

多探针扫描探针显微镜研究进展与应用

许可¹, 邵永健¹, 李鹏^{2*}, 裘晓辉^{2*}

1. 沈阳建筑大学信息与工程学院, 沈阳 110168;

2. 国家纳米科学中心, 中国科学院纳米科学卓越创新中心, 中国科学院纳米标准与检测重点实验室, 北京 100190

* 联系人, E-mail: lipeng@nanoctr.cn; xhqu@nanoctr.cn

2018-09-01 收稿, 2018-09-27 修回, 2018-10-17 接受, 2018-11-22 网络版发表

国家自然科学基金(61604048, 61327813)和辽宁省自然科学基金重点项目(20170540748)资助

摘要 扫描探针显微镜(SPM)是微纳尺度形貌表征、物性测量及微纳操作的重要工具之一. 传统的SPM只有单一探针, 功能单一, 多探针扫描探针显微镜(MP-SPM)的出现拓展了SPM的应用. MP-SPM的多个探针可充当精确定位的测量电极, 从而提供了一种无损探测样品微纳尺度电学输运性质的方法; 也可相当于多只独立活动的“手”, 相互配合实现复杂的纳米操作; 还可以探针成像, 成像信息作为其他探针操作的先验/反馈信息, 从而提高操作的效率及准确性. 本文首先介绍了MP-SPM的基本仪器结构, 多探针距离缩小及位置标定方法, 以及使用多探针技术测量材料电阻率的原理, 接着总结了近年来MP-SPM在样品微纳尺度电学输运性质测量、微纳操作、并行成像与操作以及新型力学性质测量等方面的应用, 最后探讨了该技术的前沿发展以及面临的机遇与挑战.

关键词 多探针, 扫描探针显微镜, 电学输运性质测量, 微纳操作, 并行成像与操作

扫描探针显微镜(SPM)作为纳米科技研究重要研究手段, 为纳米科技发展起到了积极的推动作用. SPM主要包括扫描隧道显微镜(STM)和原子力显微镜(AFM), 其不仅可以作为“眼”, 高分辨率观测物质表面的形貌结构(横向可优于0.1 nm, 纵向可优于0.01 nm), 以及探测多种物理化学性质, 比如电学、热学、电化学及磁学等; 还可以作为“手”, 去操纵微纳材料, 制备新颖物理、化学特性的功能器件. 但传统的SPM只有一个探针, 在某些方面如材料横向电导测量, 三维微纳操作等不足以胜任, 多探针扫描探针显微镜(MP-SPM)利用其多个可独立驱动的探针, 不仅可协同多点测量, 还可提供更多的操作“手”, 相互配合实现更为高效、更为复杂的微纳操作, 从而提供更强大的手段, 进一步拓展了SPM技术的应用.

表征材料电学输运性质信息能够有效指导合成高质量材料及制备高性能器件, 在材料领域、微电子

领域中有着非常重要的应用. 目前测量样品导电性的方法主要有利用微加工技术制作电极^[1~3], 使用集成的微型四探针芯片^[4]以及使用MP-SPM^[5]这3种. 前两种方法尚存在一些不足, 微加工技术制作电极会在材料表面残留光刻胶和有机溶剂, 从而影响材料本征的输运性质测试. 集成的微型四探针芯片, 由于制造工艺等原因使得探针间的距离不可能很小(一般在几百纳米至几微米), 且集成的芯片一旦制成探针间的距离将不会改变, 这限制了其使用的灵活性. 而MP-SPM的多个探针可作为多个测量电极, 位置灵活可控, 且探针间的距离能做到非常小(几十纳米), 从而提供了一种快速无损且灵活的微纳尺度电学输运测量手段.

微纳操作是指通过对微纳尺度对象的操作, 以实现特定功能微纳结构或器件制造的技术. 典型的微纳操作系统主要有: 基于光镊、磁镊、扫描电子显

引用格式: 许可, 邵永健, 李鹏, 等. 多探针扫描探针显微镜研究进展与应用. 科学通报, 2018, 63: 3713~3726

Xu K, Shao Y J, Li P, et al. Progress and development in multiple-probe scanning probe microscopes (in Chinese). Chin Sci Bull, 2018, 63: 3713~3726, doi: 10.1360/N972018-00909

微镜(SEM)和SPM的微纳操作系统. 光镊微纳操作系统利用激光形成的光阱来束缚微小物体, 通过移动该光阱位置从而实现对微小物体的移动及操作. 但持续的激光照射可能对样品造成损害, 且光镊的分辨率也较低. 磁镊技术则是利用磁场控制与样品相连的磁珠从而达到对样品操控的目的, 但其对物体的定量、定点操控能力较弱, 同样其分辨率也较低, 尚不能满足对纳米物质精确操作和构建纳米精确结构的要求. 基于SEM的操作系统是在SEM的真空腔内集成操作手, 在SEM实时图像指引下实现对物体的操作, SEM图像的高分辨率能力(数纳米)使得操控物体尺寸及精度可至纳米级别, 但它对样品的镀金、真空环境要求及缺少实时力反馈等限制了其应用. 基于SPM的操作系统提供了一种简单方便且高精度的微纳操作手段, 能够对物体进行推拉刻划等操作^[6-9], 但其往往局限于二维空间, 较为复杂的三维操控则不能胜任^[10]. 同时由于在操控过程中缺乏实时反馈, 需要采用扫描-操作-再扫描等耗时耗劲的工作方式, 虽然也有研究者设计了增强现实^[11]、局部扫描^[12]等方法提高了操作效率及质量, 但操控效率仍偏低. 另外, 适合操作的探针往往很难提供高质量的成像, 这就需要更换探针来进行结果检查, 使得实验操作更为繁琐. 而MP-AFM则可以解决上述问题, 它在同一区域内多个探针可协同工作, 并且通过探针间相互配合能够较为容易地实现高效的三维操作以及并行的成像/操作任务.

本文将重点论述MP-SPM的基本结构和关键技术, 及其在电学输运性质测量、微纳操作、新型力学探测及其他相关研究领域中的应用, 并探讨了该技术面临的机遇与挑战.

1 MP-SPM仪器结构及其关键技术

MP-SPM主要分为MP-STM和MP-AFM, 一般由多套可以被独立控制的探针驱动机构和位移检测机构组成. 根据受控探针数目的不同, 又可分成双探针扫描探针显微镜(DP-SPM)、三探针扫描探针显微镜(TP-SPM)和四探针扫描探针显微镜(QP-SPM).

1.1 MP-STM仪器结构

世界上第一台DP-STM, 是由日本的Masakazu Aono研究组制造^[13]. 它由2个相对的STM探针组成, 每个探针都连有压电陶瓷和惯性驱动器来进行探针

的精确定位和粗调, 其配有长焦光学显微镜用来观察探针间粗略的相对位置. 在第一台DP-STM发明后, 世界上其他的研究组也做了类似的研究, 制造了许多不同特点的DP-STM. Grube等人^[14]制造的DP-STM, 其样品台与水平方向成45°角放置, 两个探针则水平和竖直布置; Miyata等人^[15]将STM探针和集成的微型4探针芯片^[16]组合在一起制成了双探针系统; Eder等人^[17]制造了一台两个探针面对面水平布置, 而样品在探针间垂直放置的DP-STM. 为了进一步节约空间, Jaschinsky等人^[18]设置了垂直方向上排列的多层环状结构, 每个探针均由两层圆环控制, 如图1(a)所示, 其中上层的圆环控制探针在垂直方向运动, 下层的圆环控制探针在水平方向上运动.

双探针法在电学输运性质测量时, 针尖与样品间接触电阻较大, 不可忽略, 因此需多次改变探针间距再经过数据拟合提取出样品本征电阻率^[23,24]. 而四探针法通过一次测量便可获得样品本征电学性质, 消除了接触电阻影响, 因此QP-SPM得到迅速发展. Salomons等人^[19]制造的TP-STM集成了SEM和场离子显微镜(FIM), 虽然它只有3个探针但可利用自制的程序完成四探针测量, 如图1(b)所示. Cherepanov等人^[20]在圆环式紧凑结构DP-STM基础上进一步拓展, 搭建了超紧凑结构的QP-STM, 如图1(c)所示. 其四套粗调及扫描装置均集成在一个圆筒内, 直径仅50 mm. Ma等人^[21]对一台老的商业QP-STM(OMICRON Nanoprobe)进行了全方位的改造, 除了在减震、导热及SEM替换等方面改造外, 还采取了双扫描管机制, 在保证大扫描范围的同时还保证了高分辨率, 如图1(d)所示. Hobara等人^[22]制造的QP-STM, 如图1(e)所示, 其工作温度可从室温到7 K, 使得它可以测量不同温度下样品的导电性. Guise等人^[25]设计的QP-STM则是根据球坐标系粗调运动, 可较容易调节探针与样品的夹角. Higuchi等人^[26]设计的QP-STM采用样品扫描的方式不仅可较容易地同时获得4幅图像, 且能够有效防止撞针事故的发生. 目前, 已有公司将QP-STM商品化, 比较有代表性的是Scienta Omicron公司的产品, 它采用压电的惯性驱动器和堆叠扫描器, 分别实现探针的粗调和精确扫描, 如图1(f)所示.

1.2 MP-AFM仪器结构

MP-STM的发明在一定程度上满足了研究人员

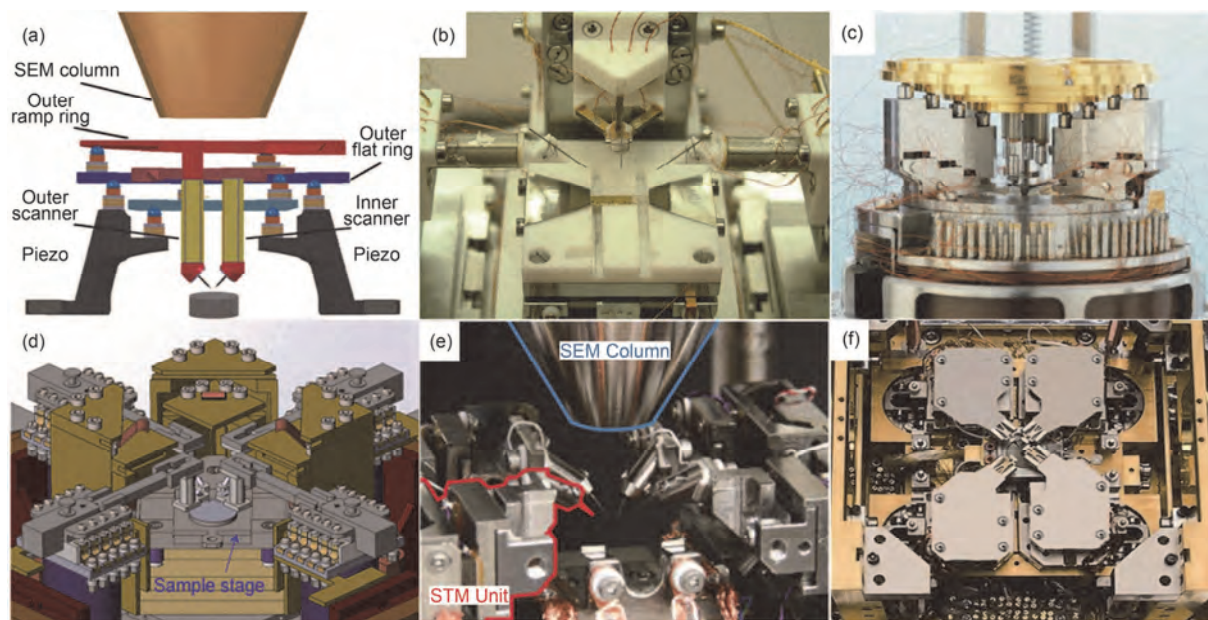


图1 多种多样的MP-STM. (a) 紧凑的多层环状结构的DP-STM^[18]示意图. (b) TP-STM^[19]的照片. (c) 超紧凑的QP-STM^[20]的照片. (d) 升级后的商业QP-STM^[21]的示意图. (e) Hobara等人^[22]制造的QP-STM的照片. (f) Scienta Omicron公司商品化QP-STM的照片

Figure 1 Various MP-STMs. (a) Schematic diagram of compact DP-STM with multilayer rings^[18]. (b) Photo of TP-STM^[19]. (c) Photo of ultra compact QP-STM^[20]. (d) Schematic diagram of an upgraded commercial QP-STM^[21]. (e) Photo of QP-STM manufactured by Hobara et al.^[22]. (f) Photo of commercial QP-STM of Scienta Omicron

的需求,但仍受样品衬底必须导电的限制,而MP-AFM则无此限制,其应用更为广泛.但相比于STM,AFM的位移检测机构较为复杂,因而对于MP-AFM来说,其结构设计显得尤为重要.MP-AFM的结构根据检测机制的不同可分为自检测探针和激光反射法两大类.前者是利用自检测探针,无需繁琐的光学部件,使得结构设计大大简化.而后者则利用常用的光杠杆检测方法,无需使用特殊的探针.

基于自检测探针的MP-AFM结构较为简单,其

位移检测是基于压电^[27]、压阻^[28]探针及石英音叉探针^[29]等方式.Satoh等人^[30]利用压阻探针制造了一台结构较为简单的DP-AFM,实现了纳米级的精确成像.日本Aono课题组^[31,32]设计了基于音叉式的四探针AFM,该系统的探针特意加长,且与样品成一定角度,可在大气/液体/真空中操作.Nanonics公司也基于音叉探针制造了QP-AFM的商业产品(MultiView 4000),如图2(a)所示,它的每个探针都可以独立地进行AFM、开尔文探针显微镜(KPFM)、静电力显微镜

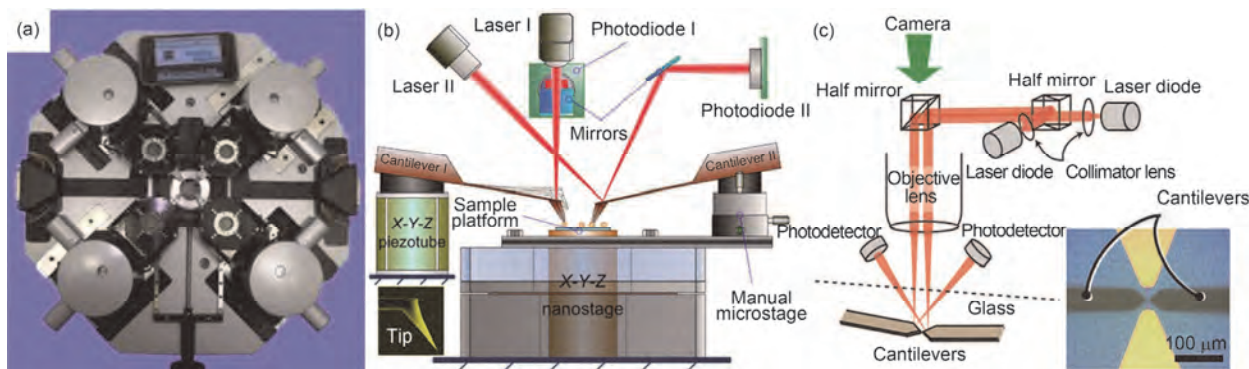


图2 不同结构的MP-AFM. (a) Nanonics公司的QP-AFM产品. (b) 激光倾斜入射的DP-AFM^[33]. (c) 激光垂直入射的DP-AFM^[34]

Figure 2 Various MP-AFMs. (a) Commercial QP-AFM of Nanonics company. (b) DP-AFM with obliquely incident laser beams^[33]. (c) DP-AFM with vertically incident laser beams^[34]

(EFM)、扫描近场光学显微镜(SNOM)等多种性质的表征,是一个强大的多物理场纳米表征及测量研究平台。

上述使用自检测探针会使系统的结构设计较为简单紧凑,且较容易与SEM等其他系统集成。但目前压阻/压电式位移检测方式精度较差,而音叉式探针其检测电路又较为复杂,并且最大的问题是其局限于特殊的探针,无法与功能化探针相兼容,因此激光反射检测方式仍有其优势。采用激光反射法的MP-AFM又分为激光倾斜入射和垂直入射两类。Xie等人^[33]设计的DP-AFM采用激光倾斜入射的方式,如图2(b)所示,可以看到其两束激光分别沿着某一倾角射向悬臂梁。而Tsunemi等人^[34]设计的DP-AFM采用激光垂直入射的方式,两束激光均经过垂直放置的物镜,分别汇聚入射至两探针的悬臂梁上,如图2(c)所示。

1.3 探针间距离缩小及其相对位置标定

MP-SPM需要各探针能在同一区域内共同工作,因此其探针间能实现的最小距离是非常关键的参数。普通的AFM探针针尖形状为倒金字塔形,其针尖位置距离最前端达数微米,因此针尖之间无法靠近,其

针尖间相互距离远大于针尖半径之和。为了解决这一问题,MP-AFM中常采用针尖向前突出的商品化探针^[35](图3(a)),或者使用聚焦离子束(FIB)将普通的探针切割成向前突出的形状^[27,30](图3(b))。对于MP-STM而言,其探针一般由金属线化学腐蚀法得到,且倾斜一定角度,因此实验中两个探针之间最小距离由两探针尖端半径所决定,一般在100 nm左右。为了进一步缩小这一距离,Kubo等人^[36]将在钨探针上生长的氧化钨纳米棒作为探针(图3(c)),在实现了原子级精确成像的同时,还使探针之间的距离达到了72 nm。但用它去测量导电性时,需在探针外镀一层Pt; Yoshimoto等人^[37]和Konishi等人^[38]将碳纳米管连接在钨探针上(图3(d)),同样减小了相对距离,为了保证稳定的电学测量,在整个探针外镀了一层PtIr。各种不同类型的探针如图3(a)~(d)所示。

多探针的相对位置标定是后序多探针并行成像、多点性质测量和协同纳米操作的前提,也可防止后序操作中出现撞针事故。目前的相对位置标定方法有3种。第1种为成像特征点识别^[26,39],其先借助光学显微镜或SEM指导多探针靠的足够近,再多探针同时成像,最后根据得到的多幅图像中的共同特征点计算出探针间相互的距离,如图3(e)所示。第2种为

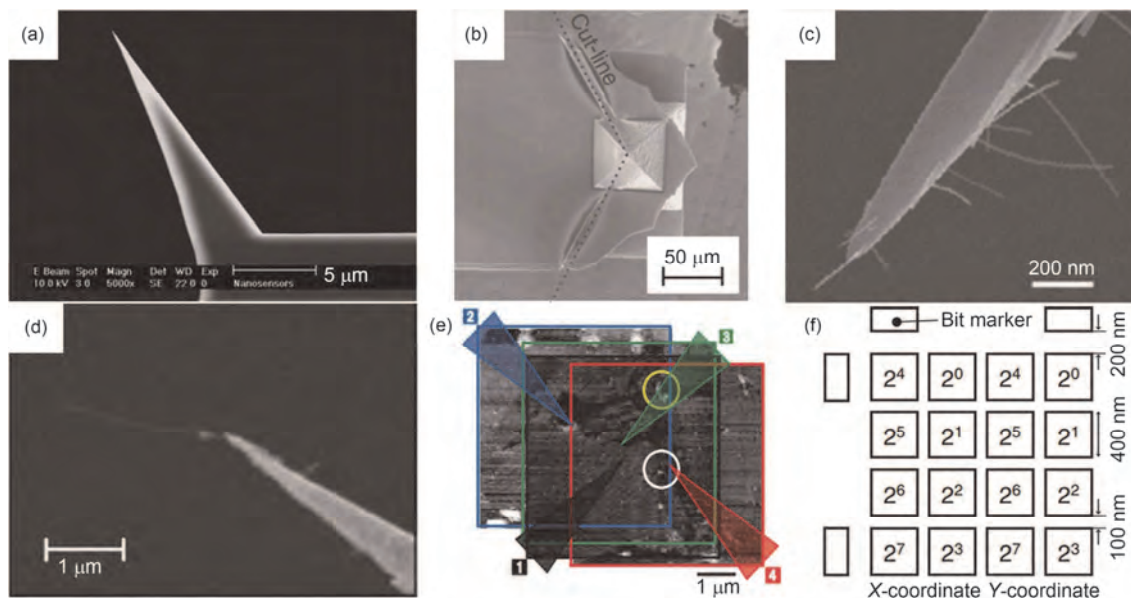


图3 探针种类及相对位置标定方法。(a) 向前突出的商品化AFM探针。(b) FIB修饰后探针^[27]。(c) 氧化钨纳米棒修饰后探针^[36]。(d) 碳纳米管修饰后探针^[37]。(e) 成像特征点识别相对距离^[26]。(f) 地址图案样品识别相对距离^[27]

Figure 3 Different types of probes in MP-SPMs and methods for evaluating the distance between probes. (a) The commercial AFM probes with protrudent tips. (b) The probe modified by FIB^[27]. (c) Epitaxially grown WO₃ nanorod probes^[36]. (d) Carbon nanotube tips^[37]. (e) Evaluation of the distance between probes through images features^[26]. (f) Evaluation of the distance between probes through address-patterned sample^[27]

借助特殊的地址样品^[27,30,40,41],样品上有代表地址的图案,各探针根据扫描的图像便能辨识出探针所在位置,进而可推算出探针间相对距离,地址样品示意图如图3(f)所示.第3种为探针互成像技术^[42],某一探针压在样品表面静止不动,另一探针将其作为样品成像,根据图像中不动探针针尖的位置可推算出各探针的相对位置.

1.4 样品电阻率多探针法测量技术

(i) 电阻率双探针法测量技术. 双探针法测量得到的电阻由样品电阻、探针电阻及探针样品间接触电阻3部分组成,探针电阻较小可忽略,但对于针尖半径数十纳米的测量中,其探针与样品接触电阻较大,可达数十千到上百兆欧姆^[43,44],这时接触电阻的影响不可忽略.但我们可以假设其接触电阻不随探针间距离的变化而变化,则对于一维材料与二维薄膜材料来说,其双探针法测量电阻 R 与探针间距离 s 的关系分别为

$$R_{1D} = \rho \frac{s}{A} + R_c, \quad (1)$$

$$R_{2D} = \frac{\rho}{2\pi t} \times \ln \frac{s^2}{ab} + R_c, \quad (2)$$

其中 R_c 为接触电阻, ρ 为材料的电阻率, A 为一维材料的横截面积, t 为二维薄膜的厚度, a, b 为两个探针的半径.由式(1)与(2)可知,通过多次测量改变探针距离 s

时的电压 U 与电流 I ,得出测量电阻 $R(U/I)$,再经过拟合可求出一维和二维材料的电阻率.但对于三维材料来说,其测量电阻与探针距离无关,所以无法通过双探针的方法得到三维物体的本征电阻率.

(ii) 电阻率四探针法测量技术. 对于四探针法来说,两探针构成电流施加回路,另两探针构成电压测量回路.电流探针间施加电流,在样品内部产生压降,再用高内阻的电压表测出另两探针的电压.电压测量回路的内阻可以认为是无穷大,因此测量的电压仅反映了外加电流引起的样品压降,而与接触电阻、探针电阻无关.表1是不同情况下四探针法所使用的公式总结^[45].

表1中, s 为探针之间的距离(假设线型布置时相邻两探针间距离相等,方块型布置时4个探针构成边长为 s 的正方形); A 为一维材料的横截面积; t 为二维薄板的厚度; R 为测量电阻; ρ_x 、 ρ_y 和 ρ_z 为各向异性物体在 x 、 y 、 z 三个方向上的电阻率; R_x 为测量各向异性物体时沿着 x 方向上的测量电阻即 V_x/I_x ; R_y 的定义与 R_x 相似,即沿着 y 方向上的测量电阻.

在上述公式中若想得到有限的二维物体的电阻率,还需在公式前乘以对应的修正因子,但这些修正因子仅能满足有限的方形或圆形样品的电阻率测量,而范德堡方法^[46,47]则可以测量任意形状的二维导体的电阻率.在使用范德堡方法时需满足以下的条件:4个探针在任意形状样品的边缘;探针的尺寸要小于

表1 四探针测量电阻公式总结

Table 1 Summary of the resistance formulas for four point measurement

样品种类		四探针线型布置	四探针方块型布置
三维物体 (半无限)	各向同性 R	$\frac{\rho}{2\pi s}$	$\frac{\rho \times (2 - \sqrt{2})}{2\pi s}$
	各向异性 R_x	$\frac{1}{2\pi s} \sqrt{\rho_x \rho_y}$	$\frac{\sqrt{\rho_x \rho_y}}{\pi s} \left[1 - \left(1 + \frac{\rho_x}{\rho_y} \right)^{-\frac{1}{2}} \right]$
	各向异性比 R_x/R_y	$\sqrt{\frac{\rho_y}{\rho_x}}$	$\frac{\sqrt{1 + \rho_x/\rho_y} - 1}{\sqrt{1 + \rho_y/\rho_x} - 1}$
二维薄板 (无限)	各向同性 R	$\frac{\rho \ln 2}{\pi t}$	$\frac{\rho \ln 2}{2\pi t}$
	各向异性 R_x	$\frac{\ln 2}{\pi t} \sqrt{\rho_x \rho_y}$	$\frac{\sqrt{\rho_x \rho_y}}{2\pi t} \ln \left(1 + \frac{\rho_x}{\rho_y} \right)$
	各向异性比 R_x/R_y	1	$\frac{\ln(1 + \rho_x/\rho_y)}{\ln(1 + \rho_y/\rho_x)}$
一维线 R		$\rho s/A$	—

样品的尺寸;样品要均匀,各向同性,且很薄;样品要单连通(样品中间不能有洞).满足以上条件后,范德堡认为材料的电阻率满足下式:

$$e^{-\frac{I}{\rho} R_{AB,CD}} + e^{-\frac{I}{\rho} R_{BC,DA}} = 1, \quad (3)$$

式中 $R_{AB,CD}$ 为 V_{CD}/I_{AB} , $R_{BC,DA}$ 为 V_{DA}/I_{BC} ; A,B,C,D为方体型布置的4探针的标号(顺时针或逆时针).

求解式(3)可得

$$\rho = \frac{\pi I}{\ln 2} \frac{R_{AB,CD} + R_{BC,DA}}{2} f, \quad (4)$$

其中 f 可由下式求得

$$\cosh \left\{ \frac{\ln 2}{f} \frac{R_{AB,CD}/R_{BC,DA} - 1}{R_{AB,CD}/R_{BC,DA} + 1} \right\} = \frac{1}{2} e^{\frac{\ln 2}{f}}. \quad (5)$$

2 MP-SPM应用

2.1 一维材料局域电学输运性质测量

一维纳米线有着独特的电学、磁学及光学性质,比如超快光学非线性响应、场致发光、场发射效应等,是构筑新型传感器、低功耗场效应晶体管及自旋电子

学纳米器件的重要功能单元,因此研究一维纳米材料的电子能带结构及输运性质,对于下一代纳米电子学器件的研发有着重要的意义. MP-SPM提供了一种能够对一维纳米线无损原位电学输运性质测量的方法. Korte等人^[48]使用自制的QP-STM探究生长过程中GaAs纳米线性质变化. 如图4(a), (b)所示,实验中GaAs纳米线底部仍连至基底,探针1位于纳米线顶部保持不动并与基底构成电流施加回路,探针2与探针3构成电压测量回路,探针2在顶部附近保持不动,而探针3沿着纳米线运动测量. 电阻随探针3位置变化及模型计算得到掺杂浓度与基底距离的曲线如图4(c)所示,实验结果表明靠近纳米线基座处掺杂度较低,推测是由于纳米线生长过程中高温所致,实验中还证明了机械弯曲并不会改变纳米线的电阻. Higuchi等人^[49]利用DP-AFM测量了聚苯胺单根纤维及交叉节点的导电性,实验配置如图4(d)所示. 测量交叉节点的过程中,为了防止其他纤维的影响,先用探针将要测量的部分单独切割. 实验结果如图4(e)所示,表明纤维电阻较为均匀,但节点处电阻远大于纤维自身. 实验还证明了节点处电阻与施加外力有关,这

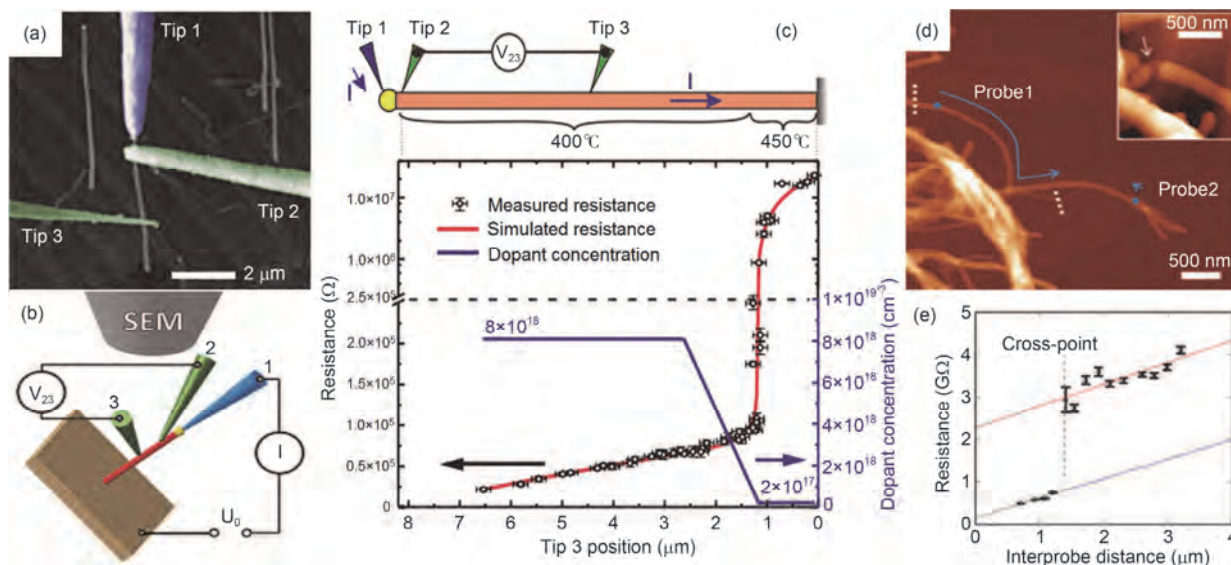


图4 MP-SPM测量纳米线电学输运性质配置及结果. (a) QP-STM的3个探针接触GaAs纳米线SEM图^[48]. (b) 四探针测GaAs纳米线时配线示意图. (c) 电阻及掺杂度随探针3位置变化曲线. (d) DP-AFM测量单根纤维及交叉节点处电阻^[49], 图中蓝点为两个探针的初始位置, 箭头为探针的运动方向, 白色虚线为实验前切割纤维的位置, 插图为切割处的放大图. (e) 所测电阻与探针之间距离的关系

Figure 4 Configuration and results of nanowire electrical transport properties measurement via MP-SPM. (a) The SEM image of GaAs nanowire contacted with three tips in QP-STM^[48]. (b) Schematic diagram including the wiring for four point resistance measurements on the GaAs nanowire. (c) The resistance and the dopant concentration profile with the change of tip 3 position. (d) Resistance measurement of PANI fiber and cross-point junction by DP-AFM^[49], blue dots indicate the initial positions of the probes and arrows indicate the direction of the tip movement, white dashed lines indicate the position where the fiber was cut, the inset shows a high-magnification image of the cut fiber. (e) Measured resistance as a function of interprobe distance

有望作为机械力响应传感器的应用。

2.2 二维材料局域电学输运性质测量

石墨烯等二维材料由于其优异的力学、电学及光学性能在柔性透明器件、场效应管、光电传感器等领域有着广阔前景。目前化学气相沉积(CVD)方法合成的大面积石墨烯往往具有多晶、褶皱等缺陷,这些缺陷会引起载流子严重散射,降低了电导及迁移率,从而制约了石墨烯的实际应用。Ma等人^[21]在自行升级改造的QP-STM基础上,采用直接四探针法测量毫米级单晶石墨烯的电导和载流子的迁移率^[50],测量过程中采用不同的测量角度测3次(每次4个探针构成的矩形相较上一次旋转120°)分别得到石墨烯在不同栅压下的电导,进一步获得了石墨烯的载流子迁移率。实验结果表明在该大面积石墨烯上电导在不同方向上几乎相同,说明了石墨烯在平面内表现为各向同性的输运性质,同时也说明了褶皱、多层石墨烯岛和其他杂质并没有破坏单晶石墨烯整体均一的特性。之后他们又用该仪器测量了双晶石墨烯中晶界(褶皱)输运性质^[51]。实验中他们先测量晶界两侧晶畴不同载流子浓度下的二维电阻,再测量不同载流子浓

度下跨越晶界的二维电阻。根据建立的晶界有效扩散模型得到了晶界或褶皱的电阻率和迁移率。通过对典型的7种晶界和2种褶皱测量,可得知晶界或褶皱对载流子有着很强散射效应,并且晶界对载流子的散射效应要远大于石墨烯褶皱。此外该方法还可拓展至其他二维材料体系,为测量二维材料晶界输运性质提供了简单有效的工具。

Kanagawa等人^[52]使用自制的QP-STM首次直接测量了固体表面单原子层的电学各向异性。实验中分别以线型和方块型布置四探针,测量体系为Si(111)4×1-In表面,方块型布置方式结果表明重构表面具有高度的电学各向异性,各向异性比高达60,而线型布置则不能很好地探测各向异性,这一点也是与表1所吻合的。此外,实验还得到了不同倾角下方块型布置测得的电阻曲线,实验结果如图5(a)~(c)所示。

Si(111)7×7因为其复杂的重构及迷人的电学性质而被广泛研究。但其电导在以往测量结果中差异较大,甚至差好几个数量级^[54,55]。Martins等人^[53]使用了TP-STM对其差异性进行了解释。其配置类似于扫描隧道电位仪(STP)^[56,57],图5(d)即为测量的原理图,在测量时两端探针加一个偏压,中间第3个探针移动

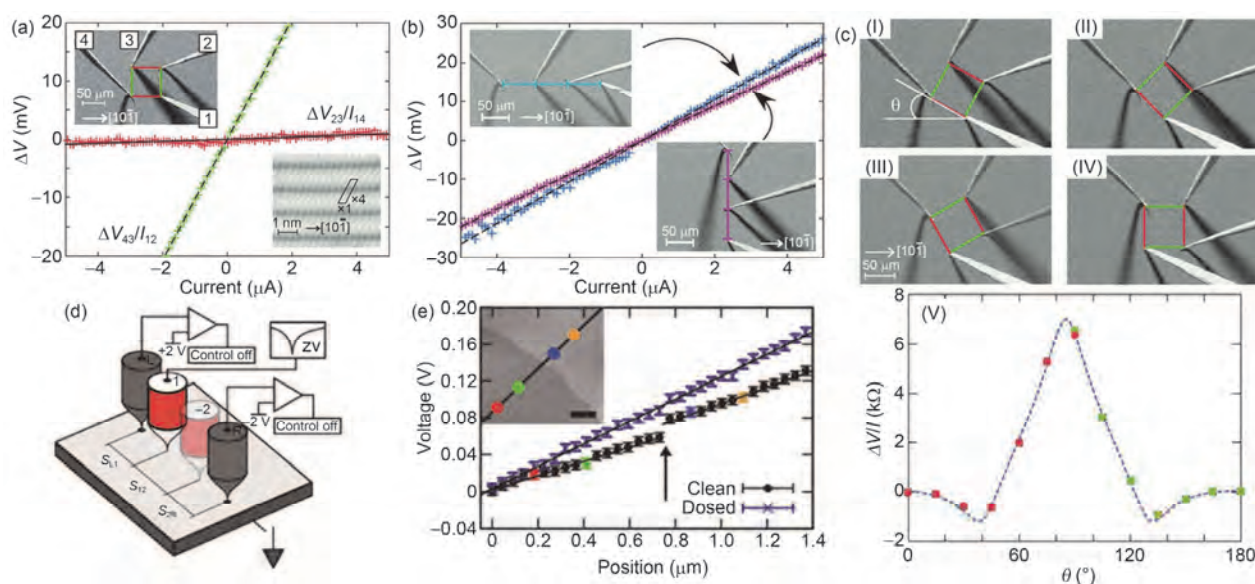


图5 MP-SPM测量二维材料电学输运性质配置及结果。(a)~(c) QP-STM测量Si(111)表面4×1-In重构的电学各向异性^[52]。4探针方形布置(a)和线型布置(b)。(c) 探针不同倾角下测得的电阻。(d)、(e) TP-STM仿四探针测量Si表面原子台阶对导电性的影响^[53]。(d) 3探针仿4探针测量时的配置图。(e) 测得电势随中间探针位置变化的曲线

Figure 5 Configuration and results of two-dimensional materials electrical transport properties measurement by MP-SPM. (a)~(c) Anisotropy in conductance of Si(111)4×1-In measured by QP-STM^[52]. Four-probe measurement (a) in square and (b) linear arrangements. (c) Angle dependence of the measured resistance. (d), (e) Four-probe measures the influence of atomic step to the conductivity of Si surface with a TP-STM^[53]. (d) Configuration of four-probe measurement with a TP-STM. (e) Potential profile with the change of the position of the middle probe

测得各点的电势分布,再根据数学模型求得样品的电导.实验结果如图5(e)所示,其中插图为一个原子台阶的SEM图, clean曲线代表干净的Si表面对应的势能曲线, dosed曲线表示该表面被其他材料覆盖后对应的势能曲线,箭头处对应原子台阶的位置.实验结果表明在Si(111)7×7表面单位长度下一个原子台阶,其电导就比无台阶变化的平坦表面小50倍,这也与Just等人^[58]的推论相似.最后他们认为多探针法测量Si表面的电导差异是由于测量过程中测量点之间包含未知个数的原子台阶所导致.

MP-SPM电学输运性质测量的应用总结如表2所示.

2.3 微纳操作

传统的基于纳米线制作的纳米操作手可以在特定条件下完成对纳米级样品的三维抓取操作^[78],但由于纳米线过于微细且柔软,使得产生的夹持力很难克服物体与基质间的黏附力导致抓取失败,尤其在空气中很难利用它们实现物体的抓取操作. DP-AFM相互独立的探针可相互配合组合成纳米“镊子”,其半径较小的针尖尖端可实现纳米级别物体的接触,且AFM悬臂可产生足够大的夹持力来克服基底黏附力影响,同时操作过程中实时力反馈也保证了操控时的微小接触力,这对柔软的样品显得尤为重要.

谢晖研究组^[79]针对不同外形的夹持器(长方体和圆柱体)进行接触力学建模推导出了接触摩擦力和接触应力大小,并认为非平行的“V”型夹持器在夹取物体上要优于平行配置的夹持器.之后又开发了用于多尺度微/纳米操作和组装应用的DP-AFM系统,该系统利用向前突出的探针(相当于“V”型夹持器),实现了微米球^[80]以及纳米线^[10]拾取-放下的三维操作.为了进一步将功能扩大化,该课题组还使用了微玻

璃管探针^[81],在操作过程中通过对微玻璃管探针的抽气和吹气来实现对微米颗粒或黏合剂的捡起和放下的三维操作.

触觉反馈的加入使得纳米操作更为直观. Iwata等人^[82]和Bolopion等人^[83]分别开发了具有实时触觉反馈的纳米镊子系统,他们采用不同的偏转检测方法, Iwata等人利用自检测探针,而Bolopion等人采用的是激光反射法.

2.4 并行成像/操作

传统的单探针AFM操作效率较低原因在于只有一个探针,无法在操作过程中实时检验操作结果,所以只能采取扫描-操作-再扫描的串行工作方式.而对于MP-AFM来说,各探针均是独立控制,可以实现在同一区域内一探针操作的同时,另一个探针成像来进行实时效果检验的并行工作模式,这极大地提高了操作效率.谢晖等人^[33,84,85]利用双探针在扫描的同时推动纳米颗粒,使纳米颗粒到达指定的位置.成像探针将操作探针及小球位置标定后,操作探针即刻运动至小球位置,以局部扫描的方式确定小球球心,再将小球推至指定位置,如图6(a), (b)所示.图6(a)中左侧为未扫描区域右侧为扫描过的区域;图6(b)为扫描的动态过程,其中I~IV代表小球依次出现,对应的箭头代表探针II任务规划中的操作方向,实验结果如图6(c)所示.相较于以往的扫描-操作-扫描这一流程,并行成像操作有效节约了时间. Loganathan等人^[86]将DP-AFM用于并行成像与机械刻蚀,成像探针提取的结果信息能够指导刻蚀探针相关参数调整,这一过程称为有反馈的刻蚀.而单探针机械刻蚀过程属于开环刻蚀,即在刻蚀过程中没有反馈使得操作结果与目标设定值不符,并且操作结果需要再次成像才能确定.有反馈的操作则能很好解决这一问题,实现

表2 MP-SPM测量材料导电性总结

Table 2 Measure the conductivity of materials using MP-SPM

类型	仪器	样品
一维	DP-STM	Pt ₂ Si ^[59] , ZnO ^[60]
	QP-STM	Bi ₂ Se ₃ ^[61] , CoSi ₂ ^[43,62]
	DP-STM	Au表面 ^[63] , 3-octylthiophene薄膜 ^[64,65] , 石墨烯 ^[66,67]
二维	DP-AFM	α-sexithiophene ^[44] , 并五苯薄膜 ^[68]
	QP-STM	石墨烯 ^[69,70] , Si(111)4×1-In ^[71] , Bi ₂ Te ₂ Se表面 ^[72,73] , Pentacene薄膜 ^[74,75] , Au表面 ^[76] , Te/Si(111)-(1×1)界面 ^[77]

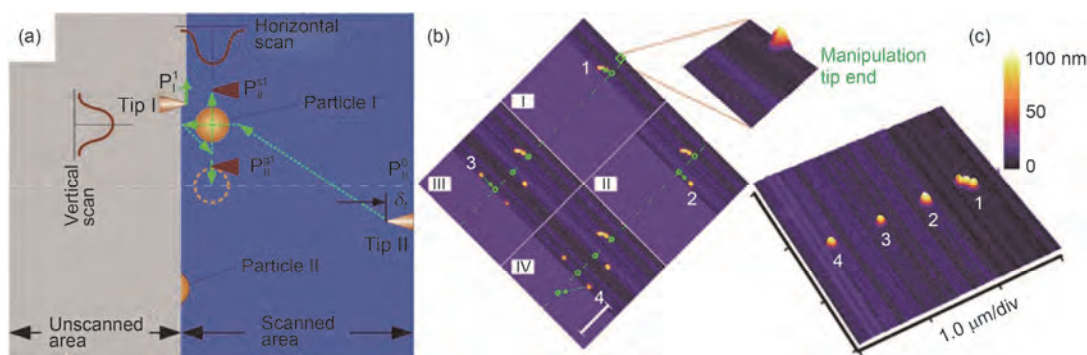


图6 DP-AFM并行成像与操作^[33]. (a) 并行成像与操作纳米颗粒的示意图. (b) 实验中动态过程. (c) 操作结果

Figure 6 Parallel imaging/manipulation with DP-AFM^[33]. (a) Schematic diagram of parallel imaging/manipulation. (b) Dynamic process in experiment. (c) The manipulation result

刻蚀深度与目标设定值相符.

2.5 新型力学测量

材料微纳尺度剥离力精确测量对微纳尺度接触界面现象的揭示有着重要的意义. 研究者将纳米管连接至无针尖的AFM悬臂^[87]或者带自检测功能的悬臂上^[88], 研究纳米管与其他材料剥离时所受力大小. 但该方法需要费力地去制备探针与样品, 降低了测量的

效率. 而DP-AFM组成的纳米镊子在进行物体三维操作的同时还具备实时力监控能力, 因而能够对操作过程中的力精确测量. Xie和Régnier^[89]利用DP-AFM测量了在不同位置处剥离硅基质上的硅纳米线所需的剥离力, 实验配置和结果如图7(a)~(c)所示. 实验结果表明将硅纳米线在不同位置处剥离所需的力有所差别, 但需要的能量则相差不大. 该实验方法可拓展至其他纳米结构, 操作尺寸也可拓展至微米范围.

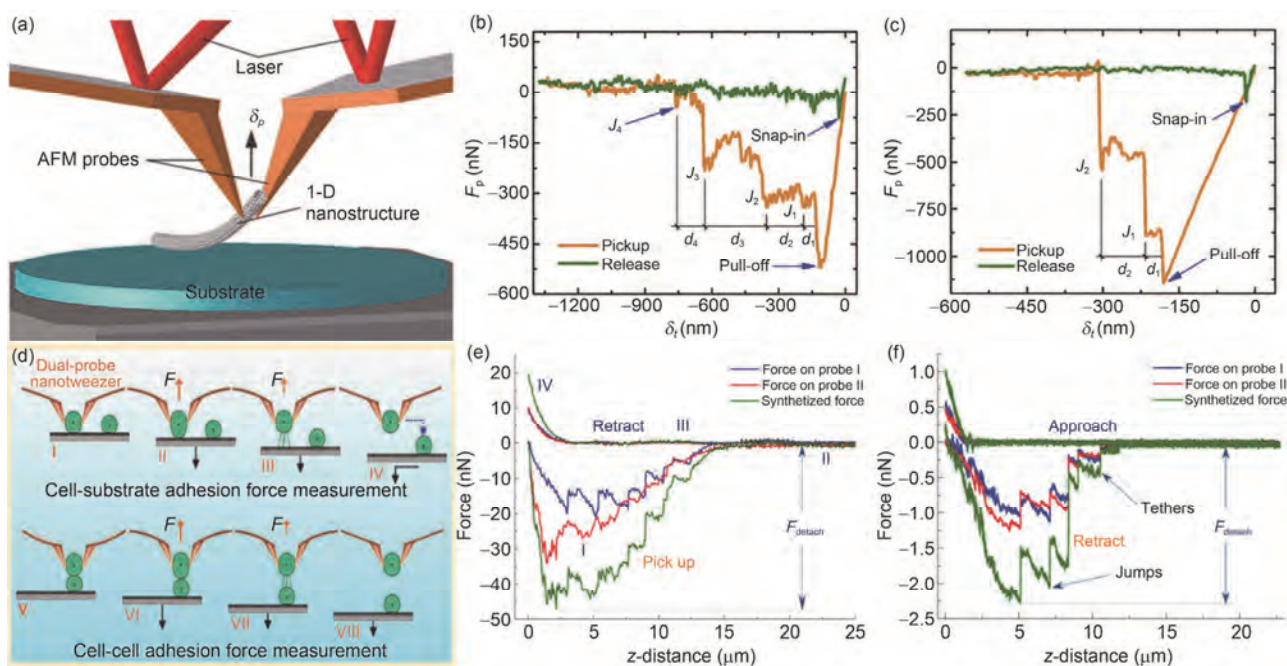


图7 DP-AFM纳米镊子黏附力测量. (a)~(c) 纳米线黏附力测量^[89]. (a) 测量示意图. 从纳米线的一端(b)和中间(c)剥离时的黏附力的曲线. (d)~(f) 细胞黏附力测量^[90]. (d) 测量示意图. 细胞-基质间(e)和细胞-细胞间(f)黏附力曲线

Figure 7 DP-AFM nanotweezer for measuring adhesion force. (a)~(c) Measurements of adhesion force of nanowires^[89]. (a) Schematic diagram of measurement. A full peeling force spectroscopy curve recorded by peeling from the end (b) and the middle (c) of nanowire. (d)~(f) Measurements of adhesion force of cells^[90]. (d) Schematic diagrams of measurement. Adhesion force spectroscopy curve of cell-substrate (e) and cell-cell (f)

细胞与其他材料表面的黏附作用可以为相关疾病的诊断提供依据,还可以对人造器官材料的选择提供指导,对于生物学和医学的研究具有重大的意义. AFM作为研究细胞微纳力学性质的常用手段,它可在液体环境下直接对细胞进行高精度、无损的测量,而DP-AFM不仅可以实现对细胞的三维操作还可以测量在操作细胞的过程中细胞与接触材料之间的黏附力,测量范围可从pN至 μ N. Xie等人^[90]在液下测量了C2C12细胞-基质间和细胞-细胞间黏附力大小,测量配置及结果如图7(d)~(f)所示. 相较于单探针测量,双探针不需要预先使用化学方法将细胞固定在悬臂上可以减小细胞的损伤,且在测量过程中可以随意更换其它细胞进行测量,在一次测量中得到多组数据使得测量更为高效. 该方法还可以测量细胞与其他生物材料之间的黏附力,定量研究不同接触面积下细胞黏附力变化以及温度对黏附力的影响.

3 总结与展望

MP-SPM被发明以来凭借多个可被独立控制的探针被广泛应用在物理、电子及生物等诸多领域中. MP-SPM简化了研究人员的操作步骤提高了测量和操作的效率,利用MP-SPM人们可以更简便、更准确且无损地测得纳米材料的电学输运性质,实现更加复杂的三维纳米操作,可以通过多个探针的相互配合来进行成像,也可以实现成像与操作同时进行的并行操作以节省操作时间,并且还可以实现边操作、边检验、边调整的反馈控制.

虽然MP-SPM已经获得了相当的发展,但其仍然面临以下问题: 其一结构较为复杂,尤其对于采用激

光反射位移检测方法的MP-AFM,繁杂的光学部件及驱动部件给显微镜的结构设计带来很大的困难,使其探针数量很难超过4个;其二操作较为繁琐,单探针AFM成像前只需要一次激光对准及光斑位置调节,但MP-AFM中探针位置移动及逼近过程,激光对准及位置调节需重复多次. 自检测探针在一定程度上缓解了上述问题,但是目前自检测探针种类少,且功能单一,无法兼容其他如热电偶、磁学及电学等功能化探针. 另外目前MP-AFM主要还是利用基本的成像模式实现简单的成像及微纳操作等应用,其他较为先进的成像模式及功能拓展尚未引入,实现功能有限.

为了解决这些问题,在接下来的研究中,研究人员们可以致力于研发性能更好,功能更强大的自检测探针,以代替激光反射法降低MP-AFM的结构复杂程度;引入先进的成像技术及加工方法,如研究人员在DP-AFM中引入KPFM去研究聚丁二炔(PDA)晶体^[91],在一个探针注入电荷的情况下利用另一个探针获得了晶体表面电势分布,证实了PDA晶体的准一维输运特性. Yang和Cui等人^[92,93]利用双探针系统实现纳米焊接及刻蚀操作,两个探针分别为光纤探针与金属AFM探针,操作时光纤探针照射加热金属探针,金属探针针尖处提供了局域的热源,可实现不同材料的焊接与刻蚀操作. 在未来的发展中,一方面可拓展多探针的功能性,如AFM与STM, SNOM等集成实现对样品不同结构物理化学特性的表征;另一方面可设计在变温、超高真空等苛刻环境下仍能工作的MP-SPM,为材料、物理、电子等基础研究提供更强大的利器.

参考文献

- 1 Mirza M M, Maclaren D A, Samarelli A, et al. Determining the electronic performance limitations in top-down-fabricated Si nanowires with mean widths down to 4 nm. *Nano Lett*, 2014, 14: 6056–6060
- 2 Storm K, Halvardsson F, Heurlin M, et al. Spatially resolved Hall effect measurement in a single semiconductor nanowire. *Nat Nanotechnol*, 2012, 7: 718–722
- 3 Ebbesen T W, Lezec H J, Hiura H, et al. Electrical conductivity of individual carbon nanotubes. *Nature*, 1996, 382: 54–56
- 4 Li J C, Wang Y, Ba D C. Characterization of semiconductor surface conductivity by using microscopic four-point probe technique. *Phys Proc*, 2012, 32: 347–355
- 5 Lord A M, Evans J E, Barnett C J, et al. Surface sensitivity of four-probe STM resistivity measurements of bulk ZnO correlated to XPS. *J Phys Condens Matter*, 2017, 29: 384001
- 6 Falvo M R, Helsen A, Chi V, et al. Nanometre-scale rolling and sliding of carbon nanotubes. *Nature*, 1999, 397: 236–238
- 7 Thalhammer S, Stark R W, Müller S, et al. The atomic force microscope as a new microdissecting tool for the generation of genetic probes. *J Struct Biol*, 1997, 119: 232–237

- 8 Vettiger P, Cross G, Despont M, et al. The “millipede”—nanotechnology entering data storage. *IEEE Trans Nanotechnol*, 2002, 1: 39–55
- 9 Sitti M, Hashimoto H. Teleoperated touch feedback from the surfaces at the nanoscale: Modeling and experiments. *IEEE/ASME Trans Mech*, 2003, 8: 287–298
- 10 Xie H, Acosta J C, Haliyo D S, et al. Pick-and-place nanomanipulation with three-dimensional manipulation force microscopy. In: 2009 IEEE/RSJ International Conference on Intelligent Robots and Systems. Louis, 2009. 1333–1338
- 11 Li G, Xi N, Yu M, et al. Development of augmented reality system for AFM-based nanomanipulation. *Mechatron IEEE/ASME Trans Mech*, 2004, 9: 358–365
- 12 Liu L, Zhang J, Li G, et al. On-line sensing and display in atomic force microscope based nanorobotic manipulation. In: 2007 IEEE/ASME International Conference on Advanced Intelligent Mechatronics. Zurich, 2007. 1–6
- 13 Nakayama T, Kubo O, Shingaya Y, et al. Development and application of multiple-probe scanning probe microscopes. *Adv Mater*, 2012, 24: 1675–1692
- 14 Grube H, Harrison B C, Jia J, et al. Stability, resolution, and tip-tip imaging by a dual-probe scanning tunneling microscope. *Rev Sci Instrum*, 2001, 72: 4388–4392
- 15 Miyata N, Hobara R, Narita H, et al. Development of a surface magneto-transport measurement system with multi-probes and the. *Jpn J Appl Phys*, 2011, 50: 36602
- 16 Shiraki I, Nagao T, Hasegawa S, et al. Micro-four-point probes in a UHV scanning electron microscope for *in-situ* surface-conductivity measurements. *Surf Rev Lett*, 2000, 7: 533–537
- 17 Eder F R, Kotakoski J, Holzweber K, et al. Probing from both sides: Reshaping the graphene landscape via face-to-face dual-probe microscopy. *Nano Lett*, 2013, 13: 1934–1940
- 18 Jaschinsky P, Coenen P, Pirug G, et al. Design and performance of a beetle-type double-tip scanning tunneling microscope. *Rev Sci Instrum*, 2006, 77: 93701
- 19 Salomons M, Martins B V C, Zikovsky J, et al. Four-probe measurements with a three-probe scanning tunneling microscope. *Rev Sci Instrum*, 2014, 85: 45126
- 20 Cherepanov V, Zubkov E, Junker H, et al. Ultra compact multitip scanning tunneling microscope with a diameter of 50 nm. *Rev Sci Instrum*, 2012, 83: 33707
- 21 Ma R, Huan Q, Wu L, et al. Upgrade of a commercial four-probe scanning tunneling microscopy system. *Rev Sci Instrum*, 2017, 88: 63704
- 22 Hobara R, Nagamura N, Hasegawa S, et al. Variable-temperature independently driven four-tip scanning tunneling microscope. *Rev Sci Instrum*, 2007, 78: 53705
- 23 Takami K, Akaikasaya M, Saito A, et al. Construction of independently driven double-tip scanning tunneling microscope. *Jpn J Appl Phys*, 2005, 44: 862–866
- 24 Takami K, Mizuno J, Akai-Kasaya M, et al. Conductivity measurement of polydiacetylene thin films by double-tip scanning tunneling microscopy. *J Phys Chem B*, 2004, 108: 16353–16356
- 25 Guise O, Marbach H, Yates J T, et al. Development and performance of the nanoworkbench: A four tip STM for conductivity measurements down to submicrometer scales. *Rev Sci Instrum*, 2005, 76: 45107
- 26 Higuchi S, Kuramochi H, Laurent O, et al. Multiple-scanning-probe tunneling microscope with nanoscale positional recognition function. *Rev Sci Instrum*, 2010, 81: 73706
- 27 Satoh N, Tsunemi E, Miyato Y, et al. Multi-probe atomic force microscopy using piezoelectric cantilevers. *Jpn J Appl Phys*, 2007, 46: 5543–5547
- 28 Iwata F, Mizuguchi Y, Ozawa K, et al. Operation of self-sensitive cantilever in liquid for multiprobe manipulation. *Jpn J Appl Phys*, 2010, 49: 8L–14L
- 29 Botaya L, Coromina X, Samitier J, et al. Visualized multiprobe electrical impedance measurements with STM tips using shear force feedback control. *Sensors*, 2016, 16: 757
- 30 Satoh N, Tsunemi E, Kobayashi K, et al. Multi-probe atomic force microscopy using piezo-resistive cantilevers and interaction between probes. *e-J Surf Sci Nanotechnol*, 2013, 11: 13–17
- 31 Higuchi S, Kuramochi H, Kubo O, et al. Angled long tip to tuning fork probes for atomic force microscopy in various environments. *Rev Sci Instrum*, 2011, 82: 43701
- 32 Higuchi S, Kubo O, Kuramochi H, et al. A quadruple-scanning-probe force microscope for electrical property measurements of microscopic materials. *Nanotechnology*, 2011, 22: 285205
- 33 Xie H, Haliyo D S, Régnier S. Parallel imaging/manipulation force microscopy. *Appl Phys Lett*, 2009, 94: 153106
- 34 Tsunemi E, Kobayashi K, Oyabu N, et al. Development of multi-environment dual-probe atomic force microscopy system using optical beam deflection sensors with vertically incident laser beams. *Rev Sci Instrum*, 2013, 84: 83701
- 35 Xie H, Haliyo D S, Régnier S. A versatile atomic force microscope for three-dimensional nanomanipulation and nanoassembly. *Nanotechnology*, 2009, 20: 215301

- 36 Kubo O, Shingaya Y, Nakaya M, et al. Epitaxially grown WO_x nanorod probes for sub-100 nm multiple-scanning-probe measurement. *Appl Phys Lett*, 2006, 88: 254101
- 37 Yoshimoto S, Murata Y, Hobara R, et al. Electrical characterization of metal-coated carbon nanotube tips. *Jpn J Appl Phys*, 2005, 44: L1563–L1566
- 38 Konishi H, Murata Y, Wongwiriyan W, et al. High-yield synthesis of conductive carbon nanotube tips for multiprobe scanning tunneling microscope. *Rev Sci Instrum*, 2007, 78: 13703
- 39 Tsunemi E, Kobayashi K, Matsushige K, et al. Development of dual-probe atomic force microscopy system using optical beam deflection sensors with obliquely incident laser beams. *Rev Sci Instrum*, 2011, 82: 2717
- 40 Satoh N, Tsunemi E, Kobayashi K, et al. Twin-probe atomic force microscopy with optical beam deflection using vertically incident lasers by two beam splitter. *Electr Commun Jpn*, 2016, 99: 92–100
- 41 Yashiro W, Shiraki I, Miki K. A probe-positioning method with two-dimensional calibration pattern for micro-multi-point probes. *Rev Sci Instrum*, 2003, 74: 2722–2725
- 42 Wu J, Lin Y, Lo Y, et al. Effective tilting angles for a dual probes AFM system to achieve high-precision scanning. *IEEE/ASME Trans Mech*, 2016, 21: 2512–2521
- 43 Yoshimoto S, Murata Y, Kubo K, et al. Four-point probe resistance measurements using PtIr-coated carbon nanotube tips. *Nano Lett*, 2007, 7: 956–959
- 44 Hirose M, Tsunemi E, Kobayashi K, et al. Influence of grain boundary on electrical properties of organic crystalline grains investigated by dual-probe atomic force microscopy. *Appl Phys Lett*, 2013, 103: 173109
- 45 Miccoli I, Edler F, Pfnür H, et al. The 100th anniversary of the four-point probe technique: The role of probe geometries in isotropic and anisotropic systems. *J Phys Condens Matter*, 2015, 27: 223201
- 46 Van der Pauw L J. A method of measuring the resistivity and hall coefficient of lamellae of arbitrary shape. *Philips Techn Rev*, 1958, 20: 220–224
- 47 Van der Pauw L J. A method of measuring specific resistivity and Hall effect of discs of arbitrary shape. *Philips Res Repts*, 1958, 13: 1–9
- 48 Korte S, Steidl M, Prost W, et al. Resistance and dopant profiling along freestanding GaAs nanowires. *Appl Phys Lett*, 2013, 103: 143104
- 49 Higuchi R, Shingaya Y, Nakayama T. Resistance of single polyaniline fibers and their junctions measured by double-probe atomic force microscopy. *Jpn J Appl Phys*, 2016, 55: 8N–9N
- 50 Ma R, Huan Q, Wu L, et al. Direct measurements of conductivity and mobility in millimeter-sized single-crystalline graphene via van der Pauw geometry. *Chin Phys B*, 2017, 26: 307–314
- 51 Ma R, Huan Q, Wu L, et al. Direct four-probe measurement of grain-boundary resistivity and mobility in millimeter-sized graphene. *Nano Lett*, 2017, 17: 5291–5296
- 52 Kanagawa T, Hobara R, Matsuda I, et al. Anisotropy in conductance of a quasi-one-dimensional metallic surface state measured by a square micro-four-point probe method. *Phys Rev Lett*, 2003, 91: 036805
- 53 Martins B V, Smeu M, Livadaru L, et al. Conductivity of $\text{Si}(111)-(7\times 7)$: The role of a single atomic step. *Phys Rev Lett*, 2014, 112: 246802
- 54 Yoo K, Weitering H H. Electrical conductance of reconstructed silicon surfaces. *Phys Rev B*, 2002, 65: 115424
- 55 Jaschinsky P, Wensorra J, Lepsa M I, et al. Nanoscale charge transport measurements using a double-tip scanning tunneling microscope. *J Appl Phys*, 2008, 104: 94307
- 56 Bannani A, Bobisch C A, Möller R. Local potentiometry using a multiprobe scanning tunneling microscope. *Rev Sci Instrum*, 2008, 79: 83704
- 57 Lüpke F, Korte S, Cherepanov V, et al. Scanning tunneling potentiometry implemented into a multi-tip setup by software. *Rev Sci Instrum*, 2015, 86: 123701
- 58 Just S, Blab M, Korte S, et al. Surface and step conductivities on $\text{Si}(111)$ surfaces. *Phys Rev Lett*, 2015, 115: 066801
- 59 Lim D K, Kubo O, Shingaya Y, et al. Low resistivity of Pt silicide nanowires measured using double-scanning-probe tunneling microscope. *Appl Phys Lett*, 2008, 92: 203114
- 60 Lord A M, Walton A S, Maffei T G, et al. ZnO nanowires with Au contacts characterised in the as-grown real device configuration using a local multi-probe method. *Nanotechnology*, 2014, 25: 425706
- 61 Fukui N, Hobara R, Hirahara T, et al. *In situ* microfabrication and measurements of Bi_2Se_3 ultrathin films in a multichamber system with a focused ion beam, molecular beam epitaxy, and four-tip scanning tunneling microscope. *e-J Surf Sci Nanotechnol*, 2014, 12: 423–430
- 62 Okino H, Matsuda I, Hobara R, et al. *In situ* resistance measurements of epitaxial cobalt silicide nanowires on $\text{Si}(110)$. *Appl Phys Lett*, 2005, 86: 233108
- 63 Li K, Zhang C, Wu Y, et al. In-plane anisotropy of quantum transport in artificial two-dimensional au lattices. *Nano Lett*, 2018, 18: 1724–1732
- 64 Takami K, Akai-Kasaya M, Saito A, et al. Control of conduction of iodine-doped poly(3-octylthiophene) thin films by double-tip scanning tunneling microscopy. *Chem Phys Lett*, 2006, 419: 250–253
- 65 Takami K, Tsuruta S, Miyake Y, et al. Electrical conduction of organic ultrathin films evaluated by an independently driven double-tip scanning tunneling microscope. *J Phys Condens Matter*, 2011, 23: 434002

- 66 Settner M, Power S R, Petersen D H, et al. Dual-probe spectroscopic fingerprints of defects in graphene. *Phys Rev B*, 2014, 90: 035440
- 67 Settner M, Power S R, Petersen D H, et al. Theoretical analysis of a dual-probe scanning tunneling microscope setup on graphene. *Phys Rev Lett*, 2014, 112: 96801
- 68 Hirose M, Tsunemi E, Kobayashi K, et al. Investigations of local electrical properties of pentacene thin films by dual-probe atomic force microscopy. *Jpn J Appl Phys*, 2010, 49: 413–437
- 69 Baringhaus J, Edler F, Neumann C, et al. Local transport measurements on epitaxial graphene. *Appl Phys Lett*, 2013, 103: 111604
- 70 Lee J, Yamazaki S, Yun H, et al. Modification of electrical properties of graphene by Substrate-Induced nanomodulation. *Nano Lett*, 2013, 13: 3494–3500
- 71 Uetake T, Hirahara T, Ueda Y, et al. Anisotropic conductivity of the Si(111)4×1-In surface: Transport mechanism determined by the temperature dependence. *Phys Rev B*, 2012, 86: 035325
- 72 Hus S M, Zhang X G, Nguyen G D, et al. Detection of the spin-chemical potential in topological insulators using spin-polarized four-probe STM. *Phys Rev Lett*, 2017, 119: 137202
- 73 Durand C, Zhang X G, Hus S M, et al. Differentiation of surface and bulk conductivities in topological insulators via four-probe spectroscopy. *Nano Lett*, 2016, 16: 2213–2220
- 74 Yoshimoto S, Tsutsui T, Mukai K, et al. Independently driven four-probe method for local electrical characteristics in organic thin-film transistors under controlled channel potential. *Rev Sci Instrum*, 2011, 82: 93902
- 75 Yoshimoto S, Miyahara R, Yoshikura Y, et al. Initial gas exposure effects on monolayer pentacene field-effect transistor studied using four gallium indium probes. *Org Electron*, 2018, 54: 34–39
- 76 Yang J, Sordes D, Kolmer M, et al. Imaging, single atom contact and single atom manipulations at low temperature using the new ScientaOmicron LT-UHV-4 STM. *Eur Phys J Appl Phys*, 2016, 73: 10702
- 77 Lüpke F, Just S, Bihlmayer G, et al. Chalcogenide-based van der Waals epitaxy: Interface conductivity of tellurium on Si(111). *Phys Rev B*, 2017, 96: 035301
- 78 Kim P, Lieber C M. Nanotube nanotweezers. *Science*, 1999, 286: 2148–2150
- 79 Xie H, Lambert P, Régnier S. Analysis of nanoscale mechanical grasping under ambient conditions. *J Micromech Microeng*, 2011, 21: 45009
- 80 Xie H, Régnier S. Development of a flexible robotic system for multiscale applications of micro/nanoscale manipulation and assembly. *IEEE/ASME Trans Mech*, 2011, 16: 266–276
- 81 Xie H, Zhang H, Song J, et al. High-precision automated micromanipulation and adhesive microbonding with cantilevered micropipette probes in the dynamic probing mode. *IEEE/ASME Trans Mech*, 2018, 23: 1425–1435
- 82 Iwata F, Takahashi M, Ko H, et al. Development of a compact nano manipulator based on an atomic force microscope: For monitoring using a scanning electron microscope or an inverted optical microscope. In: 2012 International Conference on Manipulation, Manufacturing and Measurement on the Nanoscale(3M-NANO). Xi'an, 2012. 22–27
- 83 Bolopion A, Xie H, Haliyo D S, et al. Haptic teleoperation for 3-D microassembly of spherical objects. *IEEE/ASME Trans Mech*, 2012, 17: 116–127
- 84 Xie H, Régnier S. High-efficiency automated nanomanipulation with parallel imaging/manipulation force microscopy. *IEEE Trans Nanotech*, 2012, 11: 21–33
- 85 Acosta J C, Poleselmaris J, Thoyer F, et al. Gentle and fast atomic force microscopy with a piezoelectric scanning probe for nanorobotics applications. *Nanotechnology*, 2013, 24: 65502
- 86 Loganathan M, Al-Ogaidi A, Bristow D A. Design and control of a dual-probe atomic force microscope. *IEEE/ASME Trans Mech*, 2018, 23: 424–433
- 87 Barber A H, Cohen S R, Wagner H D. Measurement of carbon nanotube-polymer interfacial strength. *Appl Phys Lett*, 2003, 82: 4140–4142
- 88 Ishikawa M, Harada R, Sasaki N, et al. Visualization of nanoscale peeling of carbon nanotube on graphite. *Appl Phys Lett*, 2008, 93: 89
- 89 Xie H, Régnier S. *In situ* peeling of one-dimensional nanostructures using a dual-probe nanotweezer. *Rev Sci Instrum*, 2010, 81: 35112
- 90 Xie H, Yin M, Rong W, et al. *In situ* quantification of living cell adhesion forces: Single cell force spectroscopy with a nanotweezer. *Langmuir*, 2014, 30: 2952–2959
- 91 Tsunemi E, Kobayashi K, Matsushige K, et al. Visualization of anisotropic conductance in polydiacetylene crystal by dual-probe frequency-modulation atomic force microscopy/Kelvin-probe force microscopy. *J Vac Sci Technol B*, 2010, 28: C4D24–C4D28
- 92 Yang L, Cui J, Wang Y, et al. Nanospot welding of carbon nanotubes using near-field enhancement effect of AFM probe irradiated by optical fiber probe laser. *RSC Adv*, 2015, 5: 56677–56685
- 93 Cui J, Yang L, Xie H, et al. New optical near-field nanolithography with optical fiber probe laser irradiating atomic force microscopy probe tip. *Integr Ferroelectr*, 2016, 169: 124–132

Summary for “多探针扫描探针显微镜研究进展与应用”

Progress and development in multiple-probe scanning probe microscopes

Ke Xu¹, Yongjian Shao¹, Peng Li^{2*} & Xiaohui Qiu^{2*}

¹ School of Information & Control Engineering, Shenyang Jianzhu University, Shenyang 110168, China;

² Chinese Academy of Sciences (CAS) Key Laboratory of Standardization and Measurement for Nanotechnology, CAS Center for Excellence in Nanoscience, National Center for Nanoscience and Technology, Beijing 100190, China

* Corresponding authors, E-mail: lipeng@nanoctr.cn; xhqi@nanoctr.cn

Scanning probe microscopes (SPM) is one of the important tools for micro/nano-scale morphological characterization, physical property measurement and micro/nano operation. However, the traditional SPM has only a single probe and its function is limited. The emergence of multiple-probe scanning probe microscopes (MP-SPM) greatly expands the application of SPM.

Characterizing the electrical transport properties of materials is very important for materials and microelectronics, because it can effectively guide synthetization of high-quality materials and fabrication of high-performance devices. Traditionally, directly measuring the conductivity of nanomaterials is performed using the nanofabricated electrodes connected to the nanomaterial of interest and the monolithic micro-four-point probes. However, photoresist and organic solvents may be left on the surface of the nanomaterial during nanofabrication, which will affect the intrinsic measurement of electrical transport properties. As for the monolithic micro-four-point probes, probe spacing is quite large due to the limitation of the manufacturing process and it will not change any more once they have been manufactured, which limits its flexibility. MP-SPM's multiple probes can act as measuring electrodes with precise localization, and the probe-sample distance can be controlled precisely, providing a means of flexible and non-destructive detection of micro/nanoscale electrical transport properties of samples.

Micro/nano-manipulation systems help to manufacture microelectronic and photonic devices, renewable energy apparatus and biomedical diagnosis chip, etc. Typical micro/nano-manipulation systems are established based on optical tweezer, magnetic tweezer, scanning electron microscopy (SEM) and SPM. However, they all have limitations. Optical tweezer may damage the sample due to the continuously irradiated lasers. Magnetic tweezer has a poor ability to control the object quantificationally, and low resolution. The system based on SEM is restricted to a vacuum working environment. On the surface of the sample, traditional single-probe SPM can perform nano-manipulation, such as push and pull, but it is limited to two-dimensional space, and three-dimensional manipulation might be a great challenge. In addition, due to the lack of real-time visual feedback during the manipulation process, it always works at a time-consuming mode, such as scanning-manipulating-scanning. Although some researchers have designed augmented reality, local scanning and other methods to improve the efficiency of manipulation, the efficiency of manipulation is still low due to the serial procedure of single probe SPM. MP-SPM have been developed to provide multiple probes which can be controlled independently, and achieve complex nano-manipulation by cooperation among the probes. Furthermore, MP-SPM can image with a certain probe; and the imaging results, serving as priori/feedback information, can guide other probes' manipulation, thereby it improves the efficiency and accuracy of micro/nano-manipulation.

In this paper, the basic structure of MP-SPM, including the structure of multiple-probe scanning tunneling microscopy and multiple-probe atomic force microscopy, is presented. Furthermore, we also introduce the methods to decrease the distance between the probes and calibrate the multiple probes' position and the principle of measuring the resistivity of materials using multiple-probe technology. Then we summarize the recent application advances of the measurement of electrical transport properties, the micro/nano-manipulation, the parallel imaging/ manipulation, and the mechanical properties at the micro/nano scale etc. Finally, we discuss the frontier development, the opportunities as well as the challenges of MP-SPM.

multiple-probe, scanning probe microscopy, electrical transport properties characterization, micro/nano manipulation, parallel imaging/manipulation

doi: 10.1360/N972018-00909