Near-Field Resonance at Far-Field Anti-Resonance: Plasmonically Enhanced Light Emission with Minimum Scattering Nanoantennas

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We demonstrate that a periodic array of optical antennas sustains a resonant Near-Field (NF) and an anti-resonant Far-Field (FF) at the same energy and in-plane momentum. This phenomenon arises in the context of coupled plasmonic lattice resonances, whose bright and dark character is interchanged at a critical antenna length. The energies of these modes anti-cross in the FF, but cross in the NF. Hence, we observe an extremely narrow bandwidth emission enhancement from quantum dots in the proximity of the array, while the antennas scatter minimally into the FF. Simulations reveal that a standing wave with a quadrupolar field distribution is the origin of this dark collective resonance.

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Resonance phenomena are at the heart of nearly any approach towards controlling the emission, manipulation, or detection of light. A resonance frequency is commonly identified as that for which the response of a system is enhanced. For a plane wave excitation, this implies that more energy is removed from an incident beam at resonance than off resonance. In the presence of surface electromagnetic waves, the definition of a resonance requires a careful reconsideration, since the spectrum of radiation may be different in the Near-Field (NF) with respect to the Far-Field (FF) [1,2]. Such differences have a profound significance for the field of metallic nanoptics, partly because the emergence of optical antennas has been largely fueled by an increased capability to manipulate optical near fields [3]. The basis for achieving such a control relies on the excitation of surface plasmon polaritons - collective oscillations of the free electrons in the metal driven by an electromagnetic field. However, not all surface plasmon modes are visible in the FF spectrum. In 2001, a class of plasmonic modes which manifest as resonances that can be excited or observed in the NF but not in the FF, so-called dark modes, were theoretically predicted [5]. These dark resonances have attracted much interest in recent years [6,7], as they hold supreme qualities for the realization of SPASERS [8,9], subwavelength guiding of optical radiation with suppressed radiative losses [10], plasmonic analogs of electromagnetically induced transparency [11,12], and cloaked sensors [13]. In particular, theoretical work by Alù and Engheta suggests that it is possible to strongly suppress the FF radiation from a nanoantenna while preserving an enhanced NF sensitivity [15,16] - an optical counterpart to radio frequency minimum scattering antennas. As pointed out by García de Abajo in Ref. [17], by having an enhanced interaction with its local environment but a minimum interaction with distant sources and detectors, such a minimum scattering antenna is effectively seeing without being seen.

In this Letter, we experimentally demonstrate that a periodic array of plasmonic nanoantennas displays a local minimum in its FF extinction but a maximum in the average NF enhancement in the plane of the array. This is evidenced from an enhanced emission of quantum dots in the vicinity of the array at an energy and in-plane momentum for which the FF extinction is minimized. Hence, a NF resonance co-exists in energy and in-plane momentum with a FF anti-resonance. Finite Difference in Time Domain (FDTD) simulations show that this phenomenon is based on the coupling of two counter-propagating surface polaritons whose energies anti-cross in the FF extinction but cross in the NF enhancement. A standing wave with a quadrupolar, i.e., dipole forbidden, field distribution is formed at the NF crossing energy.

For the experiments, we have fabricated a $2 \times 2 \text{mm}^2$ periodic array of gold nanoantennas onto a silica substrate by means of electron beam lithography. The antennas have dimensions $270 \times 80 \times 40 \text{nm}^3$, and the lattice constants are $a_x = 600 \text{ nm}$ and $a_y = 300 \text{ nm}$. We spin-coated a 600 nm layer of PbS/CdS core/shell Quantum Dots (QDs) in a polystyrene matrix, hereafter referred as the QD layer, on top of the array. The QDs emit at a peak energy of 1.33 eV with a full width at half maximum of 0.28 eV. Further details on their synthesis and optical properties may be found in the supplemental.

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The magnitude of the scattered wave vector, \( k \), is superior to the one observed for the bright mode probed from the FF there is an emission enhancement that at the same energy the FF extinction is very low. It is remarkable that at an energy and in-plane momentum for which the nanoantennas scatter minimally when the detector subtends with respect to the normal. The extinction is shown as a solid red line and the PLE as a dashed blue line for (c) \( k_{||} = 0 \) (Fig. 1(c)), a single peak in extinction associated with the bright (-1,0) SLR arises near the anti-crossing. Notice that the dispersion of the bright (-1,0) SLR flattens and the extinction increases near the anti-crossing. This indicates the formation of standing waves with an enhanced density of optical states at the band edge. A narrowing linewidth and diminishing extinction are observed for the (+1,0) SLR, which are the signature of subradiant damping [26, 27].

Figure 1(b) shows the measured variable angle PhotoLuminescence Enhancement (PLE), for the nanoantenna array described in the text. The photon energy and wave vector component parallel to surface refer to the (a) incident and (b) emitted radiation. The solid and dash-dotted white lines indicate the (+1,0) and (-1,0) Rayleigh anomalies, respectively. The inset in (a) shows a magnified view of the low \( k_{||} \) region, where the gap opens. The extinction is shown as a solid red line and the PLE as a dashed blue line for (c) \( k_{||} = 0 \), (d) \( k_{||} = 0.08 \) mrad/nm, and (e) \( k_{||} = 0.22 \) mrad/nm; the latter two cases are indicated by the dashed lines in (a) and (b).

Figure 1(a) shows the measured variable angle FF extinction, defined as \( 1 - T_0 \), with \( T_0 \) the zeroth order transmission through the sample normalized to the transmission through the substrate and QD layer. The measurements are shown as a function of the incident photon energy and the wave vector component parallel to the long axis of the antennas, \( k_{||} = k_0 \sin(\theta_{\text{em}}) \hat{x} \), where \( k_0 = \frac{2\pi}{\lambda} \) is the magnitude of the free space wave vector and \( \theta_{\text{em}} \) is the angle of incidence. The incident light is polarized parallel to the short axis of the nanoantennas, i.e., s-polarized. The (+1,0) and (-1,0) Rayleigh anomalies are indicated by the white solid and dash-dotted lines, respectively. They represent the conditions for which the corresponding diffraction orders are radiating in the plane of the array. Their dispersion is calculated from the conservation of the parallel component of the wave vector, expressed as \( k_{\text{out}}^2 = (k_x + m_1 G_x)^2 + (k_y + m_2 G_y)^2 \), with \( k_{\text{out}} \) the magnitude of the scattered wave vector, \( k_{||} = (k_x, k_y) \) the wave vector components parallel to the surface, the integers \( (m_1, m_2) \) defining the order of diffraction, and \( G = (G_x = \frac{2\pi}{a_x}, G_y = \frac{2\pi}{a_y}) \) the reciprocal lattice vector.

An effective refractive index of \( n=1.50 \) due to the underlying substrate and the QD layer was used to calculate the Rayleigh anomalies. The two peaks in extinction following the dispersion of the Rayleigh anomalies on the low energy side correspond to the excitation of Surface Lattice Resonances (SLRs), which are collective Fano resonances arising from the diffractive coupling of localized surface plasmons [19, 22]. The mutual coupling of SLRs leads to an anti-crossing in their dispersion relation, i.e., the opening of a frequency gap close to \( k_{||} = 0 \) [20]. The inset of Fig. 1(a) displays a magnified view of the gap.

Notice that the dispersion of the bright (-1,0) SLR flattens and the extinction increases near the anti-crossing. This indicates the formation of standing waves with an enhanced density of optical states at the band edge. A narrowing linewidth and diminishing extinction are observed for the (+1,0) SLR, which are the signature of subradiant damping [26, 27].
FF anti-resonance is the focus of this work. For comparison, we show in Fig. 1(e) the measurements at $k_\parallel = 0.22$ mrad/nm, where two broad features in the PLE are observed near the energies of the $(\pm 1,0)$ SLRs in extinction, as previous work has shown [23]. In what follows, we elucidate by means of FDTD simulations the conditions leading to the NF resonance at a FF anti-resonance. We investigate gold nanotriangular arrays with the same lattice constants as in the experiments, surrounded by a fully homogeneous environment of $n=1.46$. The dielectric function of gold is taken from Ref. [28], and fitted in the range of interest with a Drude model. The incident light has an in-plane momentum and a polarization vector as in the experiments.

Figure 2 shows simulations results for the (a) FF extinction and (b) average NF Intensity Enhancement (IE) defined as $IE = |E|^2/|E_0|^2$, with $E$ the total electric field at a plane intersecting the antennas at their mid-height and $E_0$ the incident field. The antenna dimensions are $L \times 110 \times 40$ nm$^3$, and the spectral response to a plane wave with $k_\parallel = 0.08$ mrad/nm is shown as a function of the antenna length $L$. Besides $L$, all other parameters are kept constant. The high and low energy resonances in both figures are the $(+1,0)$ and $(-1,0)$ SLRs, respectively. Their extinction and NF IE vary with the antenna length as a consequence of retardation and radiative damping. Notice that for $L > 250$ nm the $(+1,0)$ SLR is bright, i.e., it has a high extinction, whereas the $(-1,0)$ SLR is dark, i.e., it has a narrowing linewidth and a low extinction. The extinction of the $(-1,0)$ SLR vanishes for $L > 350$ nm. To exemplify the FF dispersion diagram in the long antenna regime, we show in Fig. 2(c) the variable angle extinction spectra calculated for an array of antennas with dimensions $450 \times 110 \times 40$ nm$^3$. We clarify that the spectra in Fig. 2(c) at $k_\parallel = 0.08$ mrad/nm and in Fig. 2(a) at $L = 450$ nm (both denoted by a white dash-dotted line) are identical. The dispersion diagram in Fig. 2(c) shows that, as the two SLR bands approach each other near normal incidence, the high energy band flattens and the extinction and linewidth of the low energy band vanish. This results in the opening of a large gap. As observed in Fig. 2(a) and 2(b), the properties of the high and low energy SLRs bands are interchanged for $L \lesssim 250$ nm. Thus, in the short antenna regime the flattening of the band occurs for the $(-1,0)$ SLR, while subradiant damping onsets for the $(+1,0)$ SLR. This explains why in a recent work the $(+1,0)$ SLR was bright and the $(-1,0)$ SLR was dark [20] (as in Fig. 2(c)), whereas in the measurements presented here (Fig. 1) this behavior is reversed. These results demonstrate that the onset of subradiant damping can be modified, or even halted for a given band, by designing the antenna length. Furthermore, a remarkable phenomenon arises at the critical antenna length for which the properties of SLRs are swapped, which occurs for $230$ nm $\lesssim L \lesssim 270$ nm. In this regime, the FF displays an energy anti-crossing characteristic of coupled surface modes, but the NF displays a crossing of the two bands. At this point the NF is resonant at an energy and in-plane momentum for which the FF is anti-resonant.

The connection between the above spectra as a function of the antenna length $L$ and the experimental results of Fig. 1 is elucidated in Fig. 3 by considering FDTD simulations for antennas of $L = 250$ nm. This length is indicated by the black dashed lines in Figs. 2(a) and
Figs. 3(a) and 3(b) at three values of $k$. The latter condition leads to a maximum NF. $k$ are broader, and shifted towards higher $k$ with respect to the FF. The condition of 0.12 mrad/nm, corresponding to the NF resonance at 0.12 mrad/nm. The antenna array is identical to the one described for Fig. 3. The illuminating plane wave has an energy of 1.37 eV and $k || = 0.12$ mrad/nm, corresponding to the NF anti-resonance in Fig. 3(d).

In conclusion, we have demonstrated that differences in the far-field and near-field spectra of radiation of optical antenna arrays can be exploited to create a medium with large local field enhancements but minimized extinction. This local field enhancement allowed us to observe an extremely narrow bandwidth emission enhancement of quantum dots in the vicinity of the array with a simultaneous minimum in extinction. Dark plasmonic resonances hold remarkable features for modified light emission and sensing, both of which depend on the local field rather than on the global symmetry of the array determining the FF extinction. Our results demonstrate that dipole inactive or dark modes are capable of enhancing the performance of photonic devices to a level comparable, or even superior, to bright modes.
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