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Enhanced Quality Factors of Surface Lattice Resonances in Plasmonic Arrays of Nanoparticles

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Key in the application of plasmonics is the realization of low loss or high quality (Q) factor resonances. Nanoparticle arrays are systems capable of sustaining remarkably high Q-factor resonances through the hybridization of plasmonic and photonic modes, known as surface lattice resonances (SLRs). SLRs result from the coupling of localized surface plasmon resonances (LSPRs) to in-plane orders of diffraction known as Rayleigh anomalies (RAs). To date, the highest Q-factors have been achieved with the (±1, 0) diffraction orders. However, these Q-factors are highly sensitive to the angle of excitation. Here, a strategy is presented to generate high Q-factor SLRs with low dispersion by coupling LSPRs to the (0, ±1) diffraction orders. 2D arrays of silver nanoparticles are investigated experimentally and numerically, and it is shown that the Q-factor of SLRs critically depends on the quality of the metal film, the detuning between RAs and LSPRs, and the absorption of adhesive layer used between the substrate and the metallic nanoparticles. These silver nanoparticle arrays can achieve Q-factors higher than 330 in the visible range. These extraordinarily high Q-factors could be increased to values above 1500 if no adhesive layer is used, which could significantly improve sensors and enhance nonlinearities in plasmonic systems.

The revolution of modern plasmonics in the latest years of the 20th century was sparked by the technology advances in producing nanoscale structures.[1] Patterned surfaces with metallic nanostructures have gained significant interest due to the large enhancement of local electromagnetic fields, which is useful to investigate light–matter interaction and nonlinearities at subwavelength scales that were inaccessible before.[2] However, the field enhancement of localized surface plasmon resonances (LSPRs) becomes negligible when the distance to the metallic nanoparticles is larger than a few tens of nanometers. In parallel to studies of localized resonances, several works have focused on spatially delocalized resonances of plasmonic nanoparticles that emerge in periodic arrays under specific conditions.[3,4] Laor and Schatz considered the dipolar coupling of metallic hemispheres in periodic clusters.[5] This work was extended by Meier et al. and Carron et al. to describe narrow resonances in the surface-enhanced Raman spectra of lattices of metallic nanoparticles.[6,7] Markel investigated 1D chains of nanoparticles using the coupled dipole approximation to describe narrow resonances emerging from diffraction coupling.[8] This work was further expanded by Zou, Schatz, and coworkers with the physical description of ultranarrow linewidths that could be achieved in arrays of silver nanospheres.[9,10]

The experimental demonstration of narrow resonances in arrays of metallic nanoparticles was more elusive due to limitations in the quality of samples and the use of focused beams.[4] Kravets et al. reported ultranarrow plasmonic resonances in asymmetric (different refractive indexes in the upper and lower media) arrays of Au nanoparticles.[11] Shortly after, Auguié and Barnes[12] and Chu et al.[13] reported narrow resonances in symmetric arrays. The potential of narrow plasmonic resonances in arrays of nanoparticles for modifying the emission of fluorophores was also demonstrated.[14,15] The origin of the narrow resonances, which are known as surface lattice resonances (SLRs), is the diffractive coupling of LSPRs through in-plane diffraction orders in symmetric media or evanescent diffraction orders—the so-called Rayleigh anomalies (RAs)—in asymmetric media. SLRs can be described as a driven damped coupled oscillator system in which one oscillator has the natural frequency of the LSPR while the other has the frequency of the diffraction order.[16]

Nanoparticle arrays are open cavities that are easy to fabricate and offer the possibility of integration with thin films or planar structures. The remarkable properties of SLRs have
led to improved surface-enhanced Raman scattering,\(^{[6,7,17]}\) sensitive bio/chemical sensing,\(^{[18–20]}\) plasmonic band-edge lasing,\(^{[21–23]}\) strong light–matter coupling,\(^{[24–27]}\) Bose–Einstein condensation,\(^{[28,29]}\) and optoelectronic devices.\(^{[30]}\) The multidisciplinary impact of SLRs has stimulated the quest toward modes with the highest possible quality (Q) factor.\(^{[8–11,29–33]}\) One strategy to obtain narrow linewidths with SLRs is by coupling multipolar resonances with different diffraction orders.\(^{[16,34,35]}\) However, Q-factors by these approaches vary significantly over momentum space.

In this communication, we focus on the impact of fabrication processes and material aspects on the quality factor of SLRs. We investigate degenerate SLRs emerging through the coupling of dipolar LSPRs with the (0, ±1) RAs, leading to a quadratic dispersion with little momentum dependence. We show how the coupling strength defines the linewidth and frequency of the SLRs. Additionally, the role of the metal quality and the absorption of the metal adhesive layer are studied to complete a set of rules for obtaining high Q-factor resonances. Using Ag nanoparticles, we demonstrate experimentally Q-factors higher than 300 and nearly independent of the angle of incidence or in-plane momentum. These remarkably high Q-factors could be further increased to values above 1500 by suppressing the parasitic absorption of the metal adhesion layer that is used between the substrate and the metal nanoparticles.

The spectrum of SLRs can be calculated from the dissipated power of a driven coupled oscillator system in which one oscillator represents the LSPRs with a natural frequency independent of the wave vector and the other is the dispersive RA. SLRs follow the dispersion of the RAs with a small shift emerging from the coupling. The dispersion of the RAs is given by the grating equation: 

\[
\pm k_R = \pm k_i + G,
\]

where \(k_i\), \(G\) correspond to the wave vectors of the Rayleigh anomaly, the in-plane wave vector component of the incident wave, and the reciprocal lattice vector, respectively. Here, \(G = (2\pi/\lambda)\mathbf{n}_{\text{eff}}\mathbf{u}_x\) and \(G = (2\pi/\lambda)\sin \theta \mathbf{u}_x\mathbf{u}_y\), with \(\lambda\), \(\mathbf{n}_{\text{eff}}\), and \(\theta\) the wavelength, effective refractive index, and incident angle with respect to the surface normal, respectively, and \(\mathbf{u}_x\) and \(\mathbf{u}_y\), the unitary vectors along the diffracted and incident directions, respectively.

\[
G = m(2\pi/p_x)\mathbf{u}_x + n(2\pi/p_y)\mathbf{u}_y,
\]

with \(p_x\) and \(p_y\) the lattice constants along the x- and y-directions, respectively, and \((m, n)\) are integers defining the diffraction order.

We restrict our investigation to the first diffraction order of the array. The dispersion of the (0, ±1) RAs is

\[
E_{\text{RA}(0,0)} = \pm \frac{hc}{n_{\text{eff}}} \sqrt{k_i^2 + \left(\frac{2\pi}{P_y}\right)^2}
\]

(1)

where \(k_i\) has only a component along the x-direction. The dispersion of the (±1, 0) RAs is given by

\[
E_{\text{RA}(1,0)} = \pm \frac{hc}{n_{\text{eff}}} \left(k_i \pm \frac{2\pi}{P_y}\right)
\]

(2)

with \(k_i = k_i\).

To generate high-Q SLRs, we have chosen Ag as the plasmonic material due to its low intrinsic (ohmic) losses in the visible range.\(^{[36,37]}\) The quality of deposited silver films has been studied in the past with various techniques.\(^{[38–40]}\) The aim of those studies was to enhance the propagation length of surface plasmon polaritons. However, how the different qualities of metal films modify the optical properties of SLRs has not been reported so far. To address this point, we first present a simple technique to modify the quality of the Ag film by tuning the evaporation rate. We then measure the extinction of different samples with different silver qualities to quantify its impact on the SLRs. Figure 1a compares the figure of merit (FoM) of a 80 nm thick silver film, defined as the ratio of the real and imaginary components of the permittivity, prepared at evaporation rates of 10 and 30 Å s\(^{-1}\), and the two most commonly used datasets found in the literature.\(^{[41,42]}\) This figure shows that it is possible to improve the FoM of the silver film by a factor of 2 over a large range of wavelengths (450–700 nm) by lowering the evaporation rate. The reduction of the metal losses can be associated with the decrease in surface roughness shown in the atomic force microscopy (AFM) images of Figure 1b,c. Particularly, the root mean squared (RMS) surface roughness of a silver film evaporated at 10 Å s\(^{-1}\) is 5.67 Å, which is three times lower than 30 Å s\(^{-1}\) where the RMS surface roughnesses are 17 and 19 nm, respectively.

Figure 1. a) Figure of merit of silver films, defined as the ratio of the real and imaginary components of the permittivity. b) AFM map of the surface of silver films deposited at 10 Å s\(^{-1}\); the surface roughness, defined in terms of the root mean square of the surface profile, is 0.567 nm. c) AFM map of a silver film deposited at 30 Å s\(^{-1}\) with RMS = 3.817 nm.
smaller than the RMS roughness of a similar film evaporated at 30 Å s$^{-1}$. The measurement techniques and optical constants of the films are described in more detail in the Experimental Section and Figure S1a,b in the Supporting Information. It should be noted that our best metal films are of lower quality than those of ref. [41], even when evaporated at almost the same rate and under improved vacuum conditions. This result stresses the relevance of characterizing the quality of deposited metals, instead of using the literature values. Another noticeable feature of the fabrication of the Ag films is that annealing does not improve the FoM, as is commonly assumed (see Figure S1c–e in the Supporting Information). In fact, the film quality decreases due to an increased surface roughness and dewetting of the Ag film on quartz substrate during annealing.

We have fabricated two arrays of silver nanorods deposited at 10 and 30 Å s$^{-1}$ to investigate the influence of the material quality on the linewidth of SLRs. The area of each array is 2.5 mm × 2.5 mm. These arrays were fabricated on a 1 mm thick quartz substrate and were subsequently embedded in a 200 nm thick polystyrene film. The pitch of the arrays along the short axis of the nanorods is $p_y = 340$ nm and along the long axis is $p_x = 420$ nm. The nanorod dimensions are 30 nm (width) × 100 nm (length) × 40 nm (height). Figure 2a displays the extinction spectra of the two arrays measured with an incident beam polarized along the short axis of the nanorods. The extinction is defined as $1 - T/T_0$, where $T$ is zero-order transmission of the sample and $T_0$ is the transmission of the reference.

As reference we use the substrate with the 200 nm film of polystyrene. The transmissions were measured with a fiber-coupled spectrometer with a multimode optical fiber. Mode coupling in the fiber led to the noise in the spectra, which was reduced by shaking the fiber during the measurements. We can see in Figure 2a that the wavelength of the SLR peaks does not change by changing the material quality. However, there is a 25% reduction of the linewidth by improving the material quality. Figure 2b shows the simulated extinction spectra of the arrays using finite differences in time domain (FDTD) and calculated with the permittivities of Figure 1a. The FDTD simulations show a good agreement with the measurements. The wavelength of LSPRs in the two samples is similar regardless of the different permittivities. Only the LSPR resonance position is slightly blueshifted (around 40 nm) by using the values from ref. [41]. This discrepancy can be explained by the abrupt film quality changes of ref. [41] around 450 nm. Another feature appearing in Figure 2b is the weak Fano resonance at 500 nm, which corresponds to the diffraction along the short axis of the unit cell.

An important parameter influencing the Q-factor and wavelength of SLRs is the detuning between RAs and LSPRs. For these experiments, we fabricated a sample consisting of the three arrays of nanoparticles shown in the scanning electron microscopy (SEM) images of Figure 3a–c (left column). The pitches ($p_x = 340$ nm and $p_y = 420$ nm) are fixed in these arrays, but the width ($w$) of the nanorods is varied ($w = 30$, 42, and 56 nm, respectively). The nominal length of the nanorods was kept constant during fabrication. However, there are small variations in this length due to the accuracy of the e-beam lithography procedure. We have measured a 27% shorter length for the nanorods with $w = 30$ nm than the nominal value of 100 nm, a 20% shorter length for the nanorods with $w = 42$ nm, and a 2% shorter length for the nanorods with $w = 56$ nm. Nevertheless, these variations do not alter the main results since these lengths of the nanorods are short enough to prevent the excitation of higher-order localized resonances. Figure 4a,b shows the measured and simulated extinction spectra of the arrays measured at normal incidence. The RAs can be seen in those spectra as a small dip at 615 nm for the three arrays. These figures show that the linewidth of the SLRs becomes narrower and it shifts toward the RAs as the LSPRs move outward of the RA frequency. Figure 4c illustrates this behavior quantitatively. Specifically, the Q-factors of the SLRs decrease from 325 to 98 and the resonant wavelength shifts from 648 to 638 nm, when the wavelength difference between the RA and the LSPRs decreases from 150 to 110 nm. These changes in Q-factor and resonance wavelength can be attributed to the change in coupling strength between the RA and LSPRs that can be altered by...
controlling their detuning.\textsuperscript{[43]} The behavior of the SLRs is less plasmonic as the detuning becomes larger, reaching a higher \( Q \)-factor. This response also translates in a weaker field confinement to the nanoparticles, which is illustrated by the SLR electric field distributions calculated at mid-height of the nanorod and illustrated in the right column of Figure 3a–c. It should be noted that the linewidth in the experiment (Figure 4a) is narrower compared to the simulation (Figure 4b). This narrower linewidth can be explained by a smaller effective size of the silver nanorods, very likely due to oxidation during a prolonged exposure to the atmosphere between the SEM measurements and the coating of the sample with a protective polymer (see the Experimental Section). We should also note that the narrow

Figure 3. a–c) SEM images (left column) of the nanorod array (scale bars correspond to 100 nm), and FDTD simulation of the normalized electric field of the SLR in a unit cell calculated in the middle plane of the nanoparticles (right column). All nanorod arrays have the same length, \( l = 98 \) nm, and height, \( h = 40 \) nm. The widths of the nanorods are a) \( w = 56 \) nm, b) \( w = 42 \) nm, and c) \( w = 30 \) nm.

Figure 4. a) Measured extinction spectra at normal incidence and for a polarization parallel to the short axis of the Ag nanorods of three arrays with the same periodicity (\( p_x = 340 \) nm, \( p_y = 420 \) nm), but different widths of nanorods, namely, \( w = 30 \) nm (blue), \( w = 42 \) nm (red), and \( w = 56 \) nm (purple). b) Simulated extinction spectra of the same structures as in part (a). c) Quality factor and wavelength of the SLRs as a function of the detuning between the LSPRs and the RA at normal incidence.
linewidths that we have measured are in agreement with the exponentially narrow resonances theoretically described by Markel,[44] and that result from the divergence of the effective extinction cross section per particle due to the retarded dipole moment of the adhesion layer in limiting the Q-factor, above 1500, is reached.

Figure 5 investigates the influence of the adhesive promoter on the linewidth of the resonance. Although several studies have reported on the effect of damping on the LSPRs,[46,47] none has reported so far whether these adhesion layers would modify the Q-factor of SLRs. Figure 5a shows the extinction of a nanorod array (20 x 100 x 42 nm³ with a 2 nm thick Ti adhesive layer) and the simulation using the geometric parameters of the experiments, measured permittivity of Ag and bulk permittivity of Ti.[41] The simulations include the coupling introduced by this conducting sublayer. The Q-factor drops significantly (a factor of 3) when we use a very thin layer of Ti as adhesive. The discrepancy between the experimental results and the simulations can be attributed to the inhomogeneous size distribution of Ag nanoparticles, imperfections on the surface of the nanoparticles, and uncertainties in the permittivity of the adhesion layer. We have also simulated the extinction of a similar sample in which the adhesion layer is suppressed. The result of this simulation is also shown in Figure 5, where an extraordinary large value of the Q-factor, above 1500, is reached. These numerical results illustrate the significant role of the adhesive layer in limiting the Q-factor in plasmonic systems and motivate a quest for new materials to replace the traditional adhesive materials.

We have also investigated the in-plane momentum dependence of the SLRs by measuring the extinction at different angles of incidence with respect to the sample normal. The extinction measurements were performed with a motorized rotational stage schematically represented in Figure 6. The samples were placed vertically on the stage and rotated around the vertical axis in steps of 0.25° in a range from −30° to 30°. The rotation axis was parallel to the length of the nanorods to measure the (0, ±1) SLRs and parallel to the width of the nanorods to measure the (±1, 0) SLRs. A polarizer was placed in front of the sample (not shown in Figure 6) to fix the polarization of the incident beam parallel to the width of the nanorods. The source was a collimated beam of white light (Energix LDLS EQ-99). The transmitted intensity in the forward direction was collected by a lens (NA = 0.04) and sent to the fiber-coupled spectrometer.

The extinction measurements are shown in Figure 7, where angle scans of the extinction of SLRs arising from the (0, ±1) RAs (Figure 7a) and of SLRs arising from the (±1, 0) RAs (Figure 7b) are displayed. These measurements show the dispersion of the SLRs as a function of the in-plane wave vector. The SLRs in Figure 7a show a quadratic dispersion and a high Q-factor. Because of the weak dispersion of these SLRs, their Q-factor is nearly independent of the angle of incidence. This response can be appreciated in the extinction spectra measured at different angles and displayed in Figure 7c. The SLRs of Figure 7b present a different behavior. Their dispersion is nearly linear, and the Q-factor depends on the angle of incidence, as can be appreciated in Figure 7d. This Q-factor decreases by a factor of 3 as we move 15° away from the normal. These differences in Q-factor are attributed to the different dispersion of the SLRs, which changes the coupling strength between the LSPRs and the RAs for different angles of incidence or in-plane momenta. As discussed above, the coupling strength between LSPRs and RAs defines the plasmonic content and the losses of SLRs. The losses in the metal due to interband transitions also limit the Q-factor of SLRs. Hence, it is very challenging to achieve high-Q SLRs at short wavelengths. This is illustrated in Figure S2 in the Supporting Information, which shows SLRs of the same sample but at shorter wavelengths (around 500 nm). These SLRs, with a lower Q-factor, arise from the coupling of LSPRs to the RAs that result from diffraction with the short pitch of the array (pₛ = 340 nm).

We have demonstrated high quality factor plasmonic resonances in arrays of Ag nanoparticles (Q > 300). These resonances, known as surface lattice resonances, emerge from the coupling of localized surface plasmon polaritons to diffraction orders in the plane of the array. The quadratic dispersion of SLRs leads to a nearly constant Q-factor over a wide range of wave vectors or angles of incidence. We have investigated the role of the intrinsic quality of the metal in the Q-factor of SLRs. We have also discussed the effect of the adhesion layer...
used between the substrate and the metal on the SLRs. The suppression of this layer can lead to SLRs with $Q$-factors larger than 1500. These extremely high $Q$-factors render arrays of metallic nanoparticles very interesting systems for plasmonic applications such as sensors, for enhanced light–matter interaction and nonlinear phenomena.

Figure 6. Schematic representation of the setup used for the measurements of the dispersion of the extinction. Note that the coordinate frame used to characterize the $(0, \pm 1)$ and $(\pm 1, 0)$ RAs is defined on the array, with $\hat{u}_x$ and $\hat{u}_y$ along the short and long axes of the nanorods, respectively. Therefore, the sample is rotated along the short nanoparticle axis to measure the dispersion of the $(\pm 1, 0)$ RAs (vertical axis as in the figure). For the $(0, \pm 1)$ RAs, the sample is rotated along the long nanoparticle axis, which corresponds to changing the orientation of the sample by 90° in the setup and rotating it along the vertical axis.

Figure 7. a) Extinction spectra as a function of the wave vector parallel to the surface, showing the dispersion of the degenerate SLRs resulting from the coupling of LSPRs in Ag nanoparticles to $(0, \pm 1)$ RAs. The LSPR at $\lambda = 475$ nm is independent of the wave vector, whereas the white curve corresponds to the $(0, \pm 1)$ RAs. b) Extinction spectra showing the dispersion of the diagonal SLRs arising from the coupling of the LSPRs to the $(\pm 1, 0)$ RAs (indicated by the white lines). c) Extinction spectra from part (a), measured at different angles. d) Extinction spectra from part (b), measured at different angles. The insets in parts (a) and (b) show a schematic representation of the angle-dependent extinction measurements, where the rectangles represent the nanorods, the orange double arrow indicates the polarization of the incident light, and the white curve illustrates the rotation direction of the sample.
Experimental Section

Sample Preparation: The silver nanoparticle arrays were fabricated by conventional electron beam lithography (Raith EBPG 5250), followed by metal deposition and a lift-off process. A piece of 2.5 x 2.5 cm² quartz substrate (Plan Optik) was coated with a 120 nm thick layer of electron beam resist ZEP520A. A pair of three different patterns (area of each pattern was 2.5 x 2.5 mm²) were defined on a single sample, on which silver (Alfa Aesar GmbH, 99.999%) was selectively deposited at rates of 10 and 30 Å s⁻¹ under the initial vacuum (∼3 x 10⁻⁶ Torr). An adhesion layer (Ti 2 nm) was used to attach silver particles to the substrate. A 4 wt% polystyrene in toluene was used to coat a thin layer of polystyrene at low spinning speed and baked at 135°C to prevent degradation of the layer. (Ti 2 nm) was used to attach silver particles to the substrate. A 4 wt% polystyrene in toluene was used to coat a thin layer of polystyrene at low spinning speed and baked at 135°C to prevent degradation of the layers. The layer thickness was kept to only 200 nm to avoid quasi-guided modes that complicate the analysis. The layer also helps in achieving a more homogeneous dielectric surrounding of the nanorods, which increases the coupling to SLRs. [13]

Metal Characterization: The morphology of 80 nm thick silver films (evaporated at rates of 10 and 30 Å s⁻¹) on a quartz substrate was examined with atomic force microscopy (Nanoscope Dimension 3100) at a scanning frequency of 0.5 Hz. The optical constants of the films were determined with a spectroscopic ellipsometer. The ellipsoid used in this study was a Woollam M2000 F1 with a spectral range of 250-1700 nm and it was used at 60° angle of incidence with respect to the surface normal. The measured ellipsoid parameters have been converted directly into permittivity with the help of the pseudo-optical constant transformation integrated into the used CompleteEase software. These calculations are described in detail by Oates et al.[40]

FDTD Simulations: The optical extinction of the nanoparticle pattern was 2.5 mm² were defined on a single sample, on which silver nanoparticles. For the different simulations, the permittivity values from refs. [41,42] or measured using ellipsometry were used.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

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