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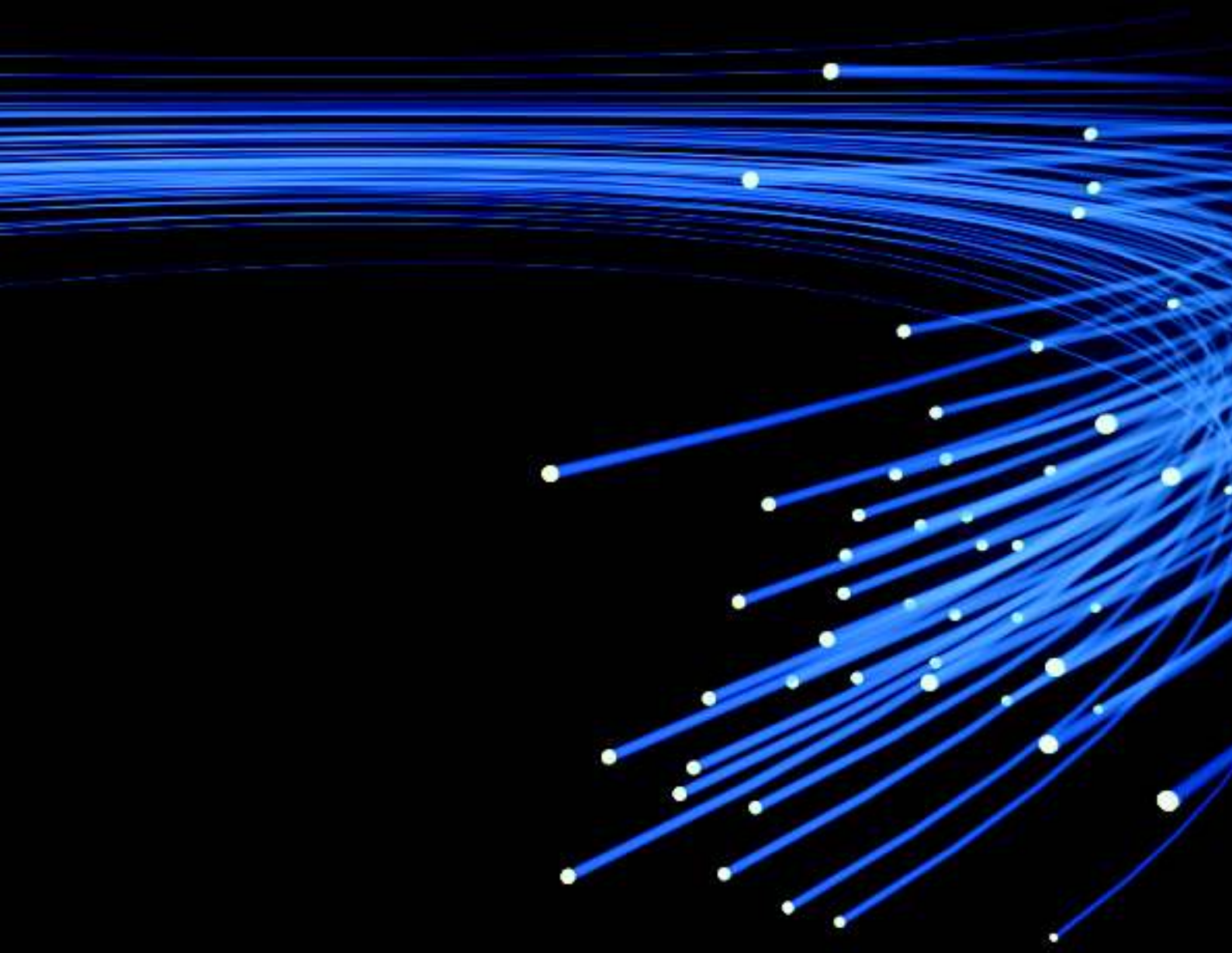

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Surface lattice resonances in plasmonic nanorod arrays

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Abstract

Radiative coupling between localized surface plasmon resonances (LSPRs) and Rayleigh anomalies (RAs) gives rise to the mixed states: surface lattice resonances (SLRs). These SLRs can have very narrow linewidth while still providing a large near field enhancement. Here we discuss the tuning of SLR modal properties in periodic arrays of metallic nanorods through controlling the nanorods' width. Variable angle light extinction measurements were done for five arrays and simulations were done to explain the results.

1. Introduction

In recent years, periodic arrays of metallic nanoparticles have attracted much attention for their ability to support collective resonances arising from the radiative coupling between localized surface plasmon resonances (LSPRs) and waves diffracted in the plane of the array (and Rayleigh anomalies (RAs)) [1-5]. These collective resonances are known as surface lattice resonances (SLRs). Despite the numerous fundamental and applied studies concerning SLRs, their general properties in terms of the coupling conditions between the participating plasmonic and photonic modes are limitedly explored.

It is the aim of this work to elucidate how the SLR characteristics depend on the spectral properties of the interacting RAs and LSPRs, which are in turn determined by simple structural design. We focus on nanorod arrays where a single first order LSPR interacts with RAs associated with two diffraction orders [(+1,0) and (-1,0)]. Experimental extinction spectra of arrays with nanorods of different width w are investigated. We focus on the case of light polarized along the width of the nanorod. The energy and linewidth of the LSPR, and thus the detuning with respect to the RAs, depend on the nanorod dimensions in arrays with fixed periodicity. The LSPR red-shifts and broadens for increasing nanorod width due to the depolarization field along this dimension. Our work analyzes how this changes the coupling of the LSPR to the RAs. We shed light into the physics through transmission and eigenmode simulations using finite element based methods. For a more complete and in depth discussion of the results, please refer to our publication on this topic [4].

2. Measurements

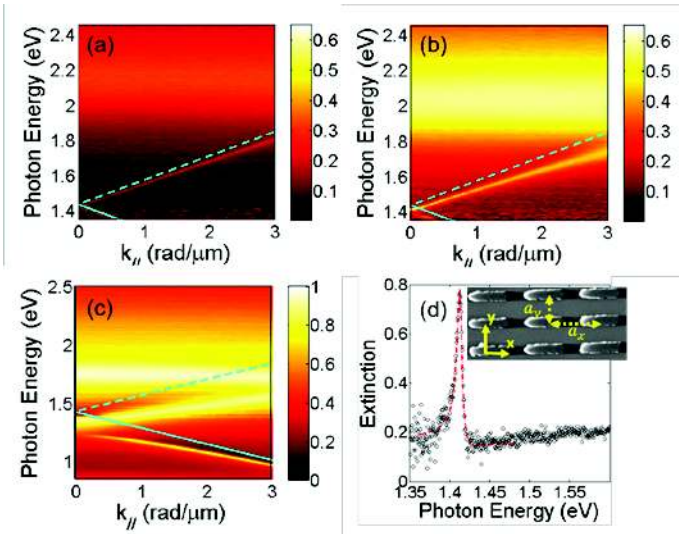

Figure 1 Experimentally measured extinction for (a) w = 70 nm, (b) w = 110 nm, (c) w = 200 nm, and (d) experimental extinction cross-section at $k_{//} = 0$ for w = 110 nm array, fitted with a Fano resonance (red dashed lines) for the upper SLR. The inset in (d) shows the SEM picture of the nanorod array with w = 110 nm.

Five gold nanorod arrays with varying nanorod width (w) of 70, 110, 160, 200 and 230 nm (in the y direction) but equal nanorod length of 450nm (in the x direction) were fabricated on a silica substrate using electron beam lithography. The arrays have dimensions of 1.5×1.5 mm^2 and lattice constants a_x = 600 nm and a_y = 300 nm. The nanorods have an approximately rectangular shape in the plane of the array, and a height of ±40 nm. The rod width was tuned by varying the exposure dose of the electron beam. The tolerances of the in-plane dimensions are on the order of ±10 nm. 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. Here we only show measurement results of 3 of the arrays.

The extinction measurements of y polarized light, defined as 1-T_0 are shown in Fig. 1(a-c) with T_0 the zeroth-order transmittance through the arrays described above. The extinction is displayed in color as a function of the incident photon energy and component of the wave vector parallel to the surface in the x direction ($k_{//}$). The samples were 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 dispersionless and broad extinction peak seen on the high energy side of the extinction spectra for all three arrays corresponds to the excitation of LSPRs in the individual nanorods. The cyan solid and dashed lines indicate the (-1,0) and (+1,0) RAs, respectively. The coupling of LSPR to the RAs yields the upper and lower SLRs. Fig.1(d) shows a cross-section plot of the extinction for w = 110 nm at $k_{//} = 0$ where one can see that the upper SLR has a Fano shaped resonance. The Fano shape is a characteristic of modes arising from a mixing between a broad and narrow resonance which in this case is the LSPR and the RA respectively. Upon fitting with the Fano function, we deduced the upper SLR linewidth to be 8.29 meV ±0.56 meV.

The correlation between the SLRs' characteristics and the spectral properties of the LSPR can be seen in Fig. 1(a-c). As the LSPR broadens and approaches the RAs in energy upon increase of w, the SLRs shift towards lower energies, their linewidths broaden, and the gap between them widens. Additionally, one can see more strong deviation of the SLR dispersion from the associated RA as the nanorod widens. This indicates that there is an increase of coupling strength between the LSPR and RAs. The broadening of the SLR linewidth, which implies increased losses, can be understood on the basis of the increased influence from the lossy LSPR.

The effect of LSPR-RA energy detuning can also be observed in the SLR dispersion of each array. As $k_{//}$ increases, the detuning between the upper SLR and LSPR diminishes. Consequently, the SLR linewidth broadens and its peak energy deviates more pronouncedly from the (+1,0) RA. In contrast, the lower SLR becomes narrower and approaches the (-1,0) RA as it becomes more detuned from the LSPR.

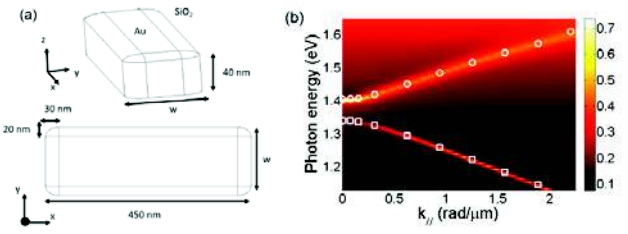

Figure 2 (a) Diagram of the simulated nanorods. (b) Simulated extinction (color plot) dispersion for w = 110 nm. White markers indicate the eigenfrequencies of the SLR modes obtained through eigenfrequency calculations. Circle and square markers indicate data points for upper and lower SLR respectively.

3. Numerical simulations

The SLR characteristics in the presence and absence of a driving optical field are examined through finite element method simulations (COMSOL). In the transmission simulations, we use plane wave illumination incoming at various angles as the driving field to obtain the extinction spectra. Fig. 2(a) shows a sketch of the nanorods in the simulated arrays. Fig. 2(b) shows the simulated extinction of the w = 110 nm array. As can be seen in Fig. 2(b), our simulations reproduce well the SLR characteristics observed in the measurements. Similar agreement was obtained for other arrays. The SLR peak energies obtained from the transmission simulations serve as an initial guess and check for the eigenenergies in the un-driven case obtained from eigenmode calculations.

With the eigenmode simulations we focused on finding the SLRs which are Bloch modes that propagate in the plane of the array along the x-direction. The SLR eigenenergies calculated at a few values of $k_{//}$ are plotted as white circles in Fig. 2(b). The good agreement of the SLR dispersion obtained with eigenmode and transmission simulations confirms that the obtained Bloch modes are indeed SLRs.

Fig. 3(a) shows a cross-section plot of the electric field in the y direction for both of the SLR modes. One can see that the upper SLR is symmetric in one cell period while the lower SLR is antisymmetric at $k_{//} = 0$. Thus, the latter is "dark" and not excitable by incoming normal incident plane waves. For more in depth analysis of the bright and dark nature of the SLRs, please refer to our publication [4-5].

Fig. 3(b) shows the magnitude of the total SLR eigenfields for arrays with different w. The upper and lower SLRs are shown in top part and bottom part respectively. As can be seen in Fig. 3(b), the near field confinement of both SLRs is enhanced as w increases for both SLRs. This effect is more clearly visible for the lower SLR, as this mode lacks a radiative component at $k_{//} = 0$. In contrast, the upper SLR retains a radiative component for all w. We observe that the magnitude of the eigenfield increases in the spatial region away from the nanorods in Fig. 3(b) for the upper SLR as w increases. Since the fields in this region are mostly radiative in character, it follows that the radiative portion of the total loss also increases with increasing w for the upper SLR. This effect, in combination with the increased near field confinement, which naturally leads to larger ohmic losses, can thus be expected to increase the linewidth of the upper SLR at $k_{//} = 0$ in the driven system. The latter observation is very clear from the experiments Fig. 1(a-c). More in depth discussion on the radiative loss and SLR eigenprofiles for non-zero $k_{//}$ can be found in our paper [4].

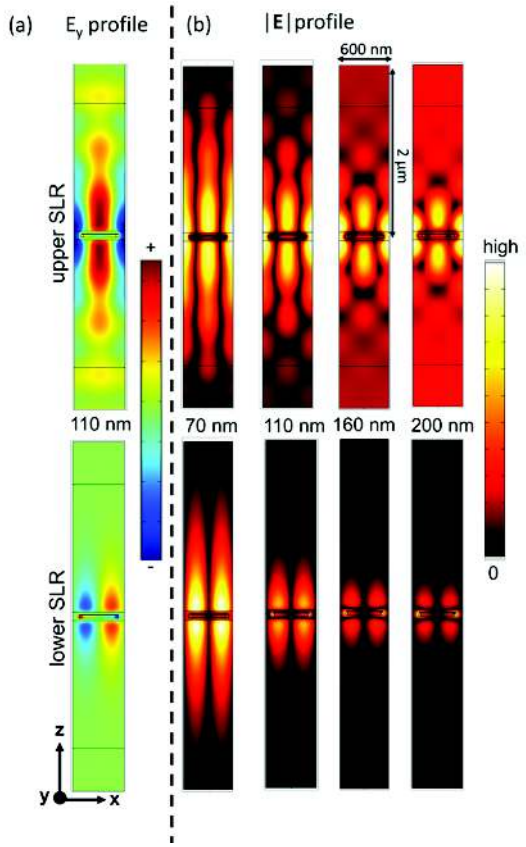

Figure 3 x-z cross-section plot through the center of the nanorod of the eigenmode (a) E_y (b) |**E**| profile of upper and lower SLR for different w at $k_{//} = 0$. Each |**E**| plot has a different normalization individually. The dimension listed is the nanorod width (w).

4. Conclusions

We have shown how the properties of the mixed states: surface lattice resonances (SLRs) depend on the spectral properties of localized surface plasmon resonances (LSPRs) and Rayleigh anomalies (RAs). The SLR dispersion, linewidth, and associated stop-gap can be tuned by controlling the energy detuning between the LSPR and RAs. In turn, the LSPR spectral characteristics can be tuned via a single geometrical parameter of the nanorods: their width.

Our results demonstrate the flexibility in tailoring the properties of hybrid plasmonic/photonic modes by simple geometrical design. The results show the interesting balance between diffraction orders, which can be exploited to control both the near- and far-field profile for diverse applications.

Acknowledgements

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