超常材料光波吸收體之簡介

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一、前言

近年來,由於溫室效應及石油危機,開發綠色能源逐漸成為世界關注之焦點。其中根據光 伏(photovoltaic)原理所發展的太陽能電池(solar cells),更是綠色能源中極重要的一員。然而,光 對電的能量轉換效率一直是太陽能電池相當大的瓶頸。而在影響能源轉換效率的各個因素中, 如何使光波有效地被材料所吸收,更是最關鍵的一環[1]。由於平板型超常材料(planar metamaterials)[2]利用其特殊電磁場共振原理,如圖一所示,可作為光吸收器(absorber)[3-6],使 光子吸收率提高;或是藉由設計組成超常材料之次波長結構,達到光捕捉(light trapping)之目的, 使光侷限在材料中,增加光路長度(optical path length),使光子吸收機率提升[7]。因此可利用平 板型超材料製作太陽能電池,提昇能量轉換效率。



圖一 超常材料完美吸收體。左圖:單位晶格之幾何結構。右圖:反射、穿透與吸收頻譜[3]

超常材料(metamaterials)[8]是由人工製造之次波長結構,在某些特別的排列下,在巨觀尺度 具有自然界不常見之物理性質,例如負磁導係數[9]及負折射率[10]等。利用這些特殊的物理特 性,可以超越電磁波或光波的繞射極限(diffraction limit),達到光學超解析(super resolution)的目 的,製作所謂的完美透鏡(perfect lens)[11]、超透鏡(superlens)及雙曲透鏡(hyperlens)等。超常材料 甚至可能製作出自然界不存在之非均勻(inhomogeneous)、非等向性(anisotropic)介質而達到隱形 (cloaking)[12,13]之功能。另一方面,超常材料具有異常光穿透(extraordinary optical transmission, EOT)[14]、異常光吸收(extraordinary optical absorption, EOA)[15]等特殊濾波性質。此外,除了電 磁波之超常材料外亦有聲波、水波、彈性波及量子波等領域之超常材料。這些多樣化的成果使得 超常材料的研究維持在重要的地位,並且在奈米製程與顯微量測技術[16,17]的不斷進步之下, 繼續開發其可能與潛在的應用。

超常材料通常是由次波長的週期結構(subwavelength periodic structure)所組成[18];因此,超 常材料之特性與準靜態(quasistatic)之共振機制有密切關聯[19,20]。由於次波長週期結構之共振波 長與幾何形狀有關,因此可藉由設計單位晶格(unit cell)的次波長結構形狀與排列,使平板超常材 料具有特殊之反射、穿透與吸收頻譜。一般常見的次波長週期結構,其基本幾何形狀有帶狀 (stripes)[21]、環狀(loops)[22]、孔洞(slits)[23],以及分裂共振環(split-ring resonator)[24-26]等。

通常在單位晶格中若只包含單一次波長結構,其共振頻譜可由 Lorentzian 共振(Lorentizian resonance)的標準線型來描述[27]

$$\mid r \mid = \sqrt{\frac{\gamma}{\left(\epsilon^2 + \gamma\right)}}$$

其中|r|為反射係數之大小, γ 為頻寬, $\epsilon = \omega - \omega_L$, ω_L 為 Lorentzian 共振頻率位置。圖二為 $\omega_L = 0.5$ 時不同 γ 值之 Lorentzian 共振之反射係數圖。



圖二 Lorentzian 共振之頻譜線型

此種由個別單一結構所產生的共振,又稱為 Mie 共振(Mie resonance)或部位共振(site resonance); 另一方面,由於結構具有週期性,因此隨著入射頻率增加,當高階繞射由消散波(evanescent wave) 開始沿著表面傳播時,會產生所謂的晶格共振(lattice resonance)或稱為 Bragg 共振(Bragg resonance),例如 Wood 異常現象(Wood's anomaly)[28-30]。通常晶格共振只與晶格常數及晶格型 態相關,與個別次波長結構之幾何形狀無關。



圖三 表面電漿示意圖[31]

金屬為次波長週期結構所經常使用之材料,而金屬與介電體的表面可以存在表面電漿共振 (surface plasmon resonance)的特性[31]。表面電漿是在導體(通常是金屬)表面傳遞的一種電磁波(或 光波)[32,33],它來導體內自由電子的集體振動與外界電磁波的偶合。由於其具有雙重性質,波 動型式的表面電漿既是橫波(電磁波的本質),又是縱波(電子波的本質)[見圖三];量子化的表面電 漿子又稱為偏極子(polariton)[34],也就是光子與電子所結合形成的粒子。因為表面電漿結合了電 子的振動,其動量比相同頻率的光波大,所以在一般情形下表面電漿是無法被激發的。必須有其 他裝置或結構的輔助,才能藉由共振(即能量與動量的相配,或頻率與波數的相配)來產生表面電 漿。這個激發機制稱為耦合(coupling)。常用的耦合方法有稜鏡偶合(prism coupling)、格柵偶合 (grating coupling)、孔洞偶合(aperture coupling)及晶格偶合(lattice coupling)等[35]。利用光速的減 低(頻率不變,因此波長減小,波數增大),或額外動量的提供(來自結構的多重散射),可以補足 動量差額來激發表面電漿。因此,表面電漿的特性不單與金屬的性質有關,還和結構的型態有密 切關係。

表面電漿的重要基本性質之一為其波動特性,這可由其色散關係(dispersion relation),即頻 率與波數(wave number)的關係來表明。頻率是時間變化的參數,波數是空間變化的參數;時間 與空間的變化描述了波動的基本特性。由於表面電漿的光學或電磁特性與金屬的材質及結構的 型態(如維度或形狀)有關,因此表面電漿的色散關係同時取決於材料色散(material dispersion)與 結構色散(structure dispersion)。前者來自金屬內自由電子的運動,後者則來自電磁波在結構中的 散射(scattering)行為。這兩種色散性質的結合,構成了表面電漿的基本波動性質[36-38]。另外, 表面電漿也是光穿遂(optical tunneling)的主要機制[39-41]。

此外表面電漿的波動特性,可進一步由其特徵模態(eigenmode)來闡明。例如使用表面電漿 來進行光吸收或光收穫(light harvest)機制,可以在高密度態(density of state)之區域設計操作。如 此可充分利用表面電漿在頻率與空間的極不均匀特性,有效率的吸收與使用光波能量,進行光 轉換及發送功能[42]。另外,利用介電-介電材料之耦合,產生類似波導之波導模態共振(guided mode resonance) [43, 44],或是共振腔模態(cavity resonance)[45],使光被捕捉(light trapping)在材 料中。

當多種(兩種或以上)共振模態耦合時,反射係數之大小之頻譜可利用 Fano 共振(Fano resonance)線型來描述[46,47]

$$|r| = \sqrt{\frac{1}{1+q^2} \frac{(\varepsilon+q)^2}{1+\varepsilon^2}}$$

其中q為非對稱參數、 $\epsilon = 2(\omega - \omega_F) / \Gamma \cdot \omega_F$ 為 Fano 共振頻率。此種多重共振耦合之現象稱為 Fano 共振[48]。



圖四 Fano 共振之頻譜線型

因此決定平面超常材料特性之共振種類可歸納如下:(1)單一次波長結構之幾何形狀產生之 Mie 共振,(2)週期結構產生之 Bragg 共振,(3)金屬-介電材料耦合產生之表面電漿共振,(4)介 電-介電材料耦合產生之波導共振,及(5)多種共振模態耦合產生之 Fano 共振。

另一方面,由於微結構尺寸小於波長,因此可使用等效介質(effective medium)理論,計算 及分析平板超常材料之等效介電常數、等效透磁係數之等效物理量[49]。此外,次波長週期結 構之共振系統亦可利用等效電路(equivalent circuit)模式來分析其共振頻譜,經由計算各種幾何 形狀之等效電容以及等效電感值來描述,例如帶狀金屬之等效電路用如圖五所示。



圖五 等效電路示意圖[50]

其中等效電容值C(l, w, x)與電感值L(l, w, x)與長度關係分別為[51]:

$$L(l,w,x) = L_0(l,w) + \frac{\left(1 - B_L(l,w)^2\right)^2 \left(1 - \frac{1}{4} B_L(l,w)^2\right) A(x) + 4B_L(l,w)^2 A(x)^2}{1 - \frac{1}{4} B_L(l,w)^2 + 2B_L(l,w)^2 \left(1 + \frac{1}{2} B_L(l,w)^2 - \frac{1}{8} B_L(l,w)^4\right) A(x) + 2B_L(l,w)^6 A(x)^2}$$

$$C(l,w,x) = C_0(l,w) + \frac{\left(1 - B_C(l,w)^2\right)^2 \left(1 - \frac{1}{4} B_C(l,w)^2\right) A(x) + 4B_C(l,w)^2 A(x)^2}{1 - \frac{1}{4} B_C(l,w)^2 + 2B_C(l,w)^2 \left(1 + \frac{1}{2} B_C(l,w)^2 - \frac{1}{8} B_C(l,w)^4\right) A(x) + 2B_C(l,w)^6 A(x)^2}$$

 $\ddagger \Psi_{C_0} = 4l \ln \left(\csc \left(1/2\pi (1-l) \right) \right) \, , \, L_0(l,w) = l \ln \left(\csc \left(1/2\pi w \right) \right) \, , \, A(x) = 1/\sqrt{1-x^2} - 1 \, , \, B_c(l,w) = \sin \left(1/2\pi (1-l) \right)$

 $B_L(l,w) = \sin(1/2\pi w)^{\circ}$

有了等效電路模型後,就可加以利用 SPICE 等軟體分析與設計。由於在光波段中,金屬需考慮 穿透深度(skin depths)之影響,故在光波段使用等效電路分析時,需要作一個修正[52]。

利用超常材料吸收體來提高光吸收率之方法大致有下列三種方法[53]。一、利用超常材料人 工結構可調變的等效光學常數之特性,使吸收材料與太陽光入射面介質達到阻抗匹配(impedance match),減少菲涅耳反射(Fresnel reflection)。二、利用超常材料次波長結構在吸收材料中產生波導 (waveguide)共振模態,增加光路徑長度,提高光吸收率。三、利用超常材料由週期結構所組成之 特性,產生高階繞射來增加光路徑長度,提高光吸收率。此方法是屬於非共振式的,只與結構之 週期相關。為了有效吸收與利用太陽光能,吸收體之能隙(band gap)需儘量接近太陽輻射能譜之峰 值(如圖六所示)。適當地選擇吸收體材質(如金、銀、鋁、鎢等金屬與如矽、鍺等介電體),及設計 有效的共振結構,可以達到幾乎完美的吸收效率。



圖六 太陽能吸收頻譜(左圖)與材料吸收頻普(右圖)

二、超常材料光波吸收體

奈米光柵結構的異常光吸收機制,來自入射光與結構及材料之間的共振現象,此共振現象主要以兩種模態表現,即,Fabry-Perot 共振腔模態及束縛於介面上的表面模態[15,54]。對於不同偏振的入射光,所能激發的模態也不相同。一般而言,橫向電偏振(transverse electric, TE)或橫向磁偏振(transverse magnetic, TM)之入射光皆可產生 Fabry-Perot 共振腔模態,而表面模態則只能由 TM 偏振光激發。目前有關探討異常光吸收的文獻中,所提出的結構皆使用金屬,利用在金屬表面產生表面電漿,以達到激發束縛表面模態的目的,因此通常其異常光吸收的效果僅限於 TM 偏振光。相較於此,有關 TE 偏振光的異常吸收乃至於非偏極化光之異常吸收則較少有討論。另外,利用表面電漿所產生之共振頻寬通常較窄,因此不適合寬頻之應用。因此,我們計畫進一步研究可針對非偏極化光之寬頻異常吸收結構,希望藉由適當之結構與材料之選擇,使得 Fabry-Perot 共振腔 模態及表面模態能夠同時被激發,並且能夠有寬頻的反應,以期更有效率地提升光捕捉的能力。

根據前述,如欲同時對 TE 及 TM 兩種偏振達到異常吸收,則必須在結構中同時激發 Fabry-Perot 共振腔模態及表面模態,因此我們目前所研究者為多層結構配合表面週期光栅,如圖 七所示,在層間及光柵之間來形成共振腔,並且利光柵的週期特性來產生提供晶格動量,進一步 在界面上激發表面模態[55,56]。



圖七 異常光吸收奈米光栅結構示意圖

另外,為了有寬頻吸收效果,除了介電體外,我們還使用在可見光波段具有高耗散之材料(例如鎢 [57,58])作為吸收體的組成結構。雖然鎢在可見光波段不具有金屬特性,無法在其表面上激發表面 電漿共振,但也因其為高損耗介質的特性,故可在其表面上形成另一種束縛表面波,即所謂的 Zenneck wave [57]。圖八為上述結構對不同偏振之正向入射光的吸收頻譜。由圖中可知該結構對 不同偏振的入射光,皆可達到寬頻高吸收的作用。具體而言,在可見光波段其吸收效率皆高於 70%,而在波長 600 nm 附近,更可達到近乎完美吸收。另外,從兩種偏振的吸收頻譜有相當大的 重疊可知,該結構對於不同偏振的吸收表現相當一致,使得該結構特別適用於針對非偏振光的應 用。



圖八 異常光吸收奈米光栅吸收頻譜

為了驗證其吸收機制,我們也分析了結構中電磁場的分布。如圖九所示,當 TE 偏振達到完美吸 收時,在光柵之間形成了 TE₁₁ 共振腔模態。此外,在中間的介電層中也有共振腔模態產生。在光 柵間形成的共振腔模態,其共振條件則可由以下公式估算[59,60]:

$$\lambda_{mn} = \frac{2}{\sqrt{\left(m/a\right)^2 + \left(m/b\right)^2}}$$

然而,由於此處的共振腔上下兩側並非封閉,導致場的侷限下降[59,61],加上真實材料會有有限 的穿透深度,因此,實際的共振頻率會相較於公式推算值紅移。除了TE₁₁模態,我們也在較高頻 處(407 nm),觀察到TE₁₂模態。然而此時由於有高階繞射的產生,因此造成吸收的下降,而高階 繞射發生的波長及方向則可由以下的光柵方程式所決定:

$$\sin\theta_m = \sin\theta + \frac{m\lambda}{d}$$

其中,θ_m為第m階繞射波的反射角度,θ為入射光角度,λ為自由空間波長,而d則為光柵週期。 相較於TE偏振,由圖十的電場分佈圖可知,TM偏振光除了可以形成共振腔模態之外,也在鎢的 表面上形成束縛表面模態,即所謂的Zenneck wave。如果以電荷振盪的觀點,亦可稱之為結構表 面電漿(structured surface plasmon)[62],這特徵與理論上預期的一致。



圖九 TE 偏振電場分佈圖; (a) 吸收高點(600 nm)之 TE₁₁ 模態及 (b) TE₁₂ 模態(407 nm)



圖十 TM 偏振吸收高點 (609 nm) 電場分佈圖; (a) 水平分量及 (b) 垂直分量

三、結論

在本文中,我們使用電磁理論分析在次波長結構內的等效波長,並考慮電磁場穿透進入真實 材料的效應所造成的紅移效果,以期能夠準確預測吸收頻譜。另外,我們也在結構中加入金屬材 料,利用表面電漿子共振,來增強局部的電磁場,提供更大的吸收。同時,配合高損耗介質的使 用,我們可彌補金屬表面電漿子共振之窄頻的缺點,增加吸收頻寬。而在針對該結構做最佳化設 計時,則可利用包括多層結構及複合光柵所提供的自由度,如圖十一所示,調整不同偏振光的吸 收頻譜。最後,以此奈米光柵的設計理論及經驗,作為進一步設計超常吸收材料吸收體的基礎。



圖十一 複合奈米光栅結構示意圖

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