组织生长方法已经可以制备无位错结构的量子点^[14-16]，这使得半导体量子点器件具有了更为诱人的发展前景。

§ 1.2 半导体中的电子态

人们已经建立了各种理论来描述量子点的电子态，最简单的是有效质量模型。我们首先讨论理想的限制情况。设势阱为无限深势阱，势能为：

$$V(\vec{r}) = \begin{cases} 0 & \vec{r} < a \\ \infty & \vec{r} \geq a \end{cases} \quad (1.1) \quad \text{其中 } a \text{ 为势阱宽度。}$$

根据薛定格方程(1.2)式及电子的能态密度函数 $\rho(E)$ (1.3)式：

$$\left[-\frac{\hbar^2}{2m^*} \nabla^2 + V(\vec{r}) \right] \psi(\vec{r}) = E \psi(\vec{r}) \quad (1.2) \quad \text{其中 } m^* \text{ 为电子有效质量。}$$

$$\rho(E) = \frac{V}{(2\pi)^3} \int \frac{1}{|\nabla_k E|} ds \quad (1.3)$$

对于三维体材料，电子运动的波函数及本征能量可表示为：

$$\psi(\mathbf{r}) = e^{i\vec{k}\cdot\vec{r}}, \quad E = \frac{\hbar^2 k^2}{2m} \quad (1.4)$$

因而电子的有效态密度为

$$\rho(E) = \frac{1}{(2\pi)^2} \cdot \left(\frac{2m^*}{\hbar^2} \right)^{\frac{3}{2}} \cdot E^{\frac{1}{2}} \quad (1.5)$$

对于二维体系，设量子阱界面垂直 z 方向，则电子运动的波函数可表示为：

$$\psi(x, y, z) = e^{i(k_x x + k_y y)} \sqrt{\frac{2}{a}} \sin \frac{n\pi z}{a}, \quad n=1, 2, 3, \dots \quad (1.6)$$

相应的本征能量: $E = E_{xy} + E_z$, 其中 E_{xy} 为连续谱, $E_{xy} = \frac{\hbar^2 k_{xy}^2}{2m^*}$, E_z 是量子化的。

电子的能态密度应为:

$$\rho(E) = \sum_n \rho_n(E) \quad \text{其中} \quad \rho_n(E) = \begin{cases} \frac{m^*}{\pi \hbar^2} & E \geq E_{nz} \\ 0 & E \leq E_{nz} \end{cases} \quad (1.7)$$

同理对于一维体系 (量子线), 同上可知:

$$\rho_{1D}(E) = \sum_{n,m} \left(\frac{2}{\pi \hbar} \right) \left(\frac{m^*}{2(E - E_x^n - E_y^m)} \right)^{\frac{1}{2}} \quad (1.8)$$

对于零维体系 (量子点), 电子在三个维度上的运动均受到限制, 其能量 E 是量子化的, 电子能态密度为: $\rho(E) = A \delta(E - E_n)$ 。如果设量子点在 x, y, z 方向上的长, 宽, 高分别为 a, b 和 c , 则为箱型量子点, 它的本征能量为:

$$E_{n,m,l} = \frac{\hbar^2 \pi^2}{2m^*} \left(\frac{n^2}{a^2} + \frac{m^2}{b^2} + \frac{l^2}{c^2} \right) \quad (1.9)$$

有效态密度为:

$$\rho_{0D}(E) = \sum_{n,m,l} \delta(E - E_x^n - E_y^m - E_z^l) \quad (1.10)$$

其中 E_x^n, E_y^m, E_z^l 为箱型量子点在 x, y 和 z 方向的量子化能级。量子化能

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