

中国激光

基于纳米连接的互连结构的电/力学性能研究进展

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摘要 纳米连接技术是纳米元器件与微系统及宏观系统整合的关键技术之一。稳定的器件性能取决于可靠的纳米互连结构, 评估纳米互连结构的力学性能及电学性能对于预测电子器件的失效模式至关重要。本文结合目前不同纳米连接技术及连接界面的特点, 对不同材料从单纳米焊接接头到宏观互连结构的电/力学性能特征进行了总结与展望, 通过对互连结构焊接及变形机制、焊接强度、疲劳特性及电学性能的探讨, 展现了激光诱导等离子体自限性低温焊接在未来纳米器件及柔性电子器件制造中的巨大潜力。

关键词 光学制造; 纳米连接; 冷焊; 激光诱导等离子体; 电学性能; 力学性能

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1 引言

随着新型电子器件向微型化、柔性化和智能化方向发展, 高速发展的信息工业对功能电子器件集成度的要求越来越高, 促使人们不断探索能够突破器件尺寸极限的途径, 相关的制造技术更是逐渐发展到了纳米尺度^[1-4]。纳米制造是全球制造技术竞争的制高点, 在《中国制造 2025》战略规划中, 我国也将纳米制造技术纳为重点发展战略。纳米连接技术作为纳米制造的关键技术, 是纳米元器件与微系统及宏观系统整合的基础, 在新一代电子器件制造中扮演越来越重要的角色。

纳米材料的尺寸效应和高的比表面积使其在连接过程中表现出与块状材料不同的焊接特征。对于具有高电导率及热导率的金属纳米材料(如 Ag、Au、Cu 等)、高热/力/电学性能的碳基纳米材料(如碳纳米管、石墨烯等)以及部分宽带隙半导体纳米材料(如 ZnO 等)来说, 在对其进行互连封装时, 高质量的纳米互连结构是使器件获得优异性能的基础,

获得可靠的纳米互连结构以及对互连接头的电/力学性能进行研究, 对于促进新一代电子器件的技术升级及预测电子器件的失效模式至关重要。本文针对目前不同的纳米连接技术以及连接界面的特点, 结合互连接头电/力学性能的对比分析, 对激光诱导纳米互连技术在未来新一代电子器件制备中的应用进行总结和展望。

2 纳米连接技术及其表征

2.1 纳米连接技术

通常, 纳米连接的目的是在两个或更多单个组件之间建立牢固、稳定的化学键连接, 保证焊接界面具有优异的电学性能和力学性能。在常温或较低温度下, 纳米材料之间会在范德瓦尔斯力作用下相互靠近并接触, 然后通过高活性的表面原子扩散行为实现物质的传输, 进而达到连接的目的。然而, 通常情况下在纳米材料表面会附着制备过程中残留的杂质和分散介质, 这不仅会降低原子或分子之间直接接触的可能性, 还会阻碍接触区域原子的扩散行为。

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为了实现纳米材料的连接,通常需要适当加载外部作用或能量,如压力、热能等,促使纳米材料之间原子的接触与扩散^[5]。

在进行纳米连接时,根据纳米材料互连时的状态可将纳米连接分为纳米固相连接和熔融连接。固相连接是指在焊接过程中不存在熔融现象的连接技术,其连接方法包括自发冷焊^[6-8]、压焊^[9-10]等。熔融连接的方法包括高温退火^[11]、焦耳热熔连接^[12]、电子束/离子束等高能束熔接^[13-15]、长脉冲激光烧蚀^[4,16-22]、光诱导表面等离子体增强自限性焊接^[23-25]等。纳米固相冷焊形成

的接头主要是随着纳米材料表面能升高以及接触表面原子扩散形成的,因此在焊接过程中要求待焊接表面尽量无杂质残留,同时具有较高的表面能。但该方法的互连尺寸具有一定的局限性。对于金属来说,低温冷焊形成的界面特征在焊接初期会出现大量位错缺陷和晶向不匹配,但随着焊接过程的进行,最终会得到近乎完美的单晶纳米结构^[8,26]。通过高温熔融得到的纳米接头的热影响区通常较大,这会对非连接部位的结构产生热影响甚至是破坏。图 1 所示为不同连接方法获得的金属 Ag 纳米线互连接头。

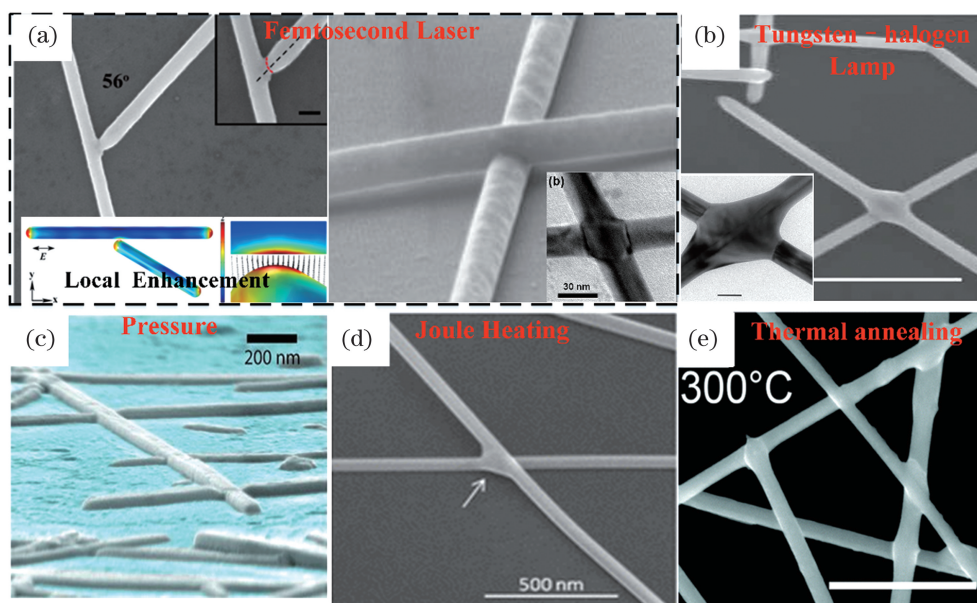


图 1 不同连接方法获得的 Ag 纳米线互连接头。(a)飞秒激光诱导纳米连接^[23-25]; (b)卤钨灯辐照^[16]; (c)低温压焊^[27]; (d)焦耳热熔连接^[12]; (e)高温退火^[11]

Fig. 1 Nanojoint of Ag nanowires obtained by different nanowelding methods. (a) Femtosecond laser induced nanowelding^[23-25]; (b) irradiation by halogen tungsten lamps^[16]; (c) low temperature pressure welding^[27]; (d) Joule heating jointing^[12]; (e) high temperature annealing^[11]

相对于其他焊接技术,通过调控光参量可以有效激发材料表面自由电子的集体振荡,具有显著的近场局域增强^[28-29]和纳米聚焦^[30-31]等特性,对非焊接部位的影响较小,能够在互连耦合间隙诱导纳米材料产生自限性低温焊接。相对于普通的光源,例如卤钨灯,激光能够激发材料表面产生更强的表面等离子体局域场增强效应,可以通过空间调制(例如激光偏振状态、能量分布等)对焊接界面进行精准调控,同时能够实现间隙(无接触)空间分布纳米材料的局域焊接。尤其是超短脉冲激光,当其脉冲宽度比电子冷却时间短时^[32],可以有效实现冷加工和冷焊接,其极高的单脉冲能量和局域强电场可以有效激发金属材料甚至是

一些键能较高的材料表面产生空键,从而形成互连结构。

2.2 纳米连接接头的电学性能表征

纳米连接接头的电学性能测量包含对单纳米接头的测量和宏观器件的测量。目前,单纳米接头的主要测量方法分为两种,一种是通过微纳米探针操作装置对纳米结构进行直接接触测量,如图 2(a)所示,另一种是基于微电子机械系统(MEMS)利用微纳米电路获得相关的电学信息,如图 2(b)所示。单纳米连接接头在电学测量过程中对引线接触电阻比较敏感,因此在测量过程中应保证接触点稳定连接。在进行原位单点电学测量时,一般会利用电子束或离子束对探针与纳米材料的接触点进行定点纳米电

极(例如 Pt 和 W 电极)沉积;而对于基于 MEMS 微电路的单点测量同样需要保证一定的接触面积,以确保电路的稳定性。对于单纳米接头,一般情况下会通过耦合扫描电镜或高倍光镜进行原位观测和测量。无论是单纳米接头,还是基于宏观互连的纳米

接头,在测量其微纳米表面电阻率时,一般会采用微纳米操作四探针测量技术,以消除引线电阻的影响。最常见的四探针测量技术分为共线探针测试法和范德堡法,其中范德堡法常被用于测量二维薄膜的电阻率。

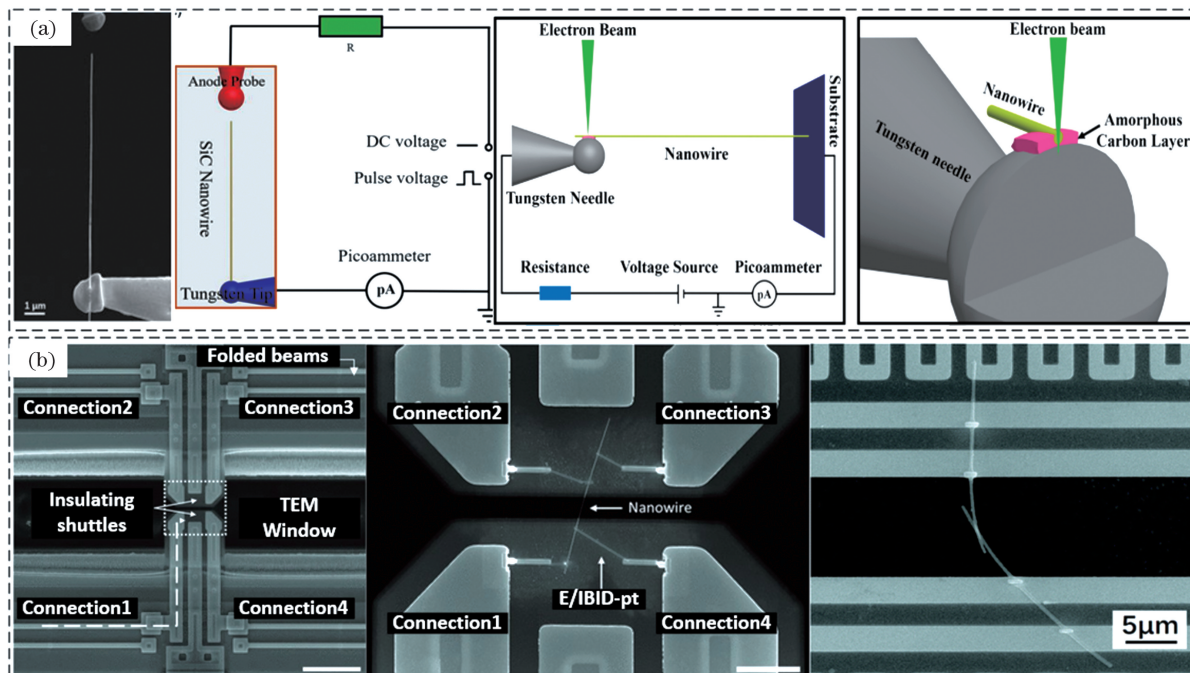


图 2 纳米互连结构的电学性能测试。(a)基于纳米操作探针的两点接触测量^[33]; (b)基于 MEMS 与扫描电镜的集成测量^[34-35]

Fig. 2 Electrical performance test of nano interconnection. (a) Two-point contact measurement based on nanometer operation probe^[33]; (b) integrated measurement based on MEMS and scanning electron microscopy^[34-35]

2.3 纳米连接接头的力学性能表征

目前,对于纳米连接接头力学性能表征的方法主要包含理论研究和实验测量两部分。理论研究大部分集中于对纳米互连界面的原子分布进行讨论分析,通过分子动力学等相关模拟软件对互连界面的力学性能进行模拟,对整个焊接、拉伸过程进行原子拟实,并对焊接界面形貌及其影响因素进行分析,获得焊接前后理论上的力学性能^[36-37]。因纳米连接的原子模拟受限于原子计算的规模尺度,因此大部分研究仍集中在直径为 20 nm 以下的一维纳米材料的互连。对于实验测量,同样包含对单纳米连接接头的测量与基于纳米连接的宏观器件的测量。随着原位表征技术的发展,目前已经可以实现对一维纳米线或二维纳米材料的直接力学拉伸。实验测量所用技术主要包括基于原子力显微镜(AFM)的直接测量技术^[38-40](如 AFM 弯曲^[41]和接触共振测试^[42])、聚焦离子束(FIB)与扫描电镜(SEM)^[43]及

透射电镜(TEM)芯片集成技术^[26]、MEMS 与电子显微镜集成技术等^[44]。基于 AFM 对纳米焊点的力学测量主要依赖于 AFM 对力-位移响应的极高分辨率,通过 AFM 探针与待测纳米材料的直接接触即可获得相关的力学信息。

近年来,随着基于 AFM 及 MEMS 的纳米操作系统在 SEM 及 TEM 内的集成,原位测量已成为研究纳米材料力学性能的理想方法,而 FIB 因可以实现对纳米材料的原位加工和操纵也被广泛应用于纳米力学试样的制备和观测。但在如此微观的尺度下对纳米焊接结构的力学性能进行直接表征,在实际操作过程中仍然面临着很多挑战,例如,测量过程中对不同工艺获得的焊接接头的转移与制样是研究人员面临的一大难题。对基于纳米互连的宏观柔性器件的力学测量,研究人员主要采用的是进行直接的疲劳强度测试和力学拉伸测试。图 3 所示为目前针对纳米连接接头力学性能进行测量的方法。

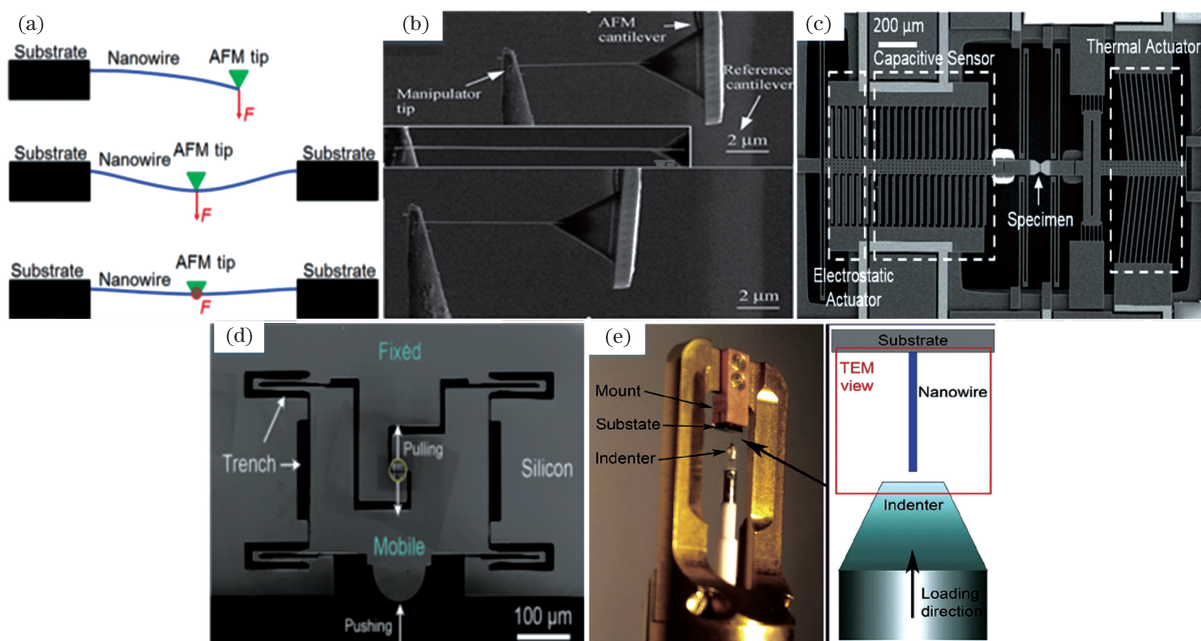


图 3 纳米互连接头力学性能的测量方法。(a)(b)基于原子力显微镜及光镜的测量^[38,45]；(c) MEMS 辅助测量^[46]；
(d)基于 TEM-SEM 芯片的测量^[47-48]；(e) TEM 原位集成测量^[26]

Fig. 3 Mechanical performance test of nano-interconnect structure. (a)(b) Measurement method based on AFM and optical microscope^[38,45]；(c) MEMS-assisted measurement^[46]；(d) measurement based on TEM-SEM chip^[47-48]；
(e) TEM-AFM *in-situ* integrated measurement^[26]

3 单纳米焊点的电/力学性能研究

目前,人们对于部分同/异质金属纳米材料以及半导体纳米材料之间单纳米焊点的电/力学性能的研究已经取得了一定进展,并且对于采用激光诱导等离子体低温焊接获得的单纳米连接接头的电学测量也获得了大量数据,但是对于激光互连单纳米焊点力学性能的直接测量获得的研究结果还相对较少,其原因还是受限于对激光诱导工艺和原位力学测量系统的耦合。由于单纳米焊点连接过程的时间和空间尺度与分子动力学能够描述的范围比较一致,因此除了利用原位测量方法对其力学性能进行直接测量外,有部分研究人员通过原子模拟的方式对纳米材料的互连与失效过程进行了拟实。模拟结果显示,焊接过程的温度越低,材料的直径越小,能够获得的焊接质量就越高^[49]。焊接参数及材料的初始形貌(包括焊接直径、焊接速度、温度和晶向)会对接头的质量产生直接影响^[50]。当焊接温度过高时,焊接界面会产生较大的堆垛层错能和残余应力,降低互连强度。与高温退火相比,激光诱导的焊接具有更少的无序原子、相变、位错以及更小的残余应力。合适的激光能量能够减少焊接时的位错与相变^[51],焊接强度和剪切强度会随着激光能量的增加

而增强,但随着激光能量的继续增加可能会导致焊接强度减弱。目前,利用原子模拟对冷焊^[50,52-53]和高温退火^[36,54-55]的纳米互连界面及接头进行模拟评价方面的研究已取得了重大进展,但对于激光诱导局域场增强自限性焊接的原子模拟,大部分还集中在作用材料对光子能量的吸收上,对于激光激发的表面等离子体效应引发的局域热点在原子尺度上的纳米互连行为演化方面的研究还不太成熟,虽然有少部分学者利用温度指数能量梯度将局域场增强效应添加到了激光的原子互连模拟中^[51],但这与实际场分布还是有所区别的,还需要进行进一步的研究和探索。

尽管目前对激光诱导等离子体激发的单纳米焊接接头的力学测量还需要进一步的研究,但是在激光互连过程中能够激发的等离子体局域自限性低温连接与低温冷焊具有一定的相似性,而原位低温自发冷焊的单纳米接头的力学测量已取得了一定进展;因此,本文将对两者接头界面的性能进行总体讨论。

3.1 同质金属纳米材料的互连

目前,人们在同质金属纳米材料互连结构的电/力学性能方面已经取得了一些重要进展。Lu 等^[7]发现,在直径为 3~10 nm 的超薄单晶 Au 纳米线之间可以实现单晶 Au 纳米冷焊工艺。他们利用扫描

隧道显微镜 (STM) 探针对纳米线焊接前后的电/力学性能进行了原位测量, 结果发现, 焊接后的纳米线与拉断前的纳米线的电阻几乎保持一致, 且纳米线在循环拉断-焊接的重复实验中表现出了稳定的电学性能。除了具有优异的电学性能外, 纳米线的力学测量结果显示, 断裂前 Au 纳米线的抗拉强度为 (600 ± 50) MPa, 断裂后的焊接强度高达 (580 ± 40) MPa (而宏观 Au 纳米材料的抗拉强度约为 100 MPa), 其在后续的循环加载实验中断裂于非焊接部位。纳米低温冷焊的力学测量结果已经证明纳米连接接头具有优异的电/力学性能, 为纳米材料的进一步工程应用提供了研究基础。

人们在后续的研究中发现, 激光诱导等离子体局域焊接不仅可以突破纳米冷焊的尺寸限制, 还能

够突破空间限制, 实现纳米材料之间的无接触焊接, 而且焊点展现出了较高的电/力学性能。这是因为当纳米线之间相距一定距离时, 局域等离子体增强效应及局域光热力的作用可以实现纳米线甚至是微米线之间的延展焊接。Yang 等^[56] 利用低能量密度的连续激光对间隙分布的直径为 $200 \sim 400$ nm 的 Ag 纳米线实现了焊接, 如图 4 所示。他们通过对单根纳米线进行聚焦离子束刻蚀构造出了可控的纳米间隙, 然后对比了纳米线在切断前后及原位焊接后的电学性能; 结果显示, 焊接后的 Ag 纳米线的电阻仅略高于切断前单根 Ag 纳米线的电阻, 搭接 Ag 纳米线在激光焊接后具有更稳定的接触电阻。因此, 激光诱导等离子体局域焊接技术可以被有效地应用于下一代纳米电子器件的构造和修复中。

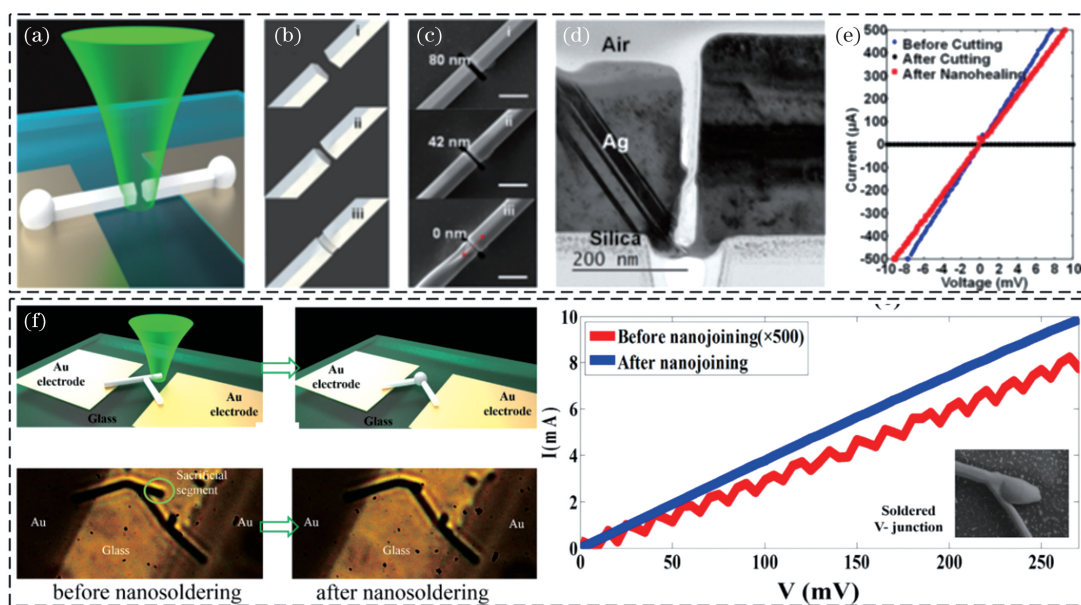


图 4 连续激光诱导 Ag-Ag 局域等离子体焊接^[56]。(a)~(d) Ag 纳米线缝隙原位焊接;(e) Ag 纳米线切断前后以及焊接后的电学性能;(f) 搭接 Ag 纳米线焊接前后的电学性能

Fig. 4 Ag-Ag local plasma welding induced by continuous laser^[56]. (a)~(d) *In-situ* welding of Ag nanowire gaps; (e) electrical properties of Ag nanowires before and after cutting and after nanoheating; (f) electrical properties of lap Ag nanowire before and after nanosoldering

3.2 异质金属纳米材料的互连

对于金属异质纳米焊点的电/力学性能进行研究, 有助于开发纳米尺度下器件的集成, 提高异种金属焊接的技术和工艺, 并加深对接头变形机理的认识, 进一步提高功能器件的集成度和可靠性。对于异质金属纳米接头的力学性能, 有学者认为同质纳米焊点的理论力学强度要比异质纳米焊点的强度高, 这是因为在焊接界面能够实现晶面匹配的焊接材料能够达到的理论力学强度要比初始结构差异较大的材料高^[53]; 但实际上, 对于一些晶面匹配度比

较高的异质金属纳米材料, 低温连接仍能够实现具有高强度和优良电学性能的纳米焊点。

对于激光诱导的金属异质互连结构的电/力学性能, 目前也取得了一些进展。Ghosh 等^[57] 利用波长为 532 nm 的连续激光将直径为 200 nm 的 Ag 纳米线焊接到 Au 电极上, 构造出了三维 Ag 纳米线拱桥结构, 如图 5(a) 所示。研究结果显示: 该结构具有稳定的电学性能, Ag 线与金属电极形成了稳定的通路, 互连后纳米材料之间的接触电阻大大降低, 而未互连相邻导线之间保持着绝缘状态; 同时, 焊接

处能够在重力为 2.6 pN 的 Au 纳米片作用下保持结构完整。可见,利用激光选区诱导构造三维纳米导电通路具有巨大的潜力。Lu 等^[7]对直径在 10 nm 以内 Ag 纳米线和 Au 纳米线进行了冷焊连接,如图 5(b)所示,他们在异质冷焊纳米互连接头的原位力学拉伸测量中发现,Ag-Au 异质界面可以实现良好的晶格匹配,断裂部位位于 Ag 纳米线的非焊接部位。Cihan 等^[58]曾基于原位透射电子显微

镜开展了 Al 和 Cu 纳米线的异质冷焊连接和 TEM 原位拉伸测试,如图 5(c)所示,他们发现:在焊接过程中,由于 Al 和 Cu 原子在较低的压力下发生了明显的相互扩散,因此焊接表面逐渐消失,形成了 Al/Cu 固溶体和 Al_3Cu_2 金属间化合物;该异质焊接接头具有超塑性,总延伸率甚至超过 100%,相应的塑性变形主要由位错的局部滑移和晶粒的旋转决定。

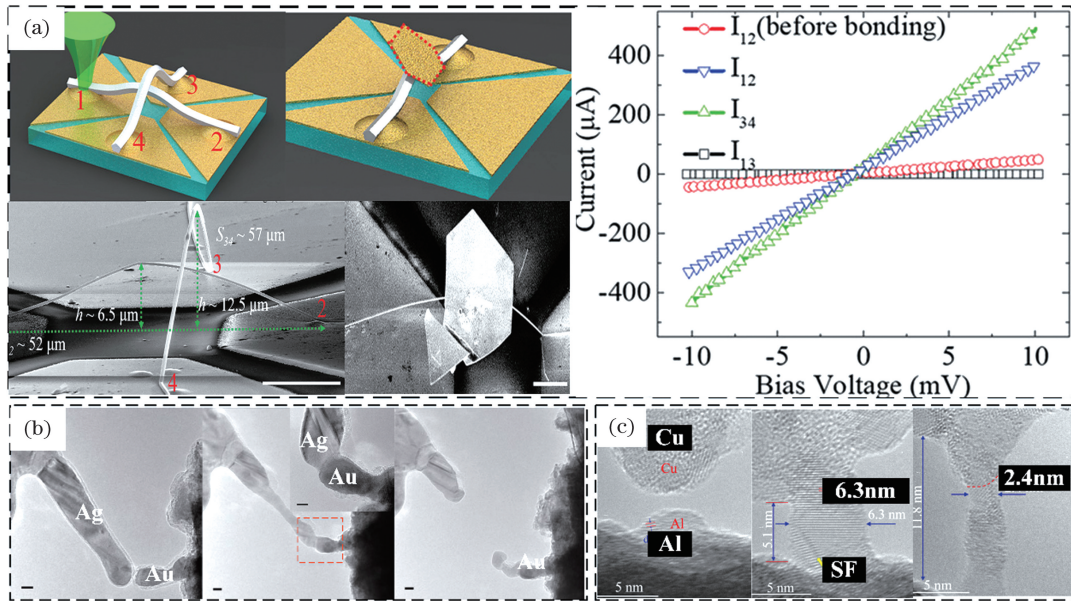


图 5 异质金属的纳米互连。(a)连续激光诱导连接 Ag 纳米线与 Au 电极构造三维拱桥结构及其电/力学性能^[57]; (b)超薄 Ag-Au 纳米线冷焊^[7]; (c)超薄 Cu-Al 纳米线冷焊^[58]

Fig. 5 Nano interconnection of heterogeneous metals. (a) Three-dimensional arch bridge structure with continuous laser-induced connection of Ag nanowires and Au electrodes and its electrical/mechanical properties^[57]; (b) ultrathin Ag-Au nanowires cold welding^[7]; (c) cold welding of ultra-thin Cu-Al nanowires^[58]

3.3 金属与半导体纳米材料的互连

对金属-半导体异质结的研究一直以来都备受催化、光电等领域研究人员的关注。先前研究人员主要采用化学气相沉积和液相外延等方法实现异质结的制备,如何高质量、高效率地实现纳米材料异质结近年来成为异质连接的热点。在构造微纳半导体功能器件时,尤其是在一些半导体材料和金属材料之间建立连接时,基于激光局域自限制的焊接能够精密调控焊接界面结构,促进了新一代传感功率电子器件的进一步发展。

Ghosh 等^[59]利用连续激光诱导构造了 ZnO 和 Ag 纳米线异质结,如图 6 所示。他们对器件的电学性能进行分析后发现:ZnO 纳米线与 Au 电极之间形成了类似肖特基界面的连接,而 Ag 纳米线与 Au 电极,以及 Ag 纳米线与 ZnO 之间更易形成欧姆连接;由 ZnO 和 Ag 纳米线及 Au 金属电极形成的器

件,在单个肖特基连接点处,能够实现 40 V 电压时响应电流最大达到 42 μA 。Lin 等^[60]利用飞秒激光脉冲实现了 Ag-TiO₂ 纳米线异质结,对其进行研究后发现:在光诱作用下,Ag 和 TiO₂ 的表面都因局域场增强效应而被修饰,TiO₂ 的润湿性增强,在纳米线耦合间隙处形成连接;在 Ag-TiO₂ 纳米线中分别产生了具有单极性和双极性的整流结,这些整流结具有不对称或对称的界面结构,为后面多材料微纳电子器件的组装和连接提供了支撑。

同时,部分学者对半导体纳米材料及金属电极的激光互连特性也进行了研究。Xing 等^[61]通过调控飞秒激光参数实现了对 Au/SiO₂/SiC 异质焊接界面的精准调控,从而构造出了三端器件,如图 7(a)~(d)所示;他们通过激光在 SiO₂ 层产生的局域场增强效应实现了 SiO₂ 层的选择性减薄,同时获得了表面覆有 SiO₂ 层的 SiC 纳米线与 Au 电极

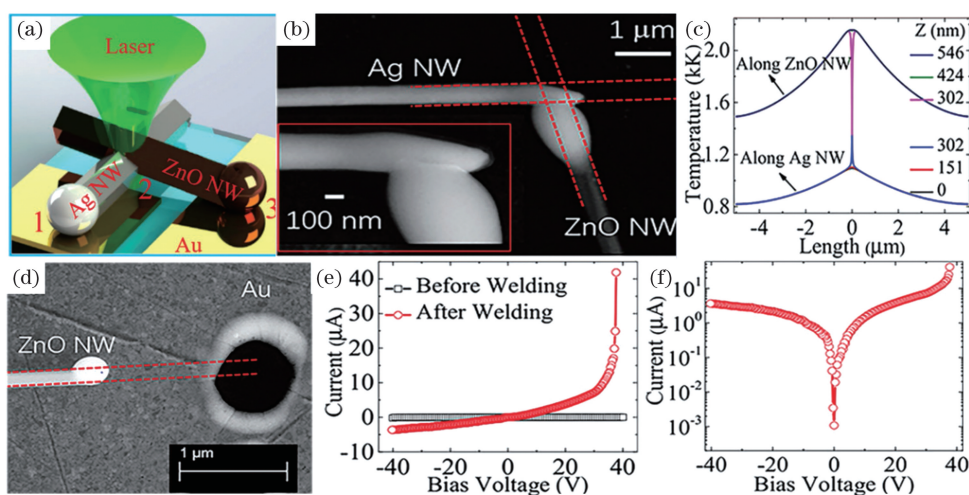


图 6 激光诱导 ZnO 与 Ag 纳米线互连^[59]。(a) 示意图；(b) 单个焊点的形貌；(c) 纳米线的温度分布；(d) ZnO 与 Au 电极的焊接形貌；(e) 焊接前后的 I - V (电流-电压) 曲线；(f) 电流-电压的半对数特征曲线

Fig. 6 Laser-induced nanowelding between ZnO and Ag nanowires^[59]. (a) Schematic; (b) morphology of single welded junction; (c) temperature distribution of nanowire; (d) welding image of ZnO and Au electrodes; (e) I - V (current-voltage) curves before and after welding; (f) semilog plot of current-voltage characteristic curve

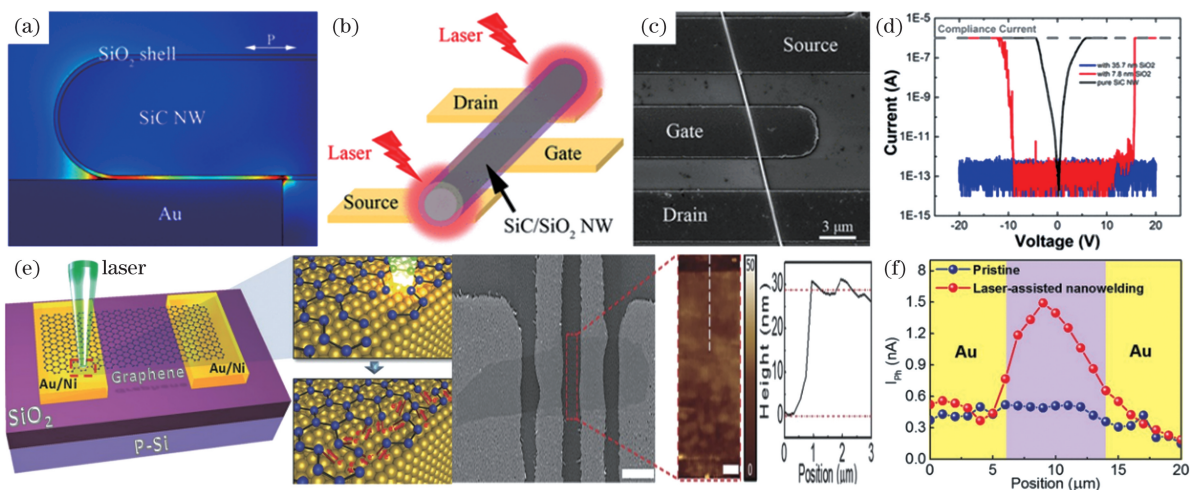


图 7 激光诱导金属电极与半导体纳米材料互连。(a)~(d) 飞秒激光诱导 Au/SiO₂/SiC 异质互连及三端器件的电学性能^[61]；(e)(f) 激光诱导金属电极与石墨烯异质互连及器件的光电性能^[63]

Fig. 7 Laser-induced nanowelding between metal electrodes and semiconductor nanomaterials. (a)–(d) Femtosecond laser-induced Au/SiO₂/SiC heterogeneous nanojoint and electrical characteristics of three-terminal devices^[61]; (e)(f) laser-induced heterogeneous nanojoint between metal electrodes and graphene and its photoelectric performance^[63]

的互连结构。同时,他们对获得的异质焊接界面进行了电/力学测量分析,结果发现,激光辐照后可以将器件的整流开关电压由 15.7 V 降到 1 V,同时该器件具有较低的漏电流。Xiao 等^[62]通过飞秒激光对 Ag-CuO 纳米线形成的金属 P 型半导体异质结的电接触性质进行了调控,并在激光诱导下通过界面处的 Ag(111)与 CuO(111)平面的晶格匹配,消除了肖特基势垒,实现了界面的欧姆接触。随着辐照时间的增加,由 Ag-CuO 纳米线异质结制成的纳米线器件从激光加工之前的双肖特基特性过渡到了整流特性。

对于一些碳基材料,例如石墨烯,其器件的电气性能在很大程度上受制于石墨烯与金属材料互连处接触电阻的限制。为了提高石墨烯的化学反应活性,促进石墨烯与金属的键合,从而最大限度地增加界面载流子输运,Keramatnejad 等^[63]采用波长为 524 nm 的连续激光照射石墨烯与金属接触区域边缘,实现了碳碳键的选择性击穿和结构缺陷的形成,如图 7(e)、(f)所示。这种方法显著降低了界面接触电阻,电阻率达到 $2.57 \Omega \cdot \mu\text{m}$ 。与未经激光处理的石墨烯/金属界面相比,激光处理后的石墨烯光电

探测器的光电流增加了 4 倍。这种无接触且高效的激光选区处理方法最大限度地保持了石墨烯器件的优越性能,使其在石墨烯器件的制造中具有无限潜力。也有学者^[64]利用飞秒激光实现了碳纳米管与金属电极的欧姆接触,说明飞秒激光可以实现对金属-半导体纳米级异质结电学性能的有效调控。

4 基于纳米连接的网络结构的电/力学性能研究

近年来,随着智能触摸交互终端、可穿戴电子设备、柔性太阳能电池等行业的快速发展,对柔性器件质轻、柔韧、高导电、大幅面和低成本等性能提出了更高要求。基于纳米互连网络的技术吸引着越来越多研究人员的关注。为了进一步推进纳米材料的工程应用,实现纳米材料性能的宏观化表征,大部分研究人员通过构造基于纳米互连的网络结构来实现功能器件性能的提高。

4.1 金属互连网络

利用激光焊接构造金属纳米互连网络通常有两种方法,一种方法是基于激光在金属离子前驱液中直接诱导光还原反应合成纳米材料,然后原位焊接金属

纳米结构^[5,20,65],另一种方法是已合成材料的图案化及大面积焊接^[66-70]。对于前者,焊接线宽的精度主要取决于激光光斑尺寸和能量的影响,而后者除了受激光参量的影响外,还受纳米材料自身形貌的调控。

目前,基于金属纳米线的互连网络被认为是制备大规模柔性透明电极的理想材料,尤其是 Ag 纳米线和 Cu 纳米线,被认为是制备下一代柔性电极极具潜力的材料^[24]。Han 等^[71]利用圆偏振激光在室温条件下对涂布的 Cu 纳米线进行超快等离子体纳米焊接,并将其与整体加热退火的焊接方式进行了对比,图 8 所示的结果表明:在环境中退火更容易导致 Cu 纳米线氧化,从而大大降低了焊接后互连器件的电学性能,其薄膜电阻值高达 $10^5 \sim 10^6 \Omega/\text{sq}$;激光诱导的 Cu 纳米线互连网络可以使 Cu 的氧化最小化,薄膜电阻为 $20 \Omega/\text{sq}$;互连后的柔性电极具有更优异的抗疲劳特性,在 20000 次的弯曲实验中以及在在高折叠角度和应变下,薄膜依然能够保持稳定的电学性能。Lee 等^[72]采用激光在聚二甲硅氧烷(PDMS)基底上构造了 Ag 纳米线透明电极(实验中所用 Ag 纳米线最长为 $500 \mu\text{m}$),结果显示,焊接后柔性电极的薄膜电阻为 $48 \Omega/\text{sq}$,即使在

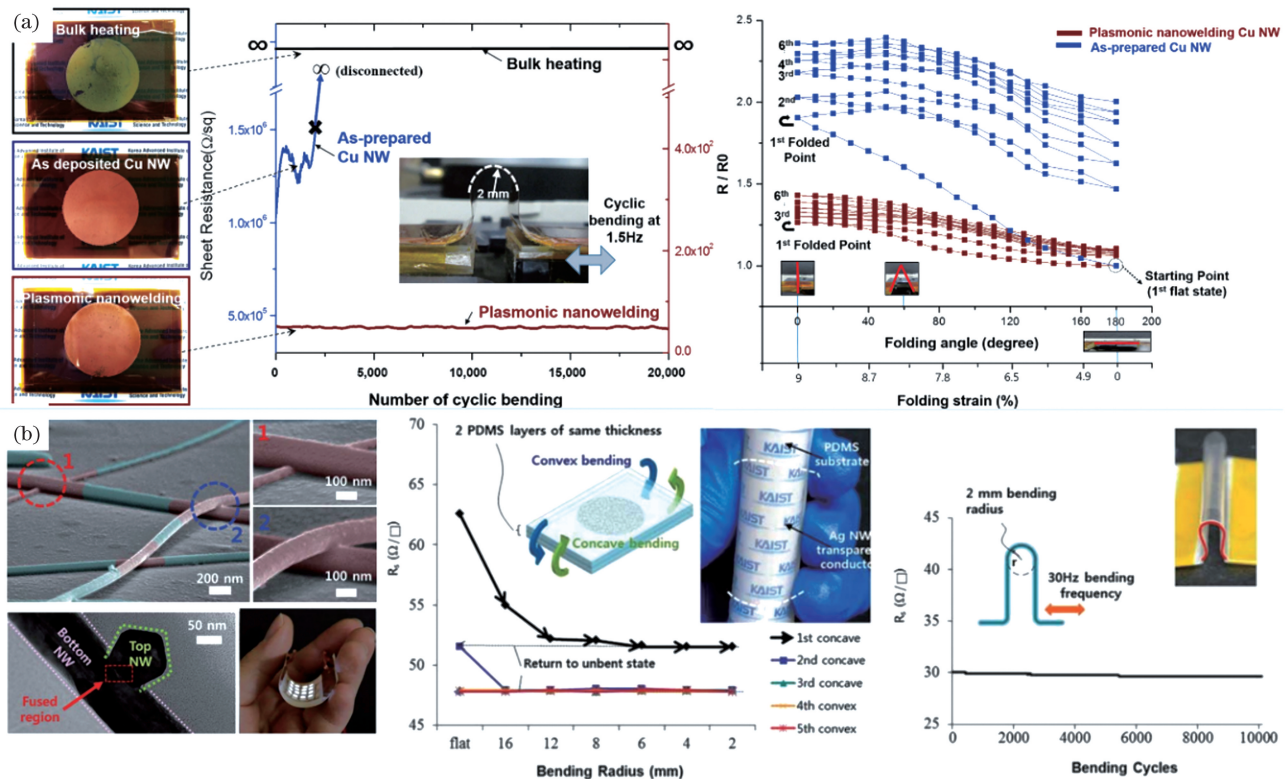


图 8 激光等离子体焊接柔性互连薄膜的电/力学性能^[71]。(a)互连 Cu 纳米线的抗疲劳性能测试;(b)激光诱导长 Ag 纳米线互连及其电/力学性能测试

Fig. 8 Electrical/mechanical properties of flexible interconnected thin films welded by laser plasma^[71]. (a) Fatigue resistance test of Cu nanowires; (b) laser induced long Ag nanowires interconnection and its electrical/mechanical performance testing

折叠半径为 2 mm,以 30 Hz 的频率加载 10000 次的情况下,薄膜电阻的变化也比较小,表现出了很强的电学稳定性。

除了实现纳米线材料自身之间的焊接外,有人发现光诱等离子体焊接也可以实现纳米线与柔性基底稳定可靠的连接。Park 等^[73]利用紫外灯照射 Ag 纳米线,实现了高电学性能互连网络(19 $\Omega/\text{sq}@90\%$ 透光率)。他们采用自行设计的双悬梁结构对 Ag 纳米线薄膜与基底开展了剥离实验,如图 9(d)所示,实验结果表明,剥离强度达到了焊接前的 310%。除了金属纳米线外,金属纳米颗粒的激光选

区烧结也被广泛应用于高性能柔性显示器件的制造。Kwon 等^[74]利用波长为 532 nm 的连续激光对 Cu 纳米颗粒进行选区烧结制造柔性显示器件,如图 9(a)~(c)所示,被辐照的 Cu 纳米颗粒的电阻率达到了 $1.67 \times 10^{-4} \Omega \cdot \text{m}$;之后他们对制造的结构进行循环弯曲测试(1000 次),测试过程中测得的电阻率变化为 $\pm 10\%$ 。之后,他们利用商用胶带对 Cu 纳米颗粒焊接结构与聚羧二甲酸乙二醇酯(PEN)基底的黏合力进行了测试,测试结果显示焊接结构具有良好的稳定性。将该结构作为触摸屏器件连接到控制器上,可以实现在柔性器件上的直接刻画输入。

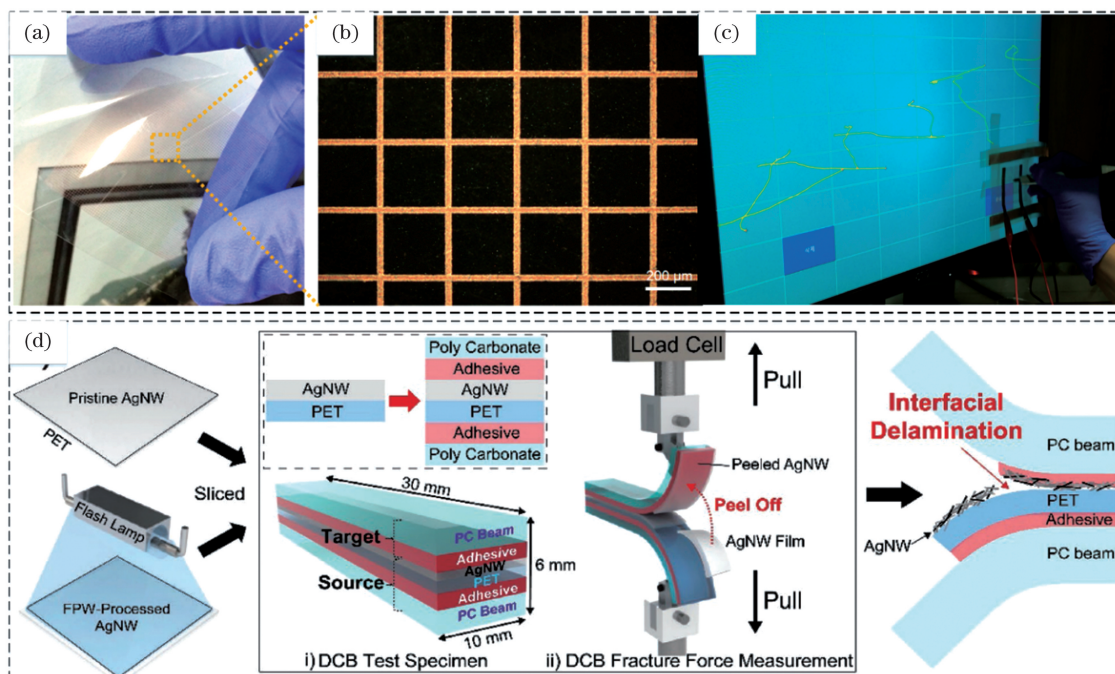


图 9 金属纳米互连网络的电/力学性能。(a)~(c)激光焊接 Cu 纳米颗粒构建柔性网格结构^[74]; (d)互连 Ag 纳米线网格与柔性基底剥离测试^[73]

Fig. 9 Electrical/mechanical performance of thin films based on metal nanowelding networks. (a)–(c) Laser-induced flexible mesh structure based on Cu nanoparticles^[74]; (d) stripping testing of interconnected Ag nanowires and flexible substrate^[73]

4.2 碳基纳米材料及半导体互连网络

碳基纳米材料作为新一代集成电路的重要组成部分,以其卓越的电/力学性能以及在应用上的无限潜力,激发着众多学者对其进行研究^[75-78]。In 等^[79]利用波长为 514 nm 的连续激光诱导垂直方向排列的碳纳米管与柔性聚合物进行连接,如图 10(a)、(b)所示,碳纳米管优异的光吸收效应导致其与聚合物板接触平面选择性加热,最大限度地降低了对聚合物的热损伤。碳纳米管的平均直径为 15 nm,密度为 $4.5 \times 10^9 \text{ cm}^{-1}$ 。In 等对互连结构进行疲劳拉伸力学测试后发现,其强度可以达到 2.8 MPa,最后断裂位置在碳纳米管的其他部位,证明了激光焊接技术能够实现碳纳米管与各种热塑性聚合物的有

效连接。Gong 等^[80]通过焦耳热焊接技术对碳纳米管进行了连接,结果发现碳纳米管表面的非晶碳有利于碳纳米管焊接过程的进行;同时,他们通过原位测量碳纳米管焊接前后的电导率变化发现,焊接后碳纳米管束的电导率提高了三个数量级,而且因焦耳热引起的温度变化与碳纳米管束电导率之间的关系符合自然对数规律。Liu 等^[81]利用连续光纤激光器对多壁碳纳米管进行了互连,如图 10(d)所示,他们认为碳纳米管在互连过程中的石墨化程度同样会极大地影响碳纳米管薄膜的电学性能。Mei 等^[82]通过调控飞秒激光能量实现了多壁碳纳米管之间的共价键连接,连接后的多壁碳纳米管互连网络薄膜的电学性能大大提高。

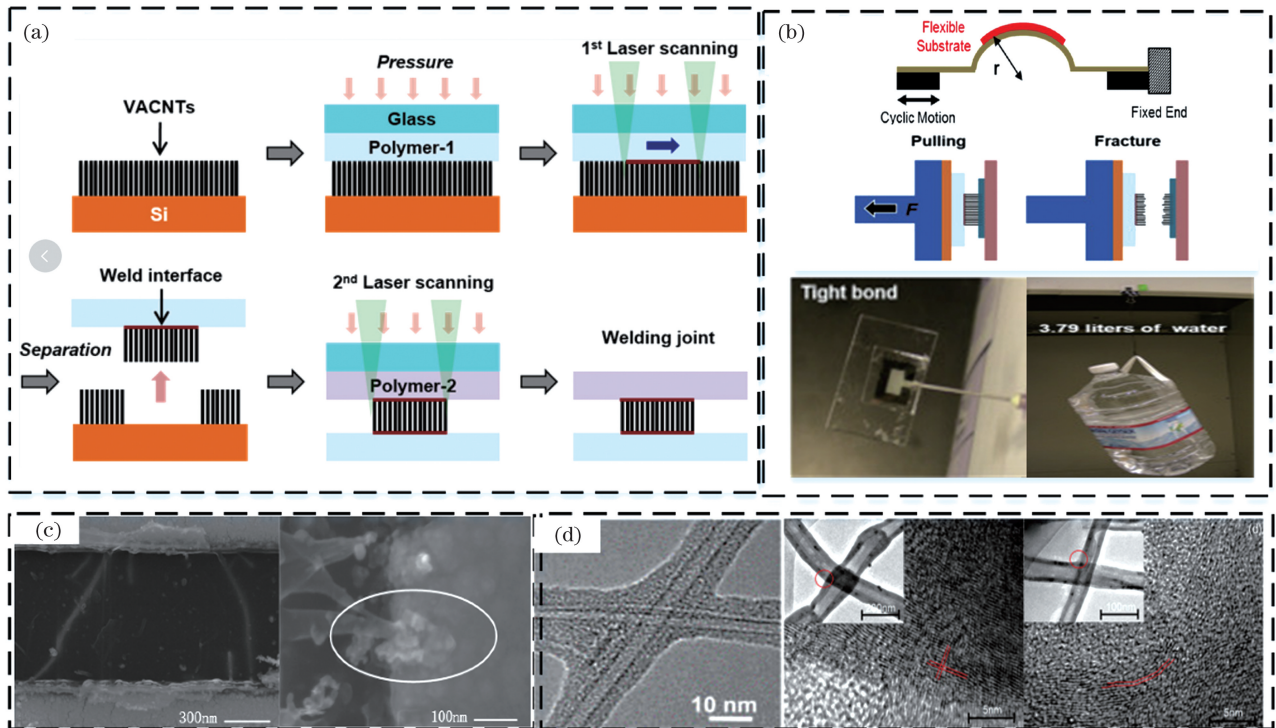


图 10 碳基纳米材料互连网络的电/力学性能。(a)(b)长脉冲激光焊接碳纳米管与柔性基底互连工艺流程及力学性能测试^[79]；(c)激光诱导碳纳米管与金属电极互连^[64]；(d)激光诱导碳纳米管互连网络^[81]

Fig. 10 Electrical/mechanical performance of thin films based on interconnection networks of carbon-based nanomaterials. (a)(b) Laser-induced nanowelding between CNTs and flexible substrate and mechanics properties testing^[79] ; (c) laser-induced carbon nanotubes interconnect with metal electrodes^[64] ; (d) laser-induced carbon nanotubes interconnection network^[81]

对于一些宽带隙半导体纳米材料, 超快激光极高的单脉冲能量能够有效实现其连接^[83]。Xing 等^[23]基于 ZnO 纳米线在飞秒激光辐照过程中的双

光子吸收效应, 实现了宽带隙纳米线之间的连接, 连接后薄膜的光电流响应大大增强, 如图 11(a)所示。Shimogaki 等^[84]通过波长为 355 nm 的激光实现了

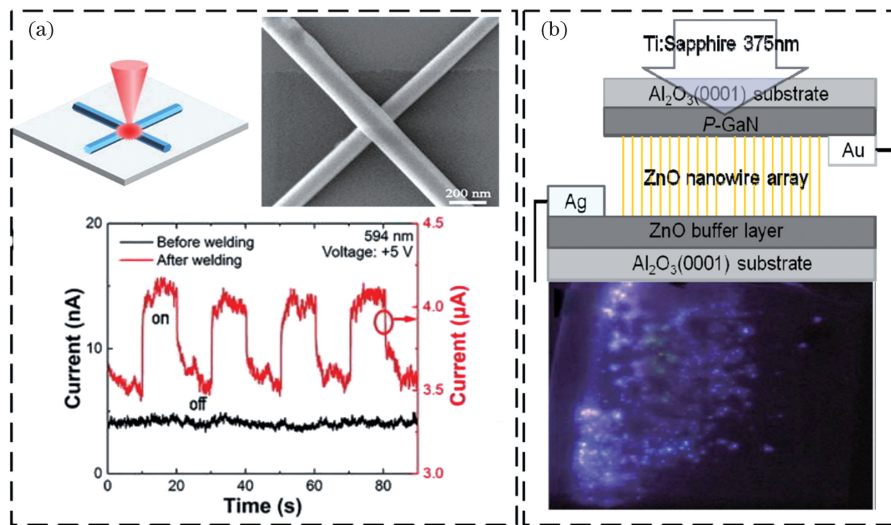


图 11 宽带隙半导体互连结构的电/力学性能。(a)飞秒激光诱导 ZnO 纳米线互连及 ZnO 纳米线的光电响应^[23]；(b)激光诱导 ZnO 与 GaN 薄膜互连及互连后器件的电致发光增强^[84]

Fig. 11 Electrical/mechanical performance of semiconductor interconnect structures. (a) Femtosecond laser-induced nanowelding between ZnO nanowires and its photoelectrical response^[23] ; (b) laser-induced interconnection of ZnO and GaN thin films and electroluminescence enhancement of the device after interconnection^[84]

N 型 ZnO 纳米线与 P 型 GaN 薄膜的连接,结果显示,器件的 $I-V$ 特性和 UV 电致发光强度都得到了明显改善,如图 11(b)所示。

5 结束语

纳米连接技术被视为未来电子产业集成互连封装的关键技术之一,该技术通过在纳米材料互连界面构建稳定的键连接促使纳米材料在互连界面仍具有优异的电/力学性能。研究和探讨纳米材料焊接后的电/力学性能,对于纳米材料未来的工程应用具有重要意义。

目前,纳米连接的主要方法包括低温冷焊^[6-8]、压焊^[9-10]、超声焊接^[77,85]、电场及化学辅助连接^[2,86-87]、高温及焦耳热熔连接^[80,88-90]、高能束(包含电子束、离子束等)熔接^[13-15]和激光等离子体低温焊接^[4,16-22]等。在纳米互连器件的制备过程中,尤其是在新一代柔性纳米电子器件的制备过程中,不仅要求互连接头具有高的电/力学性能,还要求焊接环境具备低温、低应力特征,以避免其对周围其他纳米器件及基底造成较大影响。因高温熔融得到的纳米接头常常伴随着较大的热影响面积,会对非连接部位的结构产生热影响甚至破坏,继而降低了互连结构整体的电/力学性能,因此近年来人们偏爱采用低温焊接方式来构造具有高电/力学性能的互连器件。

本文结合目前不同纳米连接技术及连接界面的特点,对单纳米焊接接头和宏观焊接结构的电/力学性能进行了探讨。与其他低温焊接技术相比,激光因能诱导纳米材料产生局域自限性低温焊接而具有无限潜力,其焊接接头不仅具有较高的电/力学性能,而且可以突破纳米冷焊的尺寸限制和空间限制,实现大尺寸和一定间距分布(无接触)的纳米材料之间的焊接。因具有显著的近场局域增强和纳米聚焦等特性,激光诱导纳米连接对非焊接部位的影响较小,而且可以通过空间调制(例如偏振状态、空间能量等)实现对焊接界面的精准调控。尤其是超短脉冲激光,其脉冲宽度比电子冷却时间短^[32],可以有效实现冷加工和冷焊接,对于金属材料,甚至是一些键能较高的材料,其极高的单脉冲能量和局域强电场可以有效激发材料表面产生空键,从而形成互连结构;而且,在连接过程中不受纳米材料种类及表面形貌质量的影响,加工过程无接触且对环境要求不高,能够灵活地实现工艺的柔性集成和大规模制造。

虽然目前对激光诱导等离子体自限性低温焊接技术的研究取得了一定进展,但是对于实现高效率、

高精度和高分辨的激光纳米可控互连制造仍面临着较大的挑战,如何实现对纳米尺度焊接点能量的精准调控以及获得纳米尺度下材料的互连机理仍需进一步研究。另外,对于线宽在纳米尺度的互连功能结构,在连接前往往还需要有效的操纵技术对其进行排布组装以及实现后续的高精度互连定位,而这个过程依赖于高精度激光纳米互连装备的集成,但目前这方面的相关研究仍亟需进一步推进。相信随着激光纳米连接技术的发展,它将在下一代电子器件互连封装中扮演着越来越重要的角色。

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Research Progress on Electrical/Mechanical Properties of Interconnection Structures Based on Nanowelding

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Abstract

Significance With the development of new electronic devices for miniaturization, flexibility and intelligence, the diversity of nanomaterial properties and limitations of traditional electrical connection methods bring new challenges in new electronic device preparation. Researchers are encouraged to continue to explore ways to break the limit of the device size. The manufacturing technology has gradually developed to the nanoscale level. Nanowelding is one of the key technologies for integrating nanomaterials with micro and macro systems.

Metal nanomaterials (e. g. , Ag, Au, and Cu) and some carbon-based nanomaterials (e. g. , carbon nanotubes, and graphene) exhibit excellent electrical and thermal properties. Besides, some wide bandgap semiconductor nanomaterials (e. g. , ZnO) have shown great potential in future electronic devices. Not only for homogenous connections but also for the study of electrical and mechanical properties of heterogeneous connections, evaluating their mechanical and electrical properties is crucial for predicting the failure modes of electronic devices.

Stable device performance depends on reliable nanointerconnected structures. The size effect and high specific surface area of nanomaterials make them exhibit different welding characteristics from bulk materials during the welding process. The study on the electrical performance of nanowelding consists of single joints and interconnection networks. The study of a single nanojoint is essential to deepen the understanding of the welding mechanism. For interconnection networks, especially with the rapid development of industries, such as smart touch interactive terminals and wearable electronic equipment flexible solar devices, their performance has attracted significant attention.

Progress Currently, the electrical and mechanical characterization of nanoconnection quality consists of two methods. The first method is aimed at electrical testing and characterization of nanoconnected single-welded joints, such as direct *in-suit* measurement of one-dimensional (1D) nanowire and nanotube-welding points. The second method indirectly characterizes macroscopic devices based on nanointerconnections, especially for some flexible film structures. For the study on the performance of 1D nanowires and single-nanometer connection joints of tubes, some researchers have used molecular dynamics-related simulation software to simulate the mechanical and electrical properties of their interface and perform atomic simulation of the entire welding process. The morphology and influencing factors are analyzed to obtain theoretical electrical performance before and after welding. For experimental measurement, if the electrical and mechanical properties are to be directly characterized at such a small scale, with the development of characterization technology, direct mechanical measurement of solder joints can be achieved. However, there are still many challenges in the actual measurement process.

The current nanowelding methods are low-temperature cold welding, pressure welding, ultrasonic welding, electric field and chemical-assisted welding, high temperature and Joule welding, high-energy beam welding (e. g. , electron and ion beam), and laser-induced plasma welding at local low temperature. During the preparation of nanointerconnection devices, especially for the new generation of flexible nanoelectronics, it is necessary to prepare interconnect joints with high electrical performance and a low-temperature and low-stress welding environment, which does not cause damage to other surrounding nanodevices and substrates. The nanojoints obtained using high-temperature melting are often accompanied by a relatively large heat-affected area, which will also have a thermal impact or even damage to the structure of the nonconnected parts, and then reduce the electrical performance of the overall interconnection structure.

Conclusion and Prospect This study summarizes and prospects the electrical and mechanical properties of different materials from the atomic scale to single welded joints, and then to macroscopic multinanoscale welded joints by combining the characteristics of current different nanowelding technologies and their welding interfaces. The discussion of welding structure and deformation mechanism, welding strength, fatigue characteristics, and electrical performance showed that laser-induced plasma welding with characteristics of self-limiting and low-temperature has great potential in fabricating nanodevices and flexible electronic devices.

Although the current study on laser-induced plasma self-limiting low-temperature welding technology has achieved a certain progress, it still faces huge challenges for achieving high-efficiency, high-precision, and high-resolution laser-induced nanocontrollable interconnection manufacturing. The realization of the energy precise control of the nanoscale joints and interconnection mechanism of materials at the nanoscale still needs further study. Besides, for interconnection functional structures with nanoscale line widths, effective manipulation techniques are often required to arrange and assemble them before the connection. It is necessary to achieve subsequent high-precision positioning. This process relies on the integration of high-precision laser nanowelding equipment; however, related technologies still need further study and development. It is believed that the continuous development of laser nanowelding technology will play a significant role in the next generation of electronic device interconnection packaging.

Key words optical fabrication; nanowelding; cold welding; laser-induced plasmon; electrical property; mechanical property

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