1.0 eV GaAs 基 InAs 量子点太阳能电池

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摘要 三结InGaP/GaAs/Ge太阳能电池理论设计中加入带隙为1.0 eV的材料代替带隙为1.4 eV的GaAs中间电池有助于解决多结串联电池的电流阻塞效应实现电流匹配, 然而带隙为1.0 eV的InGaAs和GaInNAs外延困难. 我们利用分子束外延方法外延得到In_{0.15}Ga_{0.85}As量子阱, InAs dots-in-well量子点以调整太阳能电池带隙. X射线衍射谱中观察到了量子阱的多级卫星衍射峰, 量子阱界面陡峭. 扫描透射图显示量子点呈金字塔状, 量子点的高度约为12 nm, 底边长约为27 nm. 由原子力显微镜图可知, 量子点密度约为 $2 \times 10^{10} \text{ cm}^{-2}$. 低温光致发光谱显示量子点在As₄束流下呈双模分布. 光电流响应谱显示InAs dots-in-well量子点太阳能电池吸收波长可以达到1300 nm, 相应于带隙约为1.0 eV. 器件J-V特性显示短路电流相比于GaAs标准p-i-n电池增加了37.8%. 这表明该InAs dots-in-well量子点太阳能电池有望改善多结太阳能电池的设计中的电流阻塞效应, 实现电流匹配, 在多结太阳能电池的设计中具有广阔的应用前景.

关键词 量子点, III-V半导体, 太阳能电池

近年来, GaAs基单节太阳能电池和多节太阳能电池广泛应用于空间电池技术和制造聚光太阳能电池^[1-5]. 最常用的三结太阳能电池多采用Ge作为底电池, GaAs为中间电池, InGaP为顶电池, 带隙分别为0.7, 1.4, 1.9 eV^[6,7]. 在三结太阳能电池中, GaAs结电池短路电流密度低于其他两结电池. 而三结电池的短路电流取决于子电池短路电流的最小值, 因此, GaAs结电池限制了三结太阳能电池转换效率的提高^[8].

为了扩展GaAs太阳能电池的吸收波长, 国内外学者提出在GaAs p-i-n电池的耗尽区加入量子阱结构, 应力平衡, 有效地扩展了电池的吸收波长^[9]. 根据理论计算, 在InGaP/GaAs/Ge多结电池中串联一个带隙为1.0 eV的电池替代GaAs子电池会扩展光谱

吸收增大电池效率^[10]. 常用的带隙为1.0 eV的材料有晶格匹配的GaInNAs和晶格失配的In_{0.3}Ga_{0.7}As^[11,12]. GaInNAs少子寿命短, 载流子迁移率低, 少子扩散长度小于耗尽区宽度^[13,14]; In_{0.3}Ga_{0.7}As与Ge晶格失配大, 大尺寸的异质外延很困难^[8]. 改善晶体外延质量和设计新结构的太阳能电池是提高光电转换效率的方法.

引入应力平衡的低维半导体结构, 如量子阱和量子点结构, 不仅提高了异质外延的晶体质量, 而且可以通过调节势阱带隙来调节电池的光谱吸收, 进而提高电池的光电转换效率^[15-18]. 多结太阳能电池为串联结构, 其开路电压为各子电池开路电压之和. 因此, 尽管基于低维半导体材料的太阳能电池存在降低的开路电压的缺陷, 但在多结电池串联应用中,

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并不影响多结电池的整体效率^[19]. 另外, 量子阱和量子点太阳能电池显示出优越的抗辐照性能和较高的温度系数, 在空间技术方面具有应用潜力^[20-22]. 目前, 可以通过调整生长参数来获得光致发光峰在1300 nm的InAs量子点材料, 该波长的材料主要应用于激光器的制备, 关于将该种材料应用于太阳能电池鲜有报道^[23,24].

本文研究了InGaAs量子阱和In_{0.15}Ga_{0.85}As量子阱中InAs量子点在1.0 eV电池中的适用性. 通过外延得到包含该结构的GaAs基太阳能电池, 并测试其光伏性能, 发现该结构的太阳能电池吸收波长扩展到1300 nm, 接近带隙1.0 eV, 短路电流相比于GaAs电池提高了36.7%.

1 实验

本实验的3个样品通过分子束外延(MBE)方法,

在p型GaAs(100)衬底上外延生长制得. 样品结构如图1所示, 样品A是一个标准的GaAs单结pin结构太阳能电池: p区欧姆接触层厚度为300 nm, 掺杂浓度为 $N_A=3\times10^{18}\text{ cm}^{-3}$, 基区厚度为300 nm, Be掺杂浓度为 $N_A=5\times10^{17}\text{ cm}^{-3}$, 本征区GaAs厚度为630 nm, 发射区厚度为150 nm, Si掺杂浓度为 $N_D=5\times10^{17}\text{ cm}^{-3}$, n区欧姆接触层厚度为300 nm, $N_D=3\times10^{18}\text{ cm}^{-3}$. 样品B和样品C只是本征区结构与样品A不同, 电极结构相同, 厚度相同. 样品B的厚度为630 nm, 10个8 nm的In_{0.15}Ga_{0.85}As量子阱结构均匀地穿插在11个50 nm GaAs隔离层中间, 而样品C中是在样品B中的In_{0.15}Ga_{0.85}As量子阱中插入2.2 ML(monolayer)的InAs量子点, 即在2 nm的In_{0.15}Ga_{0.85}As上生长2.2 ML的InAs量子点最后覆盖6 nm In_{0.15}Ga_{0.85}As, 本征区厚度也为630 nm. 在生长InAs dots-in-well结构时, 生长完InAs量子点后中断30 s, 促进In原子的迁移. 生长

图1 (网络版彩色)样品的结构和材料形貌表征图. (a) 器件结构: 标准GaAs太阳能电池(样品A), InGaAs/GaAs量子阱太阳能电池(样品B), InAs dots-in-well量子点太阳能电池(样品C); (b) InAs dots-in-well 太阳能电池和InGaAs/GaAs量子阱太阳能电池的X射线衍射图谱(004); (c) InAs 量子点的原子力显微镜(AFM); (d) InAs dots-in-well 太阳能电池的扫描隧道显微镜

Figure 1 (Color online) The structure of the sample and the characterization of the material. (a) Device architecture: standard GaAs solar cell (sample A), InGaAs/GaAs quantum well solar cell (sample B) and InAs/InGaAs quantum dots solar cell (sample C); (b) X-ray diffraction (w/2θ) spectra taken around the (004) reflection of InAs dots-in-well solar cell and InGaAs/GaAs quantum well solar cell; (c) atomic force microscope (AFM) image of the InAs quantum dots; (d) scanning transmission-electron microscopy (STEM) image of the InAs/InGaAs quantum dots solar cell

时As₄压保持在 1×10^{-5} Torr(1 Torr $\approx$ 133.322 Pa), GaAs生长时衬底温度保持在580℃, 生长含In组分的区域时衬底温度保持在500℃. 在样品B和样品C中生长耗尽区的GaAs势垒时, 前10 nm低温生长(500℃), 后40 nm高温生长(580℃).

外延生长完成之后, 利用常规光刻剥离流程制备出上电极图形, 通过电子束蒸发的方法, 蒸镀Ni/Au/Ge/Ni/Au(15/101/26/26/100 nm)到上电极和Ti/Au(20/100 nm)到下电极. 电池器件大小是0.5 cm $\times$ 0.5 cm. 上电极占据单个器件面积的6%. 未制作增反膜.

利用X射线衍射来观察外延片的晶体质量, 样品B和样品C的XRD衍射如图1(b)所示. 利用原子力显微镜(AFM)观察量子点表面形貌, 如图1(c)所示. 扫描投射显微镜观察InAs量子点dots-in-well结构, 如图1(d)所示.

光电流响应谱(PC)是由卓立汉光DSR100系统测得, 光致发光谱由自行搭建设备测量. 太阳能电池的光伏特性由Keithley 4200测试得出.

2 实验结果与讨论

通过改变低维纳米材料的组分和大小可以裁剪材料的带隙, 从而调整器件的光电特性. 图1(b)显示了样品InGaAs/GaAs量子阱太阳能电池(样品B)和InAs dots-in-well量子点太阳能电池(样品C)的XRD图, 从图中可以看出样品B的衍射卫星峰清晰, 强度高, 多周期样品结构完整, 界面清晰. 说明晶体质量较高, 缺陷少; 样品C的衍射峰模糊数量少, 强度低, 说明插入InAs量子点后界面粗糙度有所提高. 从图1(c)量子点的AFM图中可以看出量子点密度为 2×10^{10} cm⁻². 从图1(d)中量子点的STEM图中也可以看出金字塔型量子点高度约为12 nm, 底边长27 nm.

图2是样品C在50 K时, 在激发功率20 mW的532 nm激光照射下的光致发光谱(PL). 从图中看出量子点呈现非对称的双峰分布, 大的量子点的峰位在1243 nm, 小的量子点的峰位在1195 nm. GaAs衬底峰在850 nm.

图3上部分是3个样品的光电流响应谱(PC), 下部分是InGaAs量子阱太阳能电池和InAs dots-in-well量子点太阳能电池在220 K时的PL谱当入射光波长从500 nm变化到1400 nm时, 标准GaAs太阳能电池的光响应从600 nm开始上升, 860 nm光电流达到最大, 然后迅速降低, 超过900 nm几乎没有响应. InGaAs量

图2 (网络版彩色) InAs dots-in-well 结构在 50 K 时的光致发光谱
Figure 2 (Color online) Photoluminescence (PL) results for the 10-layers of InAs/InGaAs QDs solar cell (sample C) at the temperature of 50 K

图3 (网络版彩色) 3 种结构的太阳能电池的光电特性. (a) GaAs 标准电池、InGaAs 量子阱电池、InAs dots-in-well 量子点电池的光电流响应谱; (b) 220 K 下 InGaAs 量子阱和 InAs dots-in-well 结构的光致发光谱

Figure 3 (Color online) The optical-electrical characteristic of the three types of solar cells. (a) Photocurrent response of GaAs reference solar cell (sample A), InGaAs quantum well solar cell (sample B) and InAs dots-in-well solar cell (sample C); (b) the PL of quantum well solar cell (sample B) at room temperature and InAs dots-in-well solar cell (sample C) at 220 K

子阱太阳能电池的光响应从600 nm开始上升, 到达990 nm后迅速下降, 对应于其在220 K时的荧光峰. InAs dots-in-well量子点太阳能电池的光响应从650 nm开始上升, 到880 nm达到最大, 然后开始缓慢下降, 一直到1300 nm以后光电响应才截止. 从图3中样品的PC谱可以看出, 随着低维半导体材料, 量子阱量子点的加入可以有效地调节光谱吸收波长. InAs dots-in-well结构量子点吸收波长可以扩展到1300 nm, 对应于1.0 eV带隙. 因此本文中调制得到的InAs

dots-in-well结构的太阳能电池在多节太阳能电池的设计中有广阔的应用前景。

图4是GaAs标准电池、InGaAs量子阱、InAs dots-in-well量子点电池的 J - V 特性曲线。表1是上述3种电池的 J - V 特性表。由 J - V 特性曲线看出,在短路电流方面,InGaAs量子阱电池比GaAs标准p-i-n太阳能电池高0.784 mA,增加了7.9%。InAs dots-in-well量子点电池比InGaAs量子阱电池高2.97 mA,比GaAs标准p-i-n太阳能电池高3.754 mA,增加了37.8%。量子点的加入,降低了电池的带隙,增加了近红外波段的光吸收,短路电流增加。在开路电压方面,InGaAs量子阱电池比GaAs标准p-i-n太阳能电池低102 mV,InAs dots-in-well量子点电池比GaAs标准p-i-n太阳能电池低651 mV。量子阱和量子点的加入,使得电池的带隙降低,降低了开路电压。并且在InAs dots-in-well太阳能电池中,量子点的加入引入了大量的位错缺陷,使晶体质量变差,复合中心增多,开路电压降低,光电转换效率降低。

对于III-V族化合物电池,单片多结电池通常是在底电池上直接外延生长下一个子电池,中间通过隧穿结连接,电池的开路电压是各个子电池开路电压之和,而短路电流却等于子电池最小的短路电流。因此,尽管基于低维半导体材料的太阳能电池的效率退化机制和抑制方式仍有待进一步研究,但通过设计加入量子阱和量子点等低维半导体材料,增大了短路电流,缓解多结电池的电流阻塞效应。因此,在GaAs p-i-n子电池中引入低维纳米结构在少量增加传统三结太阳能电池的开路电压基础上,显著提高电池的短路电流,从而提升系统效率。

3 结论

本文设计了 $\text{In}_{0.15}\text{Ga}_{0.85}\text{As}$ 量子阱、InAs dots-in-well量子点太阳能电池。高分辨X射线衍射观察到了多量子阱中的多级卫星峰,表明量子阱界面陡峭。通过原子力显微镜和扫描透射显微镜观察到了量子点的形貌和高度,量子点的插入引进了大量的位错缺

图4 (网络版彩色)太阳能电池的 J - V 特性曲线。标准GaAs太阳能电池、InGaAs量子阱太阳能、InAs dots-in-well量子点太阳能电池

Figure 4 (Color online) J - V Characteristics for the InGaAs quantum well solar cell, InAs dots-in-well solar cell and GaAs reference p-i-n solar cell under 1.5 AM illumination

表1 太阳能电池大的 J - V 特性

Table 1 Device parameters for three solar cells in study

Sample	V_{oc} (mV)	J_{sc} (mA/cm ²)	Fill factor(%)	η (%)
A	215	13.68	32.7	0.94
B	768	10.71	72.3	5.95
C	866	9.926	76.4	6.57

陷,导致复合中心增多,降低晶体质量,引起室温光致发光淬灭。低温光致发光谱显示量子点在 As_4 束流下呈现双模分布。3种电池的光电流响应谱显示量子阱和量子点的插入可以调整带隙,有效增加近红外波段的光吸收,其中InAs dots-in-well量子点太阳能电池可以调整带隙到约1.0 eV,吸收波长到1300 nm,电池的 J - V 特性曲线显示InAs dots-in-well量子点太阳能电池短路电流相比于GaAs标准p-i-n电池增加了37.8%。相比于具有1.0 eV带隙的 $\text{In}_{0.3}\text{Ga}_{0.7}\text{As}$ 和GaInNAs, InAs dots-in-well量子点结构更易生长。这表明该InAs dots-in-well量子点太阳能电池有望改善多结太阳能电池的设计中的电流阻塞效应,在多结太阳能电池的设计中具有广阔的应用前景。

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Summary for “1.0 eV GaAs 基 InAs 量子点太阳能电池”

1.0 eV GaAs based InAs quantum dot solar cells

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There are great improvements in III-V semiconductor solar cells including GaAs single junction solar cell and tandem solar cells in last decades. The common commercially used triple-junction solar cell mostly utilize a Ge bottom cell with an (In)GaAs and InGaP middle and top cell respectively. The (In)GaAs middle cell absorbs limited solar spectrum and show minimum short-circuit current in multiple junction solar cell, which determine the overall current of the cell. Theoretically, higher energy conversion efficiency will be obtained when introduce a subcell with 1.0 eV band gap in series to the InGaP/GaAs/Ge tandem structures. Lattice-matched GaInNAs and lattice-mismatched In_{0.3}Ga_{0.7}As are two common choices for achieving 1.0 eV band gap. However, the addition of N to GaAs has detrimental effects on the material quality, limiting its utility to approaches that required photocurrent. And the lattice mismatch makes thick In_{0.3}Ga_{0.7}As heterogeneous epitaxial difficult. The incorporation of low band gap nanoscale materials, such as quantum well and quantum dots, into current limiting junction of a multijunction solar cell can tuning the effective bandgap and enhance the low-energy photon absorption, thereby increasing the short-circuit current.

The samples are grown by solid-source molecular beam epitaxy (V80H) on p-GaAs (001) substrates using an As₄ source. Sample A is a standard GaAs solar cell with PIN structure. Sample B contains 10 period 8 nm In_{0.15}Ga_{0.85}As QW and sample C contains 10 period InAs dots-in-well structure. The intrinsic region of these samples was at the same length of 630 nm to obtain the same build-in electric field. Besides the intrinsic region, the three samples are designed the same. The ohmic contact layer's doping concentration is $N_D/N_A=3\times10^{18}\text{ cm}^{-3}$ and its thickness is 300 nm. The emitter doping concentration is $N_D=5\times10^{17}\text{ cm}^{-3}$ and its thickness is 150 nm; The base doping concentration is $N_A=5\times10^{17}\text{ cm}^{-3}$ and its thickness is 300 nm. The As₄ pressure and the InAs QDs growth rate are $1\times10^{-5}\text{ Torr}$ and 0.1 ML/s, respectively.

The XRD pattern of sample B (QWSC) shows clearly defined and intense satellite peaks indicating steep interface and periodicity quantum well. However, the XRD pattern of sample C (the InAs dots in well solar cell) shows blurring diffraction peaks which means increased interface roughness. Uniform QD structure with density of $2\times10^{10}\text{ cm}^{-2}$ can be observed in SEM. From the STEM, we can see well-defined unrelated pyramidal quantum dots separated by a nominal 50 nm GaAs spacing. From the contrast, the pyramid InAs QDs are estimated to have the lateral dimension of 27 nm and the height of 12 nm. The PC response of sample A rise at 600 nm and fall off at 870 nm, which is corresponding to the GaAs bandgap according to the PL peak position; the PC response of QWSC (sample B) rise at 600 nm and fall off at 990 nm, which is corresponding to the In_{0.15}Ga_{0.85}As bandgap in PL spectrum. The incorporation of In_{0.15}Ga_{0.85}As quantum wells in GaAs standard solar extend the absorption of solar spectrum from 900 nm to 1000 nm. The wavelength region from 700 nm to 900 nm in PC of QWSC is weaker than the GaAs standard solar cell. The insertion of InAs QDs into QWSC can extend the infrared photon absorption to the wavelength of 1300 nm. The insertion of InAs quantum dots into In_{0.15}Ga_{0.85}As/GaAs quantum well solar cell extend the absorption of infrared light and the J_{sc} reaches 13.68 mA/cm², giving a 37.8% increase in short-circuit current over the baseline solar cell current. Because the voltage of multiple junction solar cell is the sum of the sub cell's voltage. The voltage decrease with the insertion of InAs quantum dots do not influence the overall voltage of multiple junction solar cell.

In summary, the incorporation of quantum well and quantum dots in GaAs solar cell result in extended solar spectrum absorption and contribute to the sub-band gap current gains for GaAs solar cell. Especially, InAs dots-in-well solar cell can extend the absorption spectrum to the wavelength of 1300 nm, and increase by 37.8% in short-circuit current, which shows great potential in mitigating current mismatch of multiple junction solar cell. Furthermore, designing new multiple junction solar cell becomes possible.

quantum dots, III-V semiconductor, solar cells

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