

光学学报

高品质因子表面晶格共振

杜艺璇, 敖献煜*, 蔡阳健**

山东师范大学物理与电子科学学院, 山东 济南 250358

摘要 表面晶格共振源于周期阵列的衍射耦合, 理论上可以获得高品质因子, 并可有效增强光与背景环境中物质的相互作用。本文首先介绍在垂直入射激发条件下金属及高折射率介质纳米颗粒阵列中表面晶格共振的基本性质以及实现高品质因子的技术手段; 然后, 介绍基于反射镜上高深宽比介质纳米柱阵列在非对称折射率环境实现高品质因子表面晶格共振的研究进展; 最后, 探讨目前研究存在的局限以及后续的努力方向。

关键词 表面模; 衍射; 表面等离激元; Mie 散射

中图分类号 O436 文献标志码 A

DOI: 10.3788/AOS230861

1 引言

将光场和能量限制在远小于波长尺度的空间内, 可显著增强光与物质的相互作用, 如表面增强荧光^[1]、拉曼散射^[2-3]、红外吸收^[4]等。亚波长尺度的光局域, 还关系到光子器件的小型化及集成化。目前, 在亚波长尺度实现光场局域, 主要依靠金属或高折射率介质的纳米结构^[5]。金属纳米结构表面自由电子在光激励下发生集体相干振荡, 受结构边界的约束, 可形成局域的表面等离激元共振。极端局域的光场可存在于两个金属纳米颗粒之间, 或者金属纳米颗粒与金属膜之间的纳米间隙中, 其强度比单个金属纳米颗粒表面的场强更大。然而, 电子振荡会消耗光场能量^[6]。近年来, 人们把目光转向高折射率介质的纳米结构。高折射率介质的吸收损耗比金属低得多, 可以依靠 Mie 共振来操控光场^[7]。然而, 高折射率介质主要是Ⅳ族和Ⅲ-V 族半导体, 器件的制备方法及其适用的工作波长范围有所限制^[8], 例如, 利用锗纳米结构增强三次谐波时^[9], 所产生的可见光会被结构本身吸收。

对于局域共振, 人们还关心共振的品质因子 Q。Q 因子衡量光子在体系中的存活时间, 同时, Q 因子还与局域电场强度正相关, 因此在包括激光产生、光学折射率传感、非线性光学效应增强等应用场合, 人们期望共振具有高 Q 因子。例如, 对于光学共振折射率传感, 高 Q 因子意味着能较明显地观察到光谱的细微平移。Q 因子一方面受限于所用材料本身的吸收损耗, 另一方面受限于辐射损耗。当金属纳米颗粒排成周期阵列

时, 对入射光的散射可能会产生衍射光波, 如果衍射光波恰好在阵列的周期平面内传播并与单个纳米颗粒的局域共振耦合, 将形成杂化模式——表面晶格共振 (SLR)^[10], 此时, 单颗粒的辐射损耗可通过阵列的衍射耦合抑制, 并产生更显著的场增强, 同时还表现出较高的品质因子。高折射率介质纳米颗粒的周期阵列也可以支持 SLR^[11]。

本文首先回顾金属及高折射率介质纳米颗粒周期阵列 SLR 的基本原理、性质以及获得高 Q 因子的技术手段。在垂直入射激发条件下, 金属及高折射率介质纳米颗粒阵列的衍射耦合要求阵列处于折射率均匀的环境, 且非对称的折射率环境会破坏颗粒间的长程耦合^[12-13]。然后, 介绍基于高反射率金属膜上高深宽比介质纳米柱阵列在非对称折射率环境实现高 Q 因子 SLR 的研究进展。

2 基于金属纳米颗粒阵列的 SLR

局域表面等离激元共振 (LSPR) 的激发伴随着结构表面电场的显著增强, 其共振频率取决于金属颗粒的大小、形状、材料成分和局部光学环境。对于贵金属 (Au、Ag) 纳米结构, LSPR 通常出现在可见和近红外波段。当金属纳米颗粒排成周期阵列时, SLR 通常出现在 $\lambda_{\text{SLR}} \approx n \times P$ (其中 n 为环境折射率, P 为阵列周期) 处, 即满足 Wood-Rayleigh 异常发生的条件(一级衍射光波恰好消失或出现)。为展示二者之间的联系和区别, 图 1 给出了单个金纳米颗粒及其一维周期链的示意图, 以及它们计算的正入射透射谱^[10]。在一维周期

收稿日期: 2023-04-23; 修回日期: 2023-05-19; 录用日期: 2023-05-29; 网络首发日期: 2023-06-28

基金项目: 国家重点研发计划(2022YFA1404800, 2019YFA0705000)、国家自然科学基金(12074227, 12192254, 11974218, 92250304)

通信作者: *aox@sdnu.edu.cn; **yangjian_cai@163.com

链中观察到的共振比单个纳米颗粒有更窄的线宽[周期链的半峰全宽(FWHM)为14 nm,单个颗粒的FWHM为130 nm,这相当于共振质量几乎提高了10倍]。在图1(b)中,SLR的谱呈现非对称Fano线型。Fano共振源自离散态与连续态之间的干涉,这里宽的LSPR可以看成是连续态,而Wood-Rayleigh异常则对应于离散态。作为二者相互作用的结果,SLR能够更好地收集入射波并用于实现局域场增强。如图1(c)所示,阵列中金颗粒表面场强是单个颗粒表面场强的

10倍以上^[14],相应地,模式体积急剧减小。对于荧光增强,阵列不仅可以通过局域场提高激发效率,还能对辐射进行整形^[15]。

理论上SLR可以获得极窄的共振线宽,但在实验上不容易实现,这是因为难以制备精确、无缺陷的纳米颗粒周期阵列。目前主要通过各种光刻方法定义含周期阵列的掩模,再透过掩模沉积金属。基于金属纳米颗粒阵列实现窄线宽SLR的途径大致是通过减小颗粒尺寸与周期之比,提高颗粒的均匀性并降低表面粗糙度。

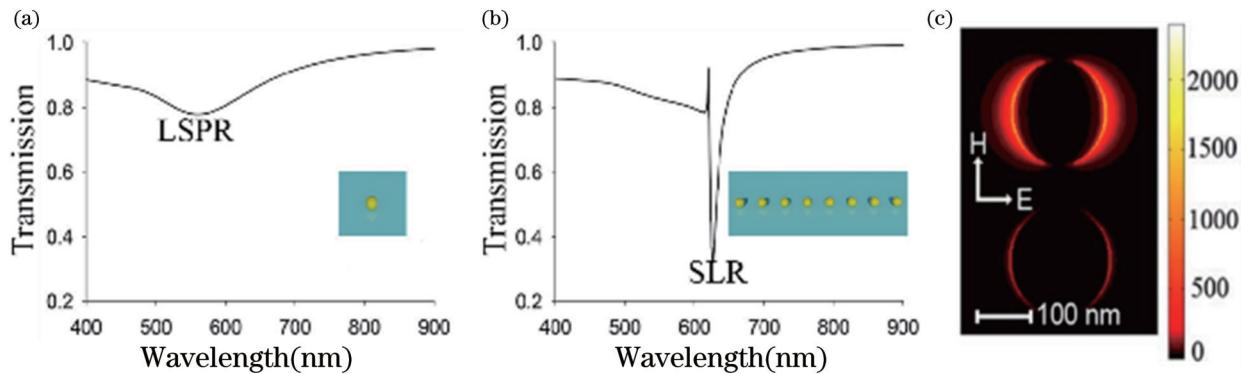


图1 LSPR和SLR的透射光谱及电场强度比较。(a)(b)单个颗粒和一维周期链的光谱,其中金颗粒的半径为80 nm,周期为620 nm,一维链包含1000个颗粒^[10];(c)直径为180 nm、厚度为40 nm的单个金圆盘在 $\lambda=930$ nm处的电场强度分布(下)和光栅常数 $P=640$ nm的二维金圆盘阵列在 $\lambda=982$ nm处的电场强度分布(上)^[14]

Fig. 1 Comparison of LSPR and SLR in transmission spectra and electric field intensity enhancement. (a)(b) Spectra for a single particle and a periodic 1D chain, the gold particles are 80 nm in radius made in air environment, the period in the chain is 620 nm, and the total number of particles in the chain is 1000^[10]; (c) electric field intensity distribution on surface of a single gold disk of 180 nm in diameter and 40 nm thick at $\lambda=930$ nm (bottom) and of a gold disk in a 2D array with $P=640$ nm at $\lambda=982$ nm (top)^[14]

关于贵金属纳米颗粒阵列SLR的品质因子,目前报道的最高实验测量值为 $Q=2340$ (在1550 nm波段)^[16],如图2(a)、(b)所示。渥太华大学的Bin-Alam等^[16]利用电子束光刻结合蒸发沉积形成矩形金纳米颗粒的矩形阵列[图2(a); $t=20$ nm, $L_x=130$ nm, $L_y=200$ nm, $P_x=500$ nm, $P_y=1060$ nm]。颗粒尺寸远小于阵列周期,可使得SLR波长远大于LSPR波长,从而明显降低金属吸收损耗的影响。然而,这样会使得每个颗粒在 λ_{SLR} 处的散射截面很小,形成SLR就需要更多的散射颗粒(即更大的阵列宽度)。他们首先分析了阵列的宽度和激发光源的相干性对所测 Q 值的影响[图2(c)]。可以看到, Q 因子会随着阵列宽度的增大而单调增加。然后,比较了宽带超连续激光和卤钨灯两种光源对测量结果的影响。在相干光束(激光)激发下每个颗粒的相位同步,与非相干光束照明比较,相干光束照明所激发的共振特征更强且线宽更窄。同时,宽带非相干光束不能被很好地准直,使得入射光束包含很多不同入射角度的分量,导致所测得的共振谱范围加宽。对于同一个 $600\text{ }\mu\text{m}\times 600\text{ }\mu\text{m}$ 的阵列,在超连续激光激发下测得 $Q=2340$,而在卤钨灯激发下测得 $Q=1000$ 。

制备方面,透过掩模利用物理气相沉积方法沉积

金属,所形成的金属纳米颗粒是多晶结构且颗粒表面形貌不规则,这一方面是由物理气相沉积过程导致的,另一方面是由掩模的缺陷导致的。为提高沉积金属颗粒形貌的均匀性、降低表面粗糙度,美国西北大学Odom课题组^[17]提出对样品进行退火处理,使颗粒重新结晶的方法。金颗粒样品的退火在900 °C的氩气氛围中进行[图3(a)]。从图3(b)可以看出,退火使得颗粒从类似圆柱形状变成半球形,从而使颗粒表面光滑、形貌均匀,同时颗粒尺寸也变小。如图3(c)所示,对于 $P=600$ nm的金纳米颗粒周期阵列,退火后SLR的FWHM为4 nm,而没有退火、同等颗粒尺寸的沉积颗粒阵列中SLR的FWHM为17 nm。退火过程使晶粒长大,也改善了材料的稳定性,退火样品在普通环境中存储一个月后光学性质保持不变。他们还测试了Ag、Al、Cu纳米颗粒,结果显示,退火处理也能改善它们的光学性质。进一步地,他们还在退火后的Cu颗粒表面通过化学气相沉积方法生长包裹石墨烯,发现在石墨烯包裹的Cu颗粒阵列中形成了FWHM低至2 nm的超窄共振。

基于退火处理的思路,复旦大学聂志鸿和石磊团队^[18]提出一种不需要真空物理沉积过程的金属纳米颗

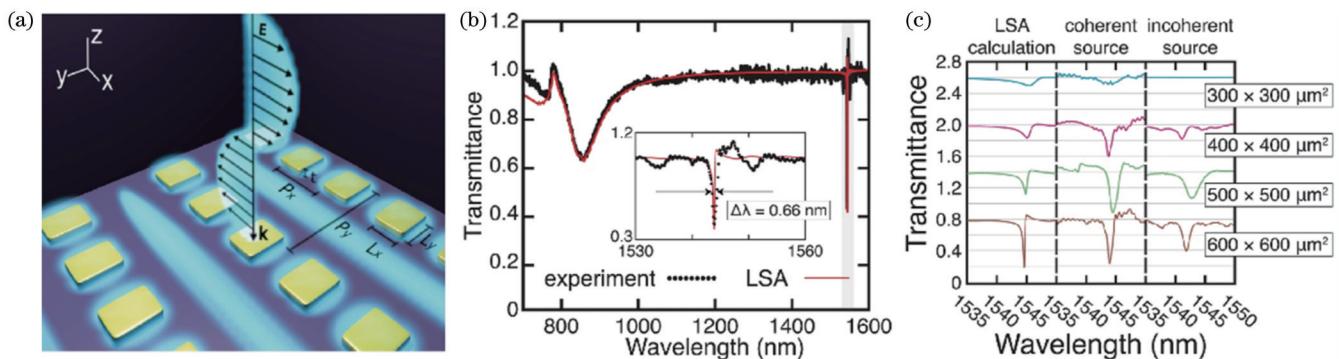


图2 矩形金纳米颗粒阵列的SLR^[16]。(a)结构示意图;(b)测量的透射光谱(黑点)和半解析计算结果(红线),插图为高Q区域的放大图;(c)阵列横向宽度及光源相干性的影响

Fig. 2 SLR in a rectangular array of gold nanoparticles^[16]. (a) Schematic of the array; (b) measured (black dots) and simulated (red line) transmission spectra, and inset is the zoomed plot of the high-Q region

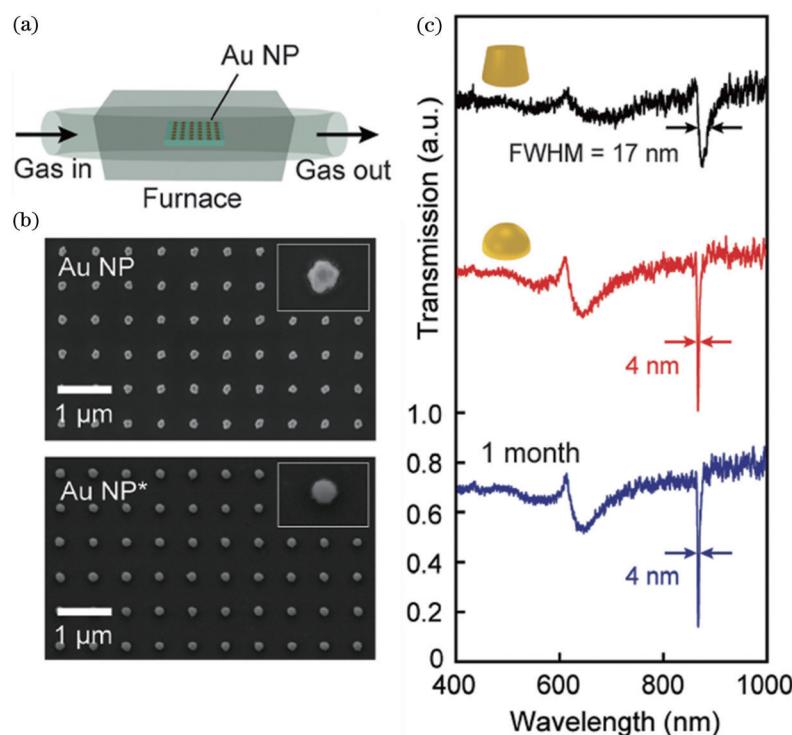


图3 退火改善金纳米颗粒阵列的SLR质量^[17]。(a)退火示意图;(b)退火前后金纳米颗粒阵列扫描电镜图;(c) $P=600\text{ nm}$ 时退火前后的金纳米颗粒阵列的透射光谱比较

Fig. 3 High-quality plasmonic SLR from annealed gold nanoparticle arrays^[17]. (a) Scheme of annealing setup; (b) SEM images of arrays before and after annealing process; (c) measured transmission spectra of untreated arrays and annealed arrays with period $P=600\text{ nm}$

粒阵列制备方法。该方法结合了溶剂辅助软光刻技术和原位定点特异性生长技术。如图4(a)所示:他们首先用包含周期孔阵列的PDMS印章吸附聚四乙烯基吡啶(P4VP)溶液,并将其转印到基片上,形成P4VP纳米柱阵列;其次,用氧气等离子体去除基片表面多余的P4VP后,将基片浸泡到HAuCl₄溶液中,使P4VP吸附Au的前驱体;然后,通过氧气等离子体去除P4VP,同时将金还原出来,并形成有序的金纳米团簇;最后,通过退火形成均匀的金颗粒。通过该方法制备周期

$P=600\text{ nm}$ 的金纳米颗粒阵列,如图4(b)所示,所获得的SLR共振线宽为4 nm,品质因子为218[图4(c)]。从计算结果看,在 $\lambda_{\text{SLR}}=870\text{ nm}$ 处,纳米颗粒周围的最大电场强度提高了约2500倍[图4(d)]。

在前文介绍的简单阵列中,一个格点上只有一个金属颗粒,并且金属颗粒表现出电偶极子类型的局域共振。实际上,还可以通过构造更加复杂的点阵类型[例如,在图5(a)、(b)中,一个格点上放置两个金属颗粒^[19-20]]或者使单个金属颗粒支持高阶多极子类型的

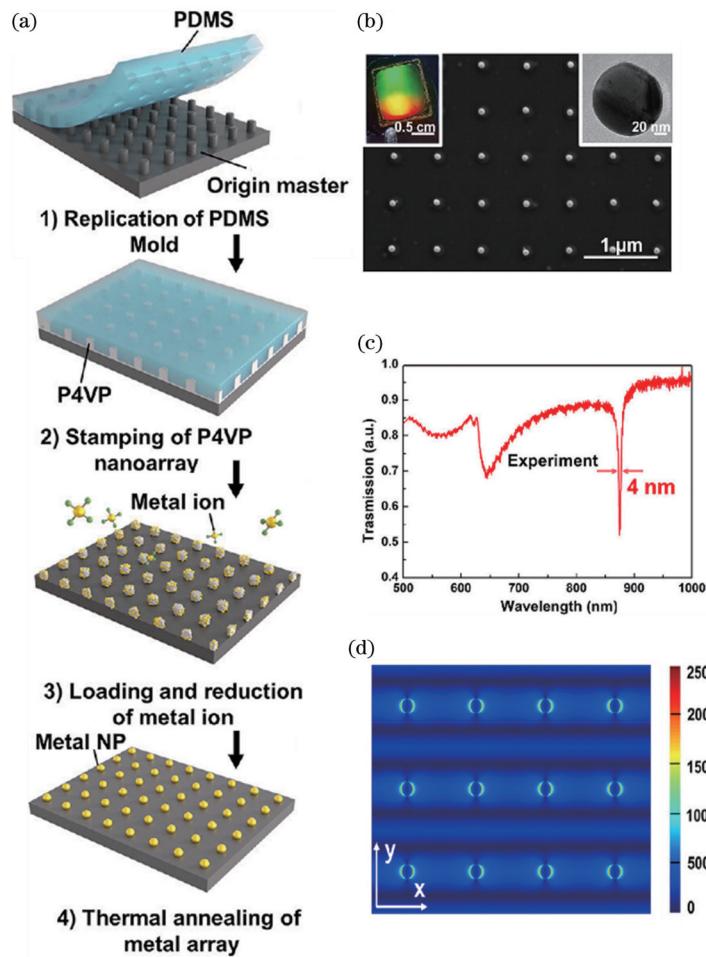


图4 模板辅助化学方法制备的金纳米颗粒阵列^[18]。(a)制备过程示意图;(b)样品照片和扫描电镜图;(c)测量的透射光谱;(d)计算的 $\lambda_{\text{SLR}}=870 \text{ nm}$ 处的电场强度分布

Fig. 4 Gold nanoparticle (GNP) arrays fabricated by template-assisted chemical method^[18]. (a) Schematic of the fabrication process; (b) photograph and SEM images of the sample; (c) measured transmission spectra of GNP array under linearly polarized light; (d) calculated near-field distribution of $|E|^2$ at $\lambda_{\text{SLR}}=870 \text{ nm}$

局域共振如电四极子^[21]来形成SLR。对双颗粒阵列的研究发现,双颗粒可以通过形成准暗态进一步降低辐射损耗,从而获得窄线宽SLR^[19-20]。同时,基于双颗粒准暗态的SLR达到同等品质因子所需的阵列宽度比单颗粒阵列小得多^[20],意味着SLR的形成对阵列的有限宽度更加不敏感。新墨西哥大学Manjavacas课题组^[22]还提出通过周期性地移除常规单颗粒点阵上的一些颗粒来构造复杂点阵[图5(c)],即在单颗粒点阵的基础上引入周期性空缺,也可以看成是两个周期不同的方阵相减得到新的阵列。这些空缺的存在:一方面在原来点阵的基础上引入了额外的周期性,从而在相应波长处产生SLR;另一方面,还引起对称性破缺,从而产生准暗态。渥太华大学Reshef课题组^[23]发现,对矩形点阵的整行颗粒进行周期性平移操作,也能在新的波长位置产生SLR。如图6(a)所示,阵列(i)的SLR发生在 $\lambda_{\text{SLR}} \approx nP_y$,阵列(ii)是将(i)的颗粒每隔一行平移 $\sigma = P_y/3$,此时有新的SLR出现在 $\lambda'_{\text{SLR}} \approx nP'_y$ ($P'_y = 2P_y/3$)处,而 λ'_{SLR} 对应着周期为 P'_y 的矩形单颗

粒阵列(iii)。如图6(b)所示,尽管阵列(ii)是由阵列(iii)每隔两行去掉一行颗粒(虚线标注位置)形成的,但是阵列(ii)和(iii)在 λ'_{SLR} 处的场分布一致。因此,这些复杂的点阵可以带来更加丰富的光学响应。

与LSPR相比,具有高态密度和低光损耗的SLR更容易与入射光有效耦合形成Fano共振,从而显著缩减光谱响应的线宽并增大场强,因此SLR能够有效实现微纳激光输出以及增强非线性响应。美国西北大学Odom课题组^[24]展示了在短波近红外区域Al和Au纳米颗粒阵列电偶极子和四极子类型的SLR模式都可以形成光学反馈并输出激光,且Al和Au表现出相似的阈值及超快动力学特性。此外,在Ag纳米颗粒阵列上涂覆半导体量子点薄膜,可形成杂化的平板波导-SLR,晶格中不同高对称点的SLR能够对应不同的激光辐射方向^[25],还可以控制偏振特性,例如获得径向或角向偏振的激光模式^[26]。除了在辐射光波长(或激发光波长)处的共振增强之外,各向异性的二维点阵还允许在激发光波长和辐射光波长处同时获得SLR,这对

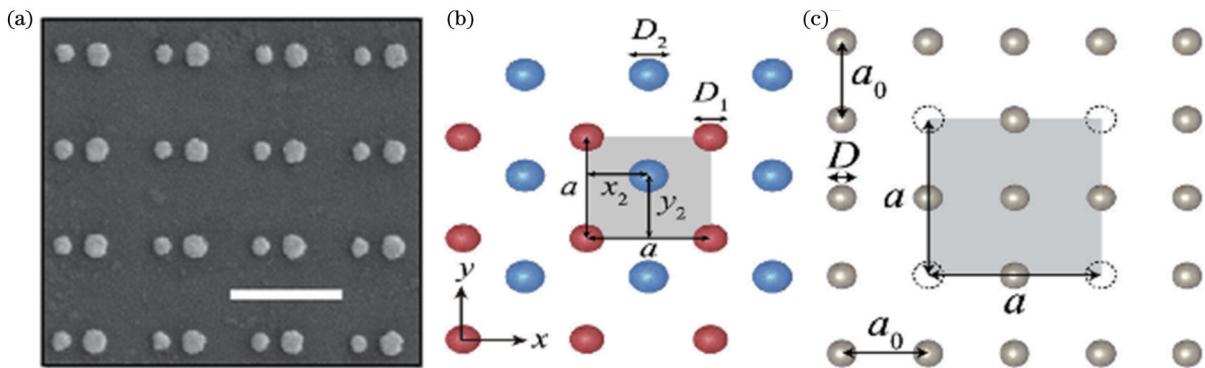


图5 复杂点阵的SLR。(a)晶格常数为450 nm的不对称圆盘二聚体阵列的扫描电镜图^[19]; (b)双颗粒阵列结构示意图^[20]; (c)在周期性纳米阵列中引入周期性空缺的示意图^[22]

Fig. 5 SLR in complex lattices. (a) SEM image of asymmetric disc dimer array with a lattice constant of 450 nm^[19]; (b) schematic of bipartite nanoparticle arrays^[20]; (c) schematic of complex lattices built by removing nanoparticles periodically^[22]

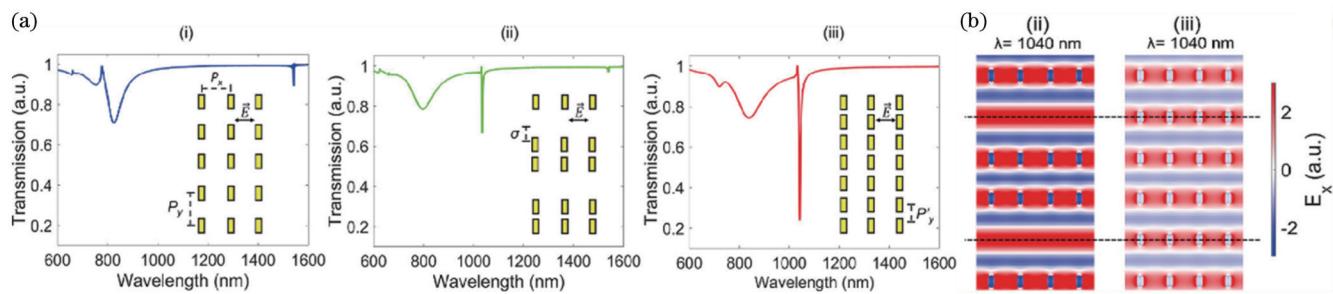


图6 对矩形点阵的整行颗粒进行周期性平移操作后产生新的SLR^[23]。(a)三种不同点阵排列的透射谱及其相应的点阵示意图; (b)点阵(ii)和(iii)在 $\lambda'_\text{SLR}=1040 \text{ nm}$ 处的 E_x 场分布

Fig. 6 SLR produced by periodically shifting the whole rows in a nanoparticle array^[23]. (a) Transmission spectra of three different lattice arrangements (i), (ii), and (iii) with their corresponding schematic of lattices; (b) E_x field profiles at $\lambda'_\text{SLR}=1040 \text{ nm}$ in lattices (ii) and (iii)

于非线性光学效应尤其重要。芬兰坦佩雷大学的Huttunen等^[27-28]设计了L形铝纳米颗粒阵列,使基频光和倍频光的SLR分别在两个正交方向上,数值计算结果表明,“双共振”相对于仅在基频光的“单共振”有额外的倍频转换效率增强。

3 基于介质纳米颗粒阵列的SLR

高折射率介质纳米颗粒可以同时支持电偶极子和磁偶极子特性的Mie散射共振。空气中半径为40~100 nm的硅纳米球最低阶的两个Mie共振落在可见光波段[图7(a)、(b)]^[11],此时材料的吸收损耗较小,其消光谱的主要贡献来自电偶极子和磁偶极子的共振散射。与类似金属纳米颗粒的局域等离激元共振相比,这些高折射率介质纳米颗粒的吸收损耗要小得多,但代价是光场增强的程度也要小很多。波士顿大学Paiella课题组^[13]分析了硅纳米圆柱阵列在近红外波段的场增强情况[图7(c)]。当阵列周期较小 [$P=700 \text{ nm}$, 图7(d)]时,不满足硅纳米圆柱中电偶极子和磁偶极子共振的衍射耦合条件,电场强度仅提升了10~30倍,即相当于单个颗粒的情形;当阵列周期满足

磁偶极子共振衍射耦合条件 [$P=1035 \text{ nm}$, 图7(e)]时,平均电场强度提升了近1000倍,计算的Q因子也达3000;当阵列周期进一步增大时,满足电偶极子共振衍射耦合条件 [$P=1500 \text{ nm}$, 图7(f)],电场的增强效果则有所降低。对比介质颗粒阵列与金属颗粒阵列的SLR场分布,可以看到二者之间存在相似性,即在颗粒之间的区域也能观察到场增强。对于介质颗粒,电偶极子与磁偶极子共振对应的SLR场分布的条纹朝向相互垂直,因此它们各自形成SLR的波长位置可以通过相应方向的周期独立调节,并且单个颗粒中电偶极子和磁偶极子共振发生的相对波长位置可以通过颗粒尺寸调控,由此可以产生很多有趣的现象如抑制背散射^[29]。

京都大学的Murai等^[30]研究了SLR线宽的调控。对于Mie共振波长小于 λ_SLR 的情况,减小颗粒尺寸或者增大阵列周期 P 会减弱共振的衍射耦合,导致线宽变窄。这一点与金属纳米颗粒阵列类似。他们在多晶硅纳米颗粒阵列上旋涂掺有染料的PMMA(基底为石英),观察到电偶极子或磁偶极子类型的SLR都能与荧光辐射耦合,考虑到PMMA层较厚,荧光增强主要来自对激发光的吸收增强以及对荧光辐射的整形。

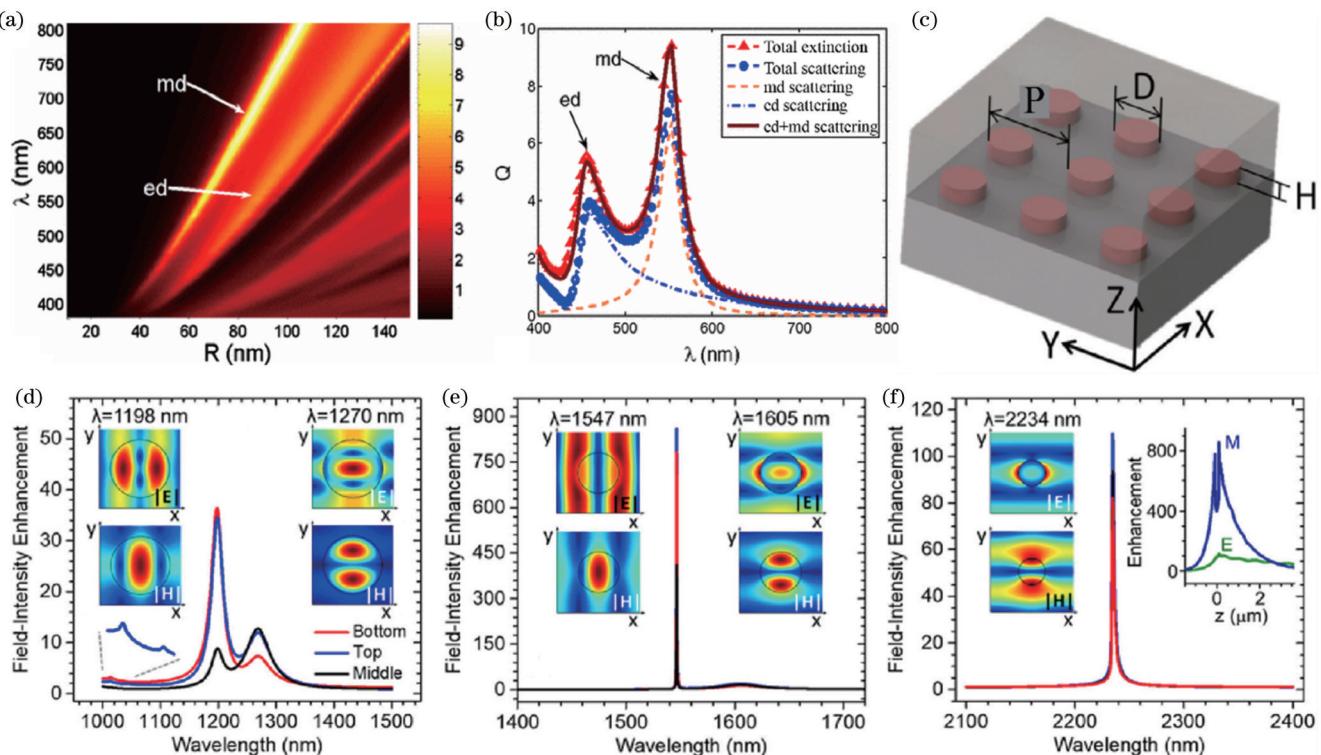


图7 高折射率介质纳米颗粒阵列的SLR^[11,13]。(a) 半径为 R 的硅球形颗粒在空气中的散射效率谱;(b) 硅颗粒半径 $R=65\text{ nm}$ 时的消光和散射光谱;(c) 嵌入聚合物的 SiO_2 衬底上的Si圆柱阵列结构示意图;(d) $P=700\text{ nm}$ 阵列在不同水平监测平面上的电场强度增强谱,插图为峰值波长处的场分布;(e) $P=1035\text{ nm}$ 阵列的电场强度增强谱;(f) $P=1500\text{ nm}$ 阵列的电场强度增强谱

Fig. 7 SLR from high-index dielectric nanoparticle arrays^[11,13]. (a) Scattering efficiency spectra of Si spherical particles with the radius R located in air; (b) extinction and scattering spectra of a Si particle with $R=65\text{ nm}$; (c) schematic of the square array of Si cylinders on a SiO_2 substrate embedded within polymer; (d) electric-field-intensity enhancement spectra of a $P=700\text{ nm}$ array, averaged over three different horizontal monitor planes, insets are field-magnitude enhancement maps at resonance peaks; (e) electric-field-intensity enhancement spectra of a $P=1035\text{ nm}$ array; (f) electric-field-intensity enhancement spectra of a $P=1500\text{ nm}$ array

介质纳米颗粒阵列还可实现其他类型的高Q共振如连续域束缚态(BIC)。BIC是开放腔的非辐射本征模,在连续谱内其品质因子可以无限大,常见的类型如“受对称性保护”的BIC因其模式具有反对称性,不能与远场辐射耦合(暗态),这在早期光子晶体平板的研究中就有提到^[31-33]。双颗粒阵列可提供较多自由度来调控暗态,使之成为准暗态。台湾交通大学陈国平团队^[34]设计出 Si_3N_4 双颗粒阵列,使BIC与SLR波长相近,如图8(a)、(b)所示,该反对称的BIC模式可看作是两个具有反相电偶极子共振类型的SLR叠加。破坏双颗粒的对称性,BIC转变为准暗态,电场强度逐渐降低[图8(b)]。此时,为了使准暗态的波长基本保持不变[图8(a)],他们在缩小一个颗粒尺寸的同时增大另一个颗粒的尺寸[图8(c)],保持总的填充因子和散射截面不变,从而可系统地研究双颗粒非对称性对阵列的品质因子,以及作为谐振反馈用途时对激光阈值的影响。

4 基于连续金属膜上周期介质阵列的SLR

在垂直入射激发条件下,金属及介质纳米颗粒阵

列的衍射耦合要求阵列所处环境的折射率均匀分布,且非对称的折射率环境会显著降低SLR的Q因子甚至会破坏SLR。此外,辐射损耗可以被抑制,吸收损耗却是材料固有的。材料的吸收损耗限制了金属及高折射率介质结构在可见光波段的应用。从紫外到中红外波段都有中/低折射率介质($n<2.5$)存在,并在宽波长范围展现出极低的吸收损耗^[35],如常用 SiO_2 制备具有超高品质因子的光学微腔^[36]。

为使局域光场离开金属表面,从而降低金属吸收损耗带来的不良影响,本课题组提出一种在连续金属膜上加载低折射率介质纳米柱阵列的方法[图9(a)],并在非对称折射率环境实现了SLR^[37-39]。在该结构中,金属膜作为反射镜,增强的电磁场被局域在介质柱上方,远离了金属表面,因此相对于局域在金属表面的等离子体共振,金属吸收损耗所产生的影响较小。采用约100 nm厚的金属膜(金、银、铝)作为反射镜,通过软压印技术制备出高深宽比的SU-8(一种在可见、近红外波段透明的聚合物,折射率 $n\approx1.58$)纳米柱阵列[图9(b)],获得较高的Q因子[$Q=320$,图9(c)]^[38-39]。此外,介质柱顶端的场增强与相应金纳米颗粒阵列中

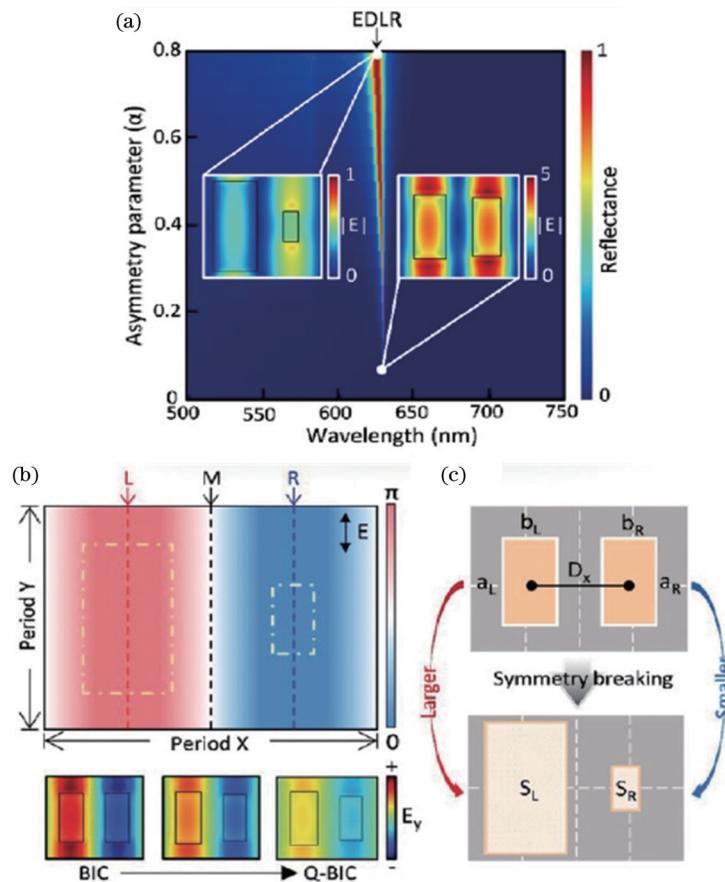


图 8 介质双颗粒阵列的 BIC 及其与 SLR 的联系^[34]。(a)计算的反射光谱,插图显示结构非对称因子 $\alpha=0.8$ 和 0.05 的近场分布;(b)反对称 BIC 模式的相位分布;(c)破坏双颗粒对称性的设计

Fig. 8 BIC in arrays of double dielectric particles and its connection with SLR^[34]. (a) Simulated reflectance spectra, the insets display the near-field distribution for asymmetry parameter $\alpha=0.8$ and 0.05; (b) phase distribution color map of asymmetry BICs; (c) design for breaking the symmetry

颗粒表面的场增强程度相当[图 9(d)]。尽管介质折射率较小,但是阵列中介质柱顶端的电场分布表现出电偶极子共振的特征。该结构不仅可调控内嵌染料分子的荧光辐射,并加快辐射衰减速率,还可提升单层 WSe₂的发光强度。介质纳米柱与背景环境的折射率比值约为 1.5,在可见光及近红外波段,这种结构设计可以推广到诸如 TiO₂、Si₃N₄纳米柱置于 SiO₂、透明聚合物、水溶液等背景环境中。该结构本身的折射率环境是非对称的,其处于非均匀分布的折射率环境也能维持高品质表面共振。在金膜上制备了 560 nm 周期的 TiO₂纳米柱阵列,并将其置于水、有机溶剂、SU-8 等背景环境中[图 9(e)~(g)],发现 SLR 能适应较大范围的背景折射率变化并可维持较窄的共振线宽。因此,可在完成制备后通过调节背景折射率来调控共振波长,或者实现光学折射率传感^[40]。低损耗的低折射率介质普遍存在,选取合适折射率比值的介质材料以及高反镜材料,可以实现高品质 SLR 在紫外到中红外波段的宽范围调谐。例如,在中红外波段可以铜为反射镜,原则上可以基于该原理实现中红外波段的窄带热辐射源^[41]。

在反射镜上加载介质纳米柱阵列形成 SLR,介质纳米柱与背景环境之间的折射率比值实际上还可以更大。中国科学院深圳先进技术研究院李光元课题组^[42]计算了空气中的中等折射率介质柱阵列,通过优化纳米柱的深宽比,发现 SLR 的 Q 因子可达数万。如图 10(a)所示,对于固定阵列参数($P=720$ nm, $d=180$ nm, $h=460$ nm):当介质柱的折射率较低($n_D=1.8$ 或 1.9)时,介质纳米柱的散射效率低,SLR 几乎不能被激发;当 n_D 从 2.0 增大到 2.2 时,SLR 的 FWHM 增大,Q 因子降低[图 10(d)];当 n_D 固定为 2.0 时,较小尺寸的介质纳米柱阵列($h=460$ nm, $d=160$ nm)的散射效率较低,随着直径增大 [$d=180\sim240$ nm, 图 10(b)]或高度增加 [$h=440\sim480$ nm, 图 10(c)],SLR 的 FWHM 增大,Q 因子降低[图 10(e)、(f)]。

当然,反射镜上的介质阵列结构不局限于纳米圆柱,其他形貌的颗粒(如纳米球),以及由纳米柱构成的二聚体等通过合理的周期排列也能获得高品质 SLR。图 11 所示为平面金膜上的纳米柱二聚体阵列的 SLR 电场强度分布^[37]。与单个纳米柱阵列的 SLR 类似,纳米柱二聚体阵列的 SLR 电场也局域在纳米柱顶端,二

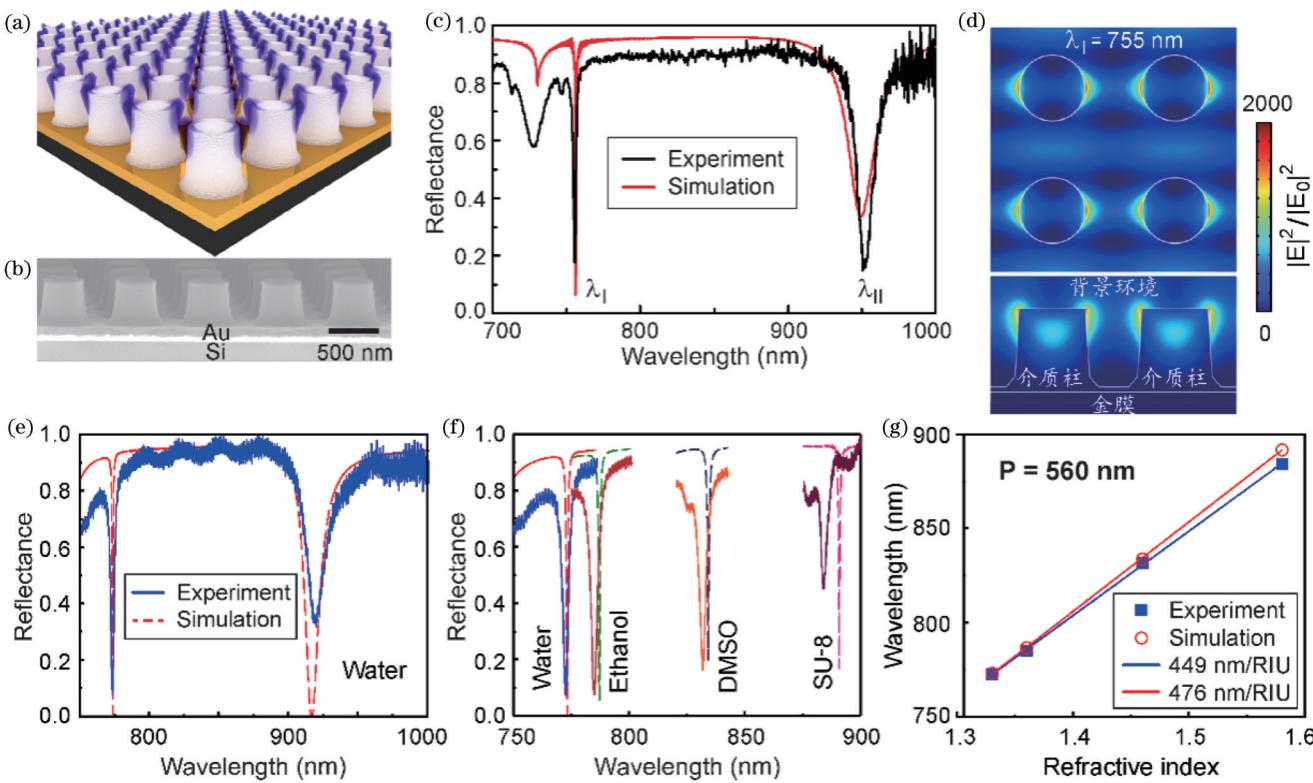


图9 反射镜上介质纳米柱阵列的SLR^[38,40]。(a)金膜上介质纳米柱的方形周期阵列示意图;(b)软压印制备的720 nm周期SU-8纳米柱阵列扫描电镜图;(c)垂直反射谱, λ_I 处的窄线宽特征对应介质表面的晶格共振模式;(d)SLR模式的电场被局域在介质柱顶端;(e)金膜上TiO₂纳米柱阵列置于水背景时的反射谱;(f)金膜上TiO₂纳米柱阵列置于不同折射率背景中的SLR;(g)金膜上TiO₂纳米柱阵列的共振波长与背景折射率的关系

Fig. 9 SLR supported by mirror-backed dielectric nanopillar arrays^[38,40]. (a) Schematic showing SLR supported by a periodic array of dielectric pillars on a gold film; (b) SEM image of SU-8 pillar array (lattice spacing is 720 nm) on a gold-coated silicon substrate; (c) reflection spectra at normal incidence and the narrow linewidth characteristic at λ_I corresponds to the lattice resonance mode on the medium surface; (d) electric field intensity distribution on the top surface and cross section of the pillar array at $\lambda_{\text{SLR}}=755 \text{ nm}$; (e) reflection spectra of SLR supported by TiO₂ pillar array (lattice spacing is 560 nm) on a gold-coated silicon substrate in water; (f) reflection spectra around SLR supported by TiO₂ pillar array (lattice spacing is 560 nm) on a gold-coated silicon substrate in different media; (g) relation between λ_{SLR} and background refractive indices of SLR supported by TiO₂ pillar array (lattice spacing is 560 nm) on a gold-coated silicon substrate

聚体中间的空隙可以提供较大的场增强区域,以增强与背景环境物质的相互作用,并且二聚体中间的间隙尺寸在100 nm量级(周期为720 nm)还能维持较大的电场强度提升($|E|^2/|E_0|^2 \approx 2000$)。合肥工业大学的Hu等^[43]进一步提出利用反射镜上非对称纳米柱二聚体阵列在两个正交方向上的超窄SLR实现窄带圆二色性增强。

5 总结与展望

介绍了在垂直入射激发条件下基于常见金属和介质纳米颗粒阵列、金属膜上周期介质阵列的高Q因子SLR。尽管在理论上可以设计出支持高Q因子SLR的贵金属纳米颗粒阵列,但制备技术的不足阻碍了高Q因子等离激元SLR的实验实现。由于介质材料本身吸收损耗比金属要低很多,因此介质纳米颗粒阵列中的SLR对颗粒表面粗糙度、形貌不规则等的敏感性

也比金属颗粒阵列低很多。半导体微纳加工工艺现已非常成熟,硅和Ⅲ-V族半导体等材料的加工可以达到纳米级的分辨率和精度,因此完全能够在材料本身的透明波段实现极高Q因子的全介质SLR。在可见光波段,金属及大多数高折射率介质都有明显的吸收损耗,而金属膜上高深宽比透明介质纳米柱阵列在实现高Q因子SLR方面有很大潜力,但是需要发展高效精确的制备方法。高Q因子SLR在传感、光学滤波、光开关和调制、荧光增强、微纳激光和非线性光学等领域有广泛应用^[10, 44-46]。

目前SLR的研究对象还包括磁性材料^[47-48]和二维材料^[49-51]等纳米颗粒阵列。引入磁性材料,一方面有望在微纳尺度通过磁场打破时间反演对称性,另一方面有望实现对磁光响应的共振增强。层状二维过渡金属硫化物材料在可见光波段表现出很高的折射率,并在室温下具有较大的激子结合能,相对于传统的半导

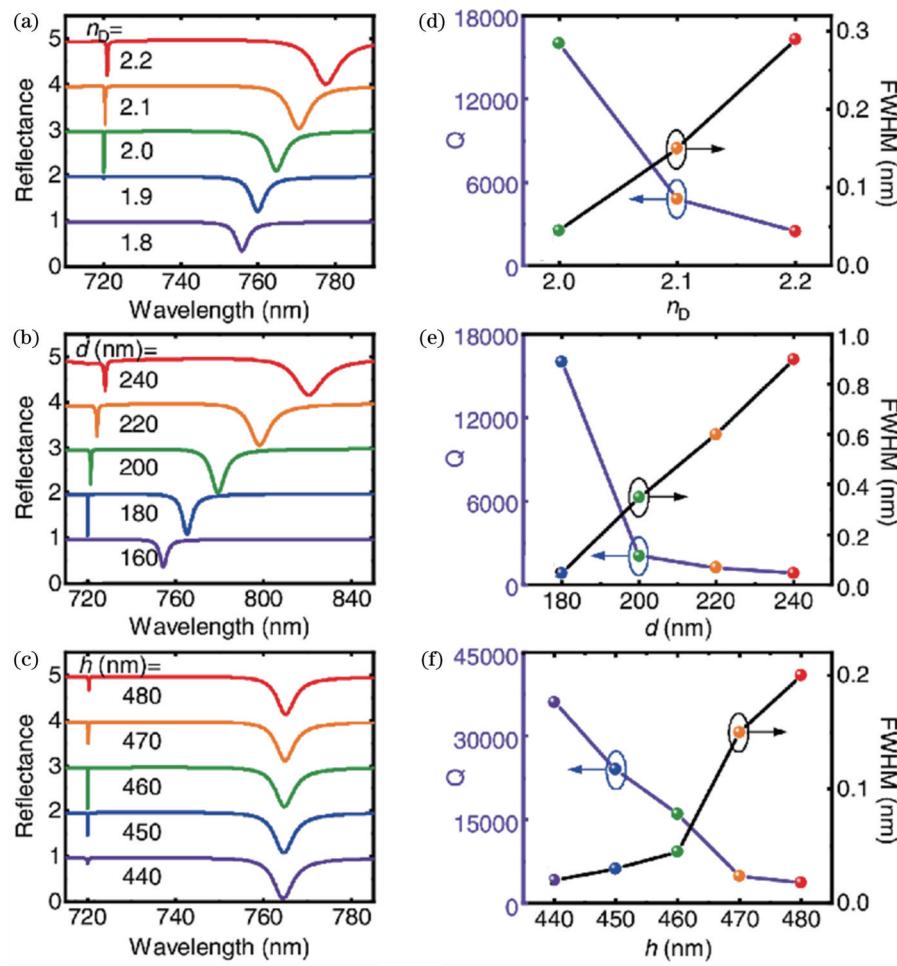


图10 反射镜上中等折射率介质纳米柱阵列的高Q因子SLR($P=720$ nm)^[42]。(a)(d)介质柱折射率变化时的反射光谱、SLR品质因子和FWHM;(b)(e)介质柱直径变化时的反射光谱、SLR品质因子和FWHM;(c)(f)介质柱高度变化时的反射光谱、SLR品质因子和FWHM

Fig. 10 High-Q SLR supported by mirror-backed dielectric nanopillars with medium refractive index ($P=720$ nm)^[42]. (a)(d) Reflectance spectra, quality factors, and FWHM of SLR for different refractive index; (b)(e) reflectance spectra, quality factors, and FWHM of SLR for different diameters; (c)(f) reflectance spectra, quality factors, and FWHM of SLR for different height

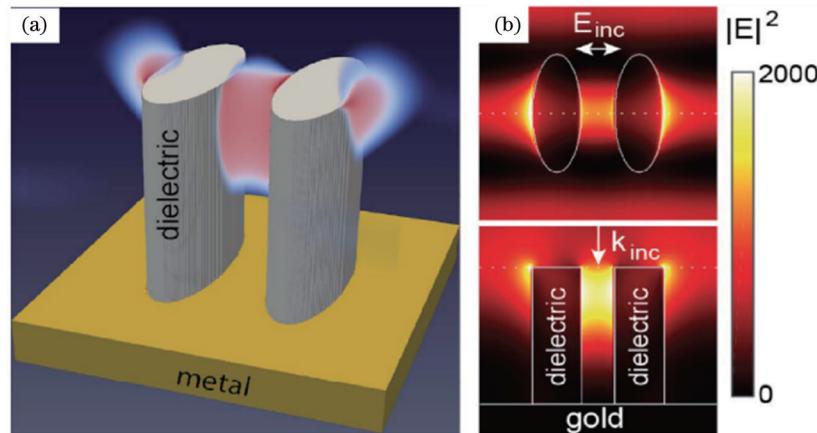


图11 反射镜上介质柱二聚体阵列的SLR^[37]。(a)镜上介质柱二聚体结构单元示意图;(b)SLR处的电场强度分布
Fig. 11 SLR supported by mirror-backed dielectric dimer arrays^[37]. (a) Schematic of dielectric-dimer-on-mirror structures; (b) electric field intensity distribution at SLR

体纳米材料展现出更大的非线性极化率。然而,由于材料本身的限制,目前基于这些材料的SLR线宽都比较宽,例如,磁性金属材料镍相比于贵金属具有更大的吸收损耗。为进一步减小共振线宽,其中一个策略是改变激发方式,可以将这些材料的纳米颗粒阵列与其他高品质因子共振机理例如导波共振相结合,以减轻或避开材料本征吸收损耗的不利影响。拓展相关研究的材料对象也是重要的研究方向,例如,探索具有绝缘体态和导电表面态的拓扑绝缘体^[52]等新兴材料。此外,SLR的动态调控方面也需进一步深入研究。先前人们展示了通过机械形变改变晶格周期^[53-54]、换背景溶液改变背景折射率^[55-56]、光致变色改变吸收损耗^[57-58]等方式调控SLR,但这些方式都存在响应慢的不足。最近希伯来大学Levy课题组^[59]将铝纳米颗粒阵列夹在铌酸锂薄膜与氧化铟锡(ITO)之间,通过铌酸锂的线性电光效应实现了对SLR波长的调谐。未来将有望通过改善制备工艺、结合新的器件原理来优化电光调制/调谐性能,例如电光聚合物可以有比铌酸锂更高的电光系数。

参 考 文 献

- [1] Fort E, Grésillon S. Surface enhanced fluorescence[J]. *Journal of Physics D*, 2008, 41(1): 013001.
- [2] Langer J, de Aberasturi D J, Aizpurua J, et al. Present and future of surface-enhanced raman scattering[J]. *ACS Nano*, 2020, 14(1): 28-117.
- [3] Stiles P L, Dieringer J A, Shah N C, et al. Surface-enhanced Raman spectroscopy[J]. *Annual Review of Analytical Chemistry*, 2008, 1: 601-626.
- [4] Neubrech F, Huck C, Weber K, et al. Surface-enhanced infrared spectroscopy using resonant nanoantennas[J]. *Chemical Reviews*, 2017, 117(7): 5110-5145.
- [5] Koenderink A F, Alù A, Polman A. Nanophotonics: shrinking light-based technology[J]. *Science*, 2015, 348(6234): 516-521.
- [6] Khurjin J B. How to deal with the loss in plasmonics and metamaterials[J]. *Nature Nanotechnology*, 2015, 10(1): 2-6.
- [7] Kuznetsov A I, Miroshnichenko A E, Brongersma M L, et al. Optically resonant dielectric nanostructures[J]. *Science*, 2016, 354(6314): aag2472.
- [8] Baranov D G, Zuev D A, Lepeshov S I, et al. All-dielectric nanophotonics: the quest for better materials and fabrication techniques[J]. *Optica*, 2017, 4(7): 814-825.
- [9] Grinblat G, Li Y, Nielsen M P, et al. Enhanced third harmonic generation in single germanium nanodisks excited at the anapole mode[J]. *Nano Letters*, 2016, 16(7): 4635-4640.
- [10] Kravets V G, Kabashin A V, Barnes W L, et al. Plasmonic surface lattice resonances: a review of properties and applications [J]. *Chemical Reviews*, 2018, 118(12): 5912-5951.
- [11] Evlyukhin A B, Reinhardt C, Seidel A, et al. Optical response features of Si-nanoparticle arrays[J]. *Physical Review B*, 2010, 82(4): 045404.
- [12] Auguié B, Bendana X M, Barnes W L, et al. Diffractive arrays of gold nanoparticles near an interface: critical role of the substrate[J]. *Physical Review B*, 2010, 82(15): 155447.
- [13] Wang X W, Kogos L C, Paiella R. Giant distributed optical-field enhancements from Mie-resonant lattice surface modes in dielectric metasurfaces[J]. *OSA Continuum*, 2018, 2(1): 32-42.
- [14] Chu Y Z, Schonbrun E, Yang T A, et al. Experimental observation of narrow surface plasmon resonances in gold nanoparticle arrays[J]. *Applied Physics Letters*, 2008, 93(18): 181108.
- [15] Laux F, Bonod N, Gérard D. Single emitter fluorescence enhancement with surface lattice resonances[J]. *The Journal of Physical Chemistry C*, 2017, 121(24): 13280-13289.
- [16] Bin-Alam M S, Reshef O, Mamchur Y, et al. Ultra-high-Q resonances in plasmonic metasurfaces[J]. *Nature Communications*, 2021, 12: 974.
- [17] Deng S K, Li R, Park J E, et al. Ultranarrow plasmon resonances from annealed nanoparticle lattices[J]. *Proceedings of the National Academy of Sciences of the United States of America*, 2020, 117(38): 23380-23384.
- [18] Yang F, Chen Q Y, Wang J J, et al. Fabrication of centimeter-scale plasmonic nanoparticle arrays with ultranarrow surface lattice resonances[J]. *ACS Nano*, 2023, 17(1): 725-734.
- [19] Humphrey A D, Meinzer N, Starkey T A, et al. Surface lattice resonances in plasmonic arrays of asymmetric disc dimers[J]. *ACS Photonics*, 2016, 3(4): 634-639.
- [20] Cuartero-González A, Sanders S, Zundel L, et al. Super- and subradiant lattice resonances in bipartite nanoparticle arrays[J]. *ACS Nano*, 2020, 14(9): 11876-11887.
- [21] Li R, Bourgeois M R, Cherqui C, et al. Hierarchical hybridization in plasmonic honeycomb lattices[J]. *Nano Letters*, 2019, 19(9): 6435-6441.
- [22] Zundel L, May A, Manjavacas A. Lattice resonances induced by periodic vacancies in arrays of nanoparticles[J]. *ACS Photonics*, 2021, 8(1): 360-368.
- [23] Lim T L, Vaddi Y, Bin-Alam M S, et al. Fourier-engineered plasmonic lattice resonances[J]. *ACS Nano*, 2022, 16(4): 5696-5703.
- [24] Li R, Wang D Q, Guan J, et al. Plasmon nanolasers with aluminum nanoparticle arrays[J]. *Journal of the Optical Society of America B*, 2019, 36(7): E104-E111.
- [25] Guan J, Sagar L K, Li R, et al. Engineering directionality in quantum dot shell lasing using plasmonic lattices[J]. *Nano Letters*, 2020, 20(2): 1468-1474.
- [26] Guan J, Sagar L K, Li R, et al. Quantum dot-plasmon lasing with controlled polarization patterns[J]. *ACS Nano*, 2020, 14(3): 3426-3433.
- [27] Huttunen M J, Reshef O, Stolt T, et al. Efficient nonlinear metasurfaces by using multiresonant high-Q plasmonic arrays[J]. *Journal of the Optical Society of America B*, 2019, 36(7): E30-E35.
- [28] Stolt T, Vesala A, Rekola H, et al. Multiply-resonant second-harmonic generation using surface lattice resonances in aluminum metasurfaces[J]. *Optics Express*, 2022, 30(3): 3620-3631.
- [29] Babicheva V E, Evlyukhin A B. Resonant lattice kerker effect in metasurfaces with electric and magnetic optical responses[J]. *Laser & Photonics Reviews*, 2017, 11(6): 1700132.
- [30] Murai S, Castellanos G W, Raziman T V, et al. Enhanced light emission by magnetic and electric resonances in dielectric metasurfaces[J]. *Advanced Optical Materials*, 2020, 8(16): 1902024.
- [31] Ochiai T, Sakoda K. Dispersion relation and optical transmittance of a hexagonal photonic crystal slab[J]. *Physical Review B*, 2001, 63(12): 125107.
- [32] Pacradouni V, Mandeville W J, Cowan A R, et al. Photonic band structure of dielectric membranes periodically textured in two dimensions[J]. *Physical Review B*, 2000, 62(7): 4204-4207.
- [33] Fan S H, Joannopoulos J D. Analysis of guided resonances in photonic crystal slabs[J]. *Physical Review B*, 2002, 65(23): 235112.
- [34] Yang J H, Huang Z T, Maksimov D N, et al. Low-threshold bound state in the continuum lasers in hybrid lattice resonance metasurfaces[J]. *Laser & Photonics Reviews*, 2021, 15(10): 2100118.
- [35] Kischkat J, Peters S, Gruska B, et al. Mid-infrared optical

- properties of thin films of aluminum oxide, titanium dioxide, silicon dioxide, aluminum nitride, and silicon nitride[J]. *Applied Optics*, 2012, 51(28): 6789-6798.
- [36] Zhang X Y, Cao Q T, Wang Z, et al. Symmetry-breaking-induced nonlinear optics at a microcavity surface[J]. *Nature Photonics*, 2019, 13(1): 21-24.
- [37] Ao X Y. Surface mode with large field enhancement in dielectric-dimer-on-mirror structures[J]. *Optics Letters*, 2018, 43(5): 1091-1094.
- [38] Ao X Y, Wang D Q, Odom T W. Enhanced fields in mirror-backed low-index dielectric structures[J]. *ACS Photonics*, 2019, 6(11): 2612-2617.
- [39] Ao X Y, Xu X N, Dong J W, et al. Unidirectional enhanced emission from 2D monolayer suspended by dielectric pillar array [J]. *ACS Applied Materials & Interfaces*, 2018, 10(41): 34817-34821.
- [40] Dong J W, Chen S, Huang G F, et al. Low-index-contrast dielectric lattices on metal for refractometric sensing[J]. *Advanced Optical Materials*, 2020, 8(21): 2000877.
- [41] Wu Q, Wang L F, Ao X Y. Narrowband mid-infrared absorber based on a mirror-backed low-index dielectric lattice[J]. *Journal of the Optical Society of America B*, 2021, 38(8): 2306-2311.
- [42] Wang Y H, Xiong L, Tian M, et al. Mirror-backed dielectric metasurface sensor with ultrahigh figure of merit based on a super-narrow Rayleigh anomaly[J]. *Applied Optics*, 2021, 60(36): 11205-11210.
- [43] Hu J G, Xiao Y X, Zhou L M, et al. Ultra-narrow-band circular dichroism by surface lattice resonances in an asymmetric dimer-on-mirror metasurface[J]. *Optics Express*, 2022, 30(10): 16020-16030.
- [44] Utyshev A D, Zakomirnyi V I, Rasskazov I L. Collective lattice resonances: plasmonics and beyond[J]. *Reviews in Physics*, 2021, 6: 100051.
- [45] Wang D Q, Guan J, Hu J T, et al. Manipulating light-matter interactions in plasmonic nanoparticle lattices[J]. *Accounts of Chemical Research*, 2019, 52(11): 2997-3007.
- [46] Wang B Q, Yu P, Wang W H, et al. High-Q plasmonic resonances: fundamentals and applications[J]. *Advanced Optical Materials*, 2021, 9(7): 2001520.
- [47] Pourjamal S, Hakala T K, Nečada M, et al. Lasing in Ni nanodisk arrays[J]. *ACS Nano*, 2019, 13(5): 5686-5692.
- [48] Kataja M, Hakala T K, Julku A, et al. Surface lattice resonances and magneto-optical response in magnetic nanoparticle arrays[J]. *Nature Communications*, 2015, 6: 7072.
- [49] Verre R, Baranov D G, Munkhbat B, et al. Transition metal dichalcogenide nanodisks as high-index dielectric Mie nanoresonators[J]. *Nature Nanotechnology*, 2019, 14(7): 679-683.
- [50] Shen F H, Zhang Z H, Zhou Y Q, et al. Transition metal dichalcogenide metaphotonic and self-coupled polaritonic platform grown by chemical vapor deposition[J]. *Nature Communications*, 2022, 13: 5597.
- [51] Babicheva V E, Moloney J V. Lattice resonances in transdimensional WS₂ nanoantenna arrays[J]. *Applied Sciences*, 2019, 9(10): 2005.
- [52] Lu H A, Yue Z J, Li Y W, et al. Magnetic plasmon resonances in nanostructured topological insulators for strongly enhanced light-MoS₂ interactions[J]. *Light: Science & Applications*, 2020, 9: 191.
- [53] Wang D Q, Bourgeois M R, Lee W K, et al. Stretchable nanolasing from hybrid quadrupole plasmons[J]. *Nano Letters*, 2018, 18(7): 4549-4555.
- [54] Yang A K, Hryniak J, Bourgeois M R, et al. Programmable and reversible plasmon mode engineering[J]. *Proceedings of the National Academy of Sciences of the United States of America*, 2016, 113(50): 14201-14206.
- [55] Abass A, Rodriguez S R K, Ako T, et al. Active liquid crystal tuning of metallic nanoantenna enhanced light emission from colloidal quantum dots[J]. *Nano Letters*, 2014, 14(10): 5555-5560.
- [56] Yang A K, Hoang T B, Dridi M, et al. Real-time tunable lasing from plasmonic nanocavity arrays[J]. *Nature Communications*, 2015, 6: 6939.
- [57] Chen S A, Huang X C, Wu Q, et al. Photochromic switching of narrow-band lattice resonances[J]. *Optics Letters*, 2022, 47(2): 337-340.
- [58] Taskinen J M, Moilanen A J, Rekola H, et al. All-optical emission control and lasing in plasmonic lattices[J]. *ACS Photonics*, 2020, 7(10): 2850-2858.
- [59] Weiss A, Frydendahl C, Bar-David J, et al. Tunable metasurface using thin-film lithium niobate in the telecom regime [J]. *ACS Photonics*, 2022, 9(2): 605-612.

High-*Q* Surface Lattice Resonances

Du Yixuan*, Ao Xianyu*, Cai Yangjian**

School of Physics and Electronics, Shandong Normal University, Jinan 250358, Shandong, China

Abstract

Significance High-*Q* resonances that confine the light energy at subwavelength scales have applications in various fields such as micro/nano-lasers, fluorescence enhancement, and optical sensing. Extreme light localization has been realized by surface plasmons squeezed in plasmonic nanogaps, whereas there is intrinsic energy dissipation by electron oscillations on metal surfaces. In contrast, high-index dielectric nanostructures supporting Mie-type electric and magnetic resonances exhibit low optical dissipation but only moderate field confinement. When plasmonic or high-index dielectric nanoparticles are arranged into periodic arrays, diffractive coupling in the plane of periodic arrays may occur. This can suppress the radiative damping of individual nanoparticles, and produce surface lattice resonance (SLR) modes with significantly higher ($|E|^2/|E_0|^2 > 10^3$) field enhancements and much higher quality factors compared with isolated nanoparticles.

The last two decades have seen significant progress in SLRs supported by metallic and high-index dielectric nanoparticle arrays under normal incident excitation. However, due to limitations in the involved materials and available

nanofabrication methods, there is still a series of challenges in achieving a high Q -factor in the visible regime, especially in asymmetric refractive-index environments. Thus, it is necessary to summarize the existing studies to guide the future development of this field more rationally.

Progress We first introduce the basic properties of SLRs in metallic and high-index dielectric nanoparticle arrays under normal incident excitation. The periodic lattices are usually generated by various top-down lithography methods. The difficulty in experimentally achieving SLRs with a high Q -factor from noble metal nanoparticle arrays is that the precise fabrication of defect-free nanoparticle arrays is hard. One strategy to overcome optical dissipation and reduce the linewidth of SLRs is to shrink the particle size relative to the lattice spacing. Reshef *et al.* at the University of Ottawa reported a Q -factor of 2340 in the telecommunication C band, and this is the ever reported highest value (Fig. 2). Another strategy is to make the particles smooth and uniform. Odom *et al.* at Northwestern University reported that thermal annealing can improve the uniformity, surface roughness, and crystallinity of metal nanoparticles produced by physical vapor deposition methods, which can lead to SLRs with dramatically improved Q factors (Fig. 3). Nie *et al.* from Fudan University proposed a method to produce metal nanoparticle arrays by combining solvent-assisted soft lithography and wet chemical with annealing processes, and thus a metal deposition process in a vacuum is not required (Fig. 4). Furthermore, SLRs can also be supported by arrays composed of complex basis or localized surface plasmons with multipolar characteristics, and these arrays show much richer optical responses compared with arrays with only one particle in a unit cell (Figs. 5 and 6). Arrays of high-index dielectric nanoparticles can support SLRs with characteristics of magnetic dipole (MD) besides electric dipole (ED), and both types of SLRs can be tuned independently (Fig. 7). By choosing lattice periods independently in each mutually perpendicular direction, Babicheva *et al.* from Georgia State University found that it is possible to make the ED-SLR and MD-SLR overlapped in a certain spectral range, which leads to the resonant suppression of the backward scattering (lattice Kerker effect).

Subsequently, we summarize the progress in achieving high- Q SLRs based on mirror-backed high-aspect-ratio dielectric nanopillar arrays in asymmetric refractive-index environments (Fig. 9). In this hybrid system, dielectric nanopillars are arranged periodically on an optically thick metal film, which blocks the light transmission completely. Therefore, the issue of a symmetric dielectric environment between the substrate and the upper cladding does not exist, in contrast to the requirement of a symmetric environment for realizing sharp lattice resonances in all-plasmonic or all-dielectric systems. Meanwhile, the electric field enhancements are comparable to lattice plasmon modes from arrays of noble metal nanoparticles, but with strongly reduced plasmonic dissipation, since the enhanced fields are away from the metal surface. The narrow linewidth resonances can be tuned over a wide wavelength range from ultraviolet to mid-infrared by simply scaling the dielectric lattices and combining them with appropriate highly reflective metals. Additionally, numerical simulations show that it is possible to achieve a Q -factor of tens of thousands on this hybrid platform (Fig. 10).

Conclusions and Prospects SLRs arise from the diffractive coupling in periodic arrays, which can theoretically achieve a high Q -factor and greatly enhance the interactions between light and matter in the background media. This prominence has brought about the development of potentially practical devices for optoelectronics, biosensing, and other applications, using common materials such as noble metals and transparent dielectrics. Nanoparticle arrays of other functional materials like magnetic metals and newly emerging materials such as two-dimensional layered materials still need new design principles to mitigate their intrinsic optical dissipation to achieve high-quality surface lattice resonances with fascinating properties.

Key words surface mode; diffraction; surface plasmon; Mie scattering