Opening stop-gaps in plasmonic crystals by tuning the radiative coupling of surface plasmons to diffracted orders

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By tuning the radiative coupling of localized surface plasmons to diffracted orders, we demonstrate how stop-gaps in plasmonic crystals of nanorods may be opened and tuned. The stop-gap arises from the mutual coupling of surface lattice resonances (SLRs), which are collective resonances associated with counter-propagating surface polaritons. We present experimental results for three different nanorod arrays, where we show how the dispersion of SLRs can be controlled by modifying the size of the rods. Combining experiments with numerical simulations, we show how the properties of the stop-gap can be tailored by tuning a single structural factor. We find that the central frequency of the stop-gap falls quadratically, the frequency width of the stop-gap rises linearly, and the in-plane momentum width of the standing waves rises quadratically, as the width of the nanorods increases. These relationships hold for a broad range of nanorod widths, including duty cycles of the array between 20% to 80%. We discuss the physics in terms of a coupled oscillator analog, which relates the tunability of the stop-gaps to the coupling strength of plasmonic modes.

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Photonic bandgaps, i.e., frequency regions within which optical propagation in a photonic crystal is forbidden, enable to control the flow of light. Gaps arise in the dispersion of photonic modes as a consequence of the optical periodicity, and the width of the gap is determined by the degree of field concentration in the different dielectric regions. A more complex scenario arises in plasmonic crystals, where light-matter interactions are no longer dominated by the optical periodicity alone. With the penetration of the electromagnetic field into the metal and the excitation of surface plasmon modes, fascinating phenomena fluorish in plasmonic crystals. For this reason, the coupling of surface modes in periodic metallic nanostructures has attracted much interest since early investigations, especially for its connection with frequency stop-gaps. Coupled surface modes have been observed in metallic grating, subwavelength hole array, nanoslit arrays, and particle arrays coupled to waveguide modes. Despite the numerous structures that have been investigated, very few studies have discussed the physical origin of these gaps. One notable exception is the work by Barnes and co-workers, where the origin of the gap in metallic sinusoidal gratings is discussed. However, the analysis therein contained is not easily extended to more complex plasmonic structures, which renders difficult the emergence of a simple, intuitive explanation on the origin of the gap.

It is the aim of this work to provide an intuitive frame-work in which the opening of frequency stop-gaps in plasmonic crystals can be understood in terms of the coupling strength of the surface modes involved. We investigate nanorod arrays supporting Surface Lattice Resonances (SLRs), which are dispersive and spectrally narrow collective resonances arising from the diffractive coupling of localized surface plasmons. This coupling occurs near the condition at which a diffraction order changes from radiating to evanescent, i.e. at the Rayleigh anomaly. In a recent work, we discussed the coupling of bright and dark SLRs in nanorod arrays. The associated stop-gap, modal symmetries, and the very high quality factors of SLRs were therein discussed. In this paper, we demonstrate how stop-gaps associated with coupled SLRs can be selectively opened by tuning the radiative coupling of localized surface plasmons to diffracted orders. We present experimental results for three different arrays with varying nanorod dimensions but equal lattice constants. By combining experiments with numerical simulations, we elucidate the influence of the width of the nanorod on the dispersion of SLRs. We also find scaling laws for the properties of the gap as a function of the nanorod width. As we will show, these scaling laws are related to the coupling strength of the surface modes involved, and they are valid for a wide range of duty cycles.

We have investigated experimentally three 3 × 3 mm2 arrays of gold nanorods fabricated on a silica sub-
strate using Substrate Conformal Imprint Lithography (SCIL) \cite{15}. The arrays were embedded in a uniform surrounding medium by placing a silica superstrate preceded by n = 1.45 index matching fluid to ensure good optical contact. The three arrays have lattice constants a_x = 600 nm and a_y = 300 nm, but they comprise nanorods which differ in size. The nanorods have an approximately rectangular shape in the plane of the array, and a height = 38 ± 2 nm. The rod size (length × width) was tuned by varying the exposure dose of the electron beam when preparing the master for nanoimprint. This procedure yielded rods of size (a) 420 × 85 nm², (b) 430 × 95 nm², and (c) 450 × 115 nm², which correspond to the measurements in Figures 1(a), 1(b), and 1(c), respectively. The tolerances of these in-plane dimensions are on the order of ±10 nm. Figures 1(a) and 1(c) show a Scanning Electron Microscope (SEM) image of the corresponding array, and a cartesian triad in the inset which we use to describe the measurements next.

Figure 1 shows the extinction, defined as 1 − T with T the zeroth order transmittance, for the three arrays described above. The extinction is displayed as a function of the reduced frequency, i.e., the angular frequency normalized by the speed of light in vacuum, and the component of the incident wave vector parallel to the surface, which is given by \( k_\parallel = \frac{2\pi}{a} n \sin(\theta) \hat{x} \) with n the refractive index of silica and \( \theta \) the angle of incidence. The sample was rotated around the y-axis while the y-polarized collimated beam from a halogen lamp impinged onto the sample, probing the short axis of the nanorods. The broad, dispersionless extinction peak seen on the high frequency side of the spectra for all three arrays corresponds to the excitation of Localized Surface Plasmon Resonances (LSPRs) in the individual nanorods. The black solid and dashed lines indicate the (+1,0) and (−1,0) Rayleigh anomalies of the arrays, respectively. The Rayleigh anomalies are solutions to the equation

\[
\begin{align*}
k_{\text{out}}^2 &= k_{m}^2 \sin^2(\theta) + m_x^2(2\pi/a_x)^2 + m_y^2(2\pi/a_y)^2 + 2k_{in} \sin(\theta)m_x(2\pi/a_x),
\end{align*}
\]

where \( k_{m} \) and \( k_{\text{out}} \) are the modulus of the incident and scattered wave vectors and \( m_j \) \((j = 1,2)\) are the integers defining the order of diffraction. Physically, these so-called anomalies represent the frequency and wave vector for which the corresponding diffracted orders are propagating grazing to the surface of the array. The coupling of LSPRs to the Rayleigh anomalies gives rise to the SLRs, which manifest in the measurements as narrow and dispersive peaks in extinction at slightly lower frequencies than the associated Rayleigh anomalies.

Figures 1(a)-(c) show the gradual opening of a stop-gap in the dispersion relation of SLRs as the nanorod width increases. The gap is centered near 7 mrad/nm in Fig. 1(a), but its central frequency is lowered and its width \( \Delta \omega_{\text{gap}} \) increases as the nanorods become wider. We note that this is not a complete photonic band-gap, since it only exists for light polarized parallel to the short axis of the nanorods and with an in-plane wave vector component parallel to the long axis of the nanorods. For light polarized parallel to the long axis of the nanorods, the dipolar LSPR lies at lower energies than the (±1,0) Rayleigh anomalies are degenerate, leading to degenerate (±1,0) SLRs and therefore no gap \cite{17}.

Inspired by previous work explaining electromagnetic resonance phenomena in terms of coupled oscillators \cite{15,17}, we recently introduced an analog to the plasmonic crystal consisting of three mutually coupled harmonic oscillators \cite{15}. In this analogy, the conduction electrons in the nanorod driven by the electromagnetic field are modeled...
The measurements in Fig. 2 show two main effects on the SLR lineshape as the width of the nanorods increases: i) the peak resonance frequency is increasingly detuned from the Rayleigh anomaly, and ii) the linewidth broadens. As it is shown next, this behavior is explained by the change of the coupling constant between the LSPR and the Rayleigh anomaly. In Fig. 3(a) we plot the detuning of the SLR from the Rayleigh anomaly, $\omega_{RA} - \omega_{SLR}$, and the linewidth at Full-Width Half Maximum (FWHM) of the SLR as a function of the width of the nanorods. It is remarkable that these two quantities are in quantitative agreement, which indicates a direct connection between radiative losses and the peak resonance frequency with respect to the corresponding Rayleigh anomaly. The implications of this connection for sensing small changes to the bulk refractive index by means of plasmonic nanoparticle arrays have been recently discussed. A universal scaling of the figure of merit of plasmonic sensors, which is a function of the detuning $\omega_{RA} - \omega_{SLR}$ alone, was therein found. In Fig. 3(b) we plot $\omega_{RA} - \omega_{LSPR}$ as a function of the nanorod width, which displays the diminishing frequency difference between the LSPR and the Rayleigh anomaly. This decrease in the magnitude of $\omega_{RA} - \omega_{LSPR}$ with a simultaneous broadening of the LSPR promotes a stronger radiative coupling of localized surface plasmons to diffracted orders, i.e., an increase in $\Omega_{12}$. Figure 3(c) shows the values of $\Omega_{12}$ used to fit the measurements in Fig. 2. Although in the fitted range $\Omega_{12}$ may well be described by a linear function, we have used a quadratic function for a reason that will be clarified further in the text. We see that increasing $\Omega_{12}$ detunes the SLR from the Rayleigh anomaly and broadens it in the right amount to have an excellent agreement with the measurements. A crucial understanding in attributing the observed behavior to $\Omega_{12}$ mainly lies in the fact that changing $\gamma_2$ (the intrinsic damping of oscillator 2) can not lead to the right detuning $\omega_{RA} - \omega_{SLR}$. We have verified this through several calculations (not shown here).
in which $\gamma_2$ and $\Omega_{12}$ were varied independently and/or simultaneously. Next, we consider the more general case of inclined incidence light, where the three oscillators are mutually coupled.

Inspection of Fig. 1 points towards the connection between the formation of standing waves in the high frequency SLR band and the opening of the gap. We consider the gap to open at the slowdown point for the high frequency SLR band and the opening of the gap. We consider the gap to open at the slowdown point for the high frequency SLR band and the opening of the gap. We consider the gap to open at the slowdown point for the high frequency SLR band and the opening of the gap. We consider the gap to open at the slowdown point for the high frequency SLR band and the opening of the gap. We consider the gap to open at the slowdown point for the high frequency SLR band and the opening of the gap. We consider the gap to open at the slowdown point for the high frequency SLR band and the opening of the gap. We consider the gap to open at the slowdown point for the high frequency SLR band and the opening of the gap. 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In order to investigate the properties of the gap for a broader range of nanorod widths, we have performed Finite Difference in Time Domain (FDTD) simulations with an in-house developed software. We have calculated dispersion relations in extinction for arrays with lattice dimensions as constants. We have calculated dispersion relations in extinction for arrays with lattice dimensions as constants.

To validate the simulation results, we compare the measured and calculated dispersion relations near the gap for a set of gold nanorods of 80 nm width in both cases. From the simulations of the three experimental arrays previously presented, it can be recognized that the nanorods in the simulations are about 7% shorter in length. Nevertheless, a good qualitative agreement is observed between measurements and simulations.

In Fig. 6(a) we plot the frequency width of the stop-gap, $\Delta \omega_{gap}$, and the $k_{\parallel}$ width of the standing waves in the high frequency SLR band, $\Delta k_{SW}$, both as function of the width of the nanorods. $\Delta \omega_{gap}$ is taken to be the frequency difference between the slowdown point and the (+1,0) SLR, both evaluated at the $k_{\parallel}$ value of the slowdown point. $\Delta k_{SW}$ is equal to the value of $k_{\parallel}$ at the

![Image](image_url)

**FIG. 5.** (Color online) Figure (a) is a zoom into the stop-gap displayed in the measurements of Figure 1(a). Figure (b) shows FDTD simulations results for an array of comparable antennas. The extinction is displayed by the same color scale for both measurements and simulations. The black solid and dashed lines represent the (+1,0) and (-1,0) Rayleigh anomalies, respectively.

In Fig. 6(a) we plot the frequency width of the stop-gap, $\Delta \omega_{gap}$, and the $k_{\parallel}$ width of the standing waves in the high frequency SLR band, $\Delta k_{SW}$, both as function of the width of the nanorods. $\Delta \omega_{gap}$ is taken to be the frequency difference between the slowdown point and the (+1,0) SLR, both evaluated at the $k_{\parallel}$ value of the slowdown point. $\Delta k_{SW}$ is equal to the value of $k_{\parallel}$ at the
TABLE I. Coupling and damping frequencies used in equation 1 to reproduce the spectra shown in Figure 4. In the entries for which a minimum estimate is given, the value in parenthesis is the value used in the model yielding the spectra in Figure 8. All quantities are given in units of mrad/nm.

| k1 | ω2  | ω3  | Ω12 | Ω13 | Ω23 | γ2  | γ3  |
|----|-----|-----|-----|-----|-----|-----|-----|
| 0.13 | 7.31 | 7.01 | 3.0 ± 0.1 | < 1.0 (0.4) | 1.0 ± 0.1 | < 0.02 (0.008) | 0.08 ± 0.03 |
| 0.18 | 7.34 | 6.93 | 3.2 ± 0.1 | < 1.0 (0.5) | 1.2 ± 0.1 | < 0.02 (0.008) | 0.08 ± 0.02 |
| 0.35 | 7.40 | 6.69 | 3.5 ± 0.1 | < 1.0 (0.8) | 1.5 ± 0.1 | < 0.02 (0.008) | 0.08 ± 0.01 |

slowdown point. In Fig. 6(b) we plot the detuning of the (±1, 0) SLRs from their respective theoretically predicted Rayleigh anomalies, also as a function of the nanorod width. The open data points are taken from the measurements of the three arrays, and the solid and dashed curves result from the simulations. A central finding is that both Δω_{gap} and the (-1,0) detuning are linearly increasing functions of the nanorod width, while Δk_{SW} and the (+1,0) detuning are both quadratically increasing functions of the nanorod width. We therefore find the frequency width of the gap to be correlated with the (-1,0) detuning, and the k∥ width of the standing waves to be correlated with the (+1,0) detuning. In terms of the coupled oscillator model, this can be interpreted as follows: the gap opens with a linear increase in Ω13 (coupling of LSPR to the (-1,0) order), whereas the in-plane momentum width of the standing waves broadens with a quadratic increase in Ω12 (coupling of LSPR to the (+1,0) order). The latter dependance is the reason for which Ω12 was fitted with a quadratic function in the normal incidence spectra, i.e., the dashed curve in Fig. 3(c).

Further quantifying the properties of the gap, in Fig. 7(a) we plot the central frequency of the stop-gap, ω_{gap}, which falls quadratically for increasing nanorod width. In Fig. 7(b) we plot the gap-midgap ratio, Δω_{gap}/ω_{gap}, which rises quadratically for increasing nanorod width. Figure 7(a) shows that for nanorods wider than those considered in the experiments, the central frequency of the gap falls into the infrared part of the spectrum. Limited by the spectral response of our acquisition system (based on a silicon detector), we are unable to perform measurements for the wider nanorods. Nevertheless, in light of the good agreement between measurements and simulations we believe Figs. 6(a) and 7 accurately encompass all properties of the gap as function of the nanorod width, and provide a suitable recipe upon which stop gaps in plasmonic crystals may be opened and tuned. Comparing to previous work, where it was found that the gap’s width is a linearly increasing function of the modulation amplitude in metallic sinusoidal gratings, we have also found a linear dependance of the gap’s width on a single structural factor. However, the connection between these two works is not an obvious one, since whereas Barnes and co-workers have varied a dimension out of the plane of propagation, we have investigated structures of equal height but variable dimensions in the plane of propagation. From a fabrication point of view, a precise in plane structuring of plasmonic crystals may offer a higher degree of versatility in how stop-gaps may be tuned and at a greater ease.

We now discuss the validity range of the previously discussed SLR properties on the nanorod width. Figure 8 shows the extinction of two arrays with lattice constants a_x = 600 nm, a_y = 300 nm, nanorods of length= 450 nm and height = 40 nm, and embedded in a homogeneous environment of n=1.45; these conditions are the same as in the experiments presented in Fig. 1. The width of the nanorods in Fig. 8 is (a) 60 nm, and (b) 240 nm, which correspond to the extremes of the curves plotted in Figs. 6 and 7. Due to the very different extinction efficiency of the two arrays in the spectral region of interest, the extinction is displayed by a logarithmic color scale common to both graphs, which allows quantitative comparison of the spectra. The low extinction and remarkably narrow resonances seen in Fig. 8(a) can be explained in terms of the coupled oscillator model as a consequence of the low coupling strength of LSPRs to diffracted orders. This point can be inferred from the

FIG. 7. (Color online) Figure (a) shows the central frequency of the stop-gap, ω_{gap}, with black open squares representing measurements and a black solid curve representing FDTD simulation results. Figure (b) shows the gap-midgap ratio, with blue open circles representing measurements and a blue solid curve representing FDTD simulations results. All quantities in (a) and (b) are plotted as a function of the nanorod width.
Quantities given in Table 1, where all coupling frequencies $\Omega_{jk}$ are seen to decrease as the nanorod width decreases. Lower $\Omega_{12}$ and $\Omega_{13}$ therefore translate into a lower coupling strength of LSPRs to diffracted orders, whereas a lower $\Omega_{23}$ translates into a lower coupling between the $\pm (1,0)$ and $\pm (1,0)$ SLRs and therefore a smaller gap. As the width of the nanorods increases and the $\Omega_{jk}$ terms increase, the extinction at the SLRs also increases, the resonance linewidth broadens, and the frequency gap widens, eventually reaching the case displayed in Fig. 8(b) for a nanorod width of 240 nm. Figure 8(b) displays a dipolar LSPR near 6.8 mrad/nm, which is lower in frequency than the diffraction edge at normal incidence. The $(+1,0)$ SLR can still be recognized in the spectrum from the non-dispersive feature near 6.0 mrad/nm (in the red tail of the LSPR). However, for wider nanorods the LSPR shifts to lower frequencies, thereby making the $(+1,0)$ SLR indistinguishable from the LSPR. As the energy of the dipolar LSPR becomes substantially lower that the $(\pm 1,0)$ diffraction orders, diffractive coupling of dipolar LSPRs becomes very weak, and the properties of coupled SLRs leading to the opening of the stop-gap significantly deviate.

In conclusion, we have shown how the radiative coupling strength of localized surface plasmons to diffracted orders in periodic arrays of nanorods can be tuned. This tuning is achieved experimentally by modifying the width of the nanorods, and results in the opening of frequency stop-gaps whose properties are therefore tunable. A quadratic dependence of both the frequency width of the stop-gap and the $(-1,0)$ SLR detuning was found on the nanorod width. A linear dependence of both the in-plane momentum width of the standing waves in the high-frequency SLR band and the $(+1,0)$ SLR detuning was found on the nanorod width. In light of a coupled oscillator analog to the plasmonic crystal, we have associated these two correlations with the coupling strength of localized surface plasmons to the $(1,0)$ and $(\pm 1,0)$ diffracted orders. Supporting experiments with numerical simulations for a wider set of nanorod widths, we have analyzed the properties of the gap and discussed the limiting cases where the scaling laws we present deviate. Although we have only considered nanorod arrays in this work, similar results are expected to hold in periodic arrays of particles with different geometries whereby diffractive coupling of localized surface plasmons is possible. Our results therefore pave the road towards nanoscale light manipulation in 2D plasmonic crystals, since stop-gaps allow to selectively enhance or suppress light-matter interactions.

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**FIG. 8.** (Color online) FDTD simulations results for the extinction of an array of gold nanorods of width (a) 60 nm and (b) 240 nm. Both arrays have nanorods of length $= 450$ nm, height $= 40$ nm, in a lattice with constants $a_x = 600$ nm and $a_y = 300$ nm. The black solid and dashed lines indicate the $(+1,0)$ and $(-1,0)$ Rayleigh anomalies, respectively. The extinction is displayed by a logarithmic color scale common to both graphs.
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