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Scaling of losses with size and wavelength in nanoplasmonics and metamaterials

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We show that, for the resonant metal-dielectric structures with sub-wavelength confinement of light in all three dimensions, the loss cannot be reduced considerably below the loss of the metal itself unless one operates in the far IR and THz regions of the spectrum or below. Such high losses cannot be compensated by introducing gain due to Purcell-induced shortening of recombination times. The only way low loss optical meta-materials can be engineered is with, as yet unknown, low loss materials with negative permittivity. © 2011 American Institute of Physics. [doi:10.1063/1.3664105]

Recent years have seen significant progress in two inter-related fields of nanoplasmonics (NP) and metamaterials (MMs).[1–3] These fields rely upon the most remarkable feature of sub-wavelength metallic objects—a high degree of concentration of energy achievable in their vicinity, well beyond the concentration allowed by the diffraction limit. In turn, the ability to concentrate energy near these "artificial atoms" allows one to arrange them in a regular manner and thus engender new MMs with optical properties that are unattainable in nature, the elusive negative index[3] being just one of them. While significant steps in developing functional NP and MM devices have been made, widespread practical implementation of them has been impeded by many factors, the most significant of which remains the inherent loss associated with the absorption in the metal. It is well known that the rate of energy loss in the metal, determined mostly by electron-phonon and to a lesser degree electron-electron scattering inside the conduction band, is on the order of $2\gamma \sim 10^{14}\,\mathrm{s}^{-1}$ for noble metals and it gets even larger at shorter λ's where the band-to-band absorption arises. In the Drude model (DM) approximation, one can represent the dielectric constant of a metal as $\varepsilon(\omega) = \varepsilon_r - j\varepsilon_i = 1 - \omega_p^2/(\omega^2 + j\omega\gamma)$, where ω_p is the plasma velocity and γ is the velocity relaxation rate, which is exactly one half of the energy loss rate. The quality factor of metal $Q_m = |\varepsilon_r|/\varepsilon_i \approx \omega/\gamma$ is less than 40 even under most optimistic projections and in reality far worse than that once surface scattering has been factored in. Q_m in turn determines the Q of the whole device and thus its ability to perform its requisite function. While significant efforts are being devoted to the search for alternative negative materials, such as highly doped semiconductors[4] or more exotic ones[5] as well as to compensating the loss with gain,[6] such efforts have not yet yielded practical results. While reduced total losses for different material combinations and λ's have been widely reported, these results are not systematic and often are no more than the consequence of moving to longer λ's, where interband absorption can be avoided. Overall, the most impressive MM results have been obtained in the THz region of the spectrum and beyond,[7] while results in the visible and especially UV have been far more modest. This problem has been first tackled in the pivotal works[8,9] where the difficulty in scaling

to the short λ's is explained by the rise of "kinetic inductance" associated with the inertia of electrons in small nanostructures. But these studies explain only one side of the scaling difficulties associated with the inertia of electrons—no systematic study of limitations associated with electron-related loss has been made. In this work, we perform a simple study of the Q-factor of two representative NP and MM elements—a split ring resonator (SRR) and an elliptical nanoparticle (ENP)—as a function of their dimensions and λ and demonstrate that the apparent improvement of Q in the mid IR is purely material-related (avoidance of interband absorption), while the improvement in the THz region and beyond is of a more fundamental nature (i.e., present in DM) and is associated with an increase in the conductivity current relative to the displacement current. The point of this study is to show that high Q-factors in optical/plasmonic regime $\omega \gg \gamma$ are fundamentally unattainable with existing metals.

A simple general rationalization of this statement can be given before we embark on the more involved derivations using the Maxwell equation $\nabla \times \boldsymbol{H} = j\omega\varepsilon\varepsilon_0\boldsymbol{E} = j\omega\varepsilon_r\varepsilon_0\boldsymbol{E} + \sigma\boldsymbol{E}$, where $\sigma = \omega\varepsilon_i\varepsilon_0$ is the conductivity. Now, if we assume that the scale on which the electric field is confined is roughly a, integrating over the area πa^2 and invoking Stokes theorem yields this relation for the field magnitudes

$$|H| \sim \frac{a}{2}|j\omega\varepsilon\varepsilon_0 + \sigma||E| = \frac{a\omega}{2}\sqrt{\varepsilon_r^2 + \varepsilon_i^2}\,\varepsilon_0|E|. \qquad (1)$$

Now, near the resonance, E and H (with current J) are oscillating 90° out of phase with each other. Hence, half of the time all the energy is stored in electro-static form $U_E = \varepsilon_0^2\varepsilon_r^2E^2V/2$, while the other half of the time the energy is expected to be stored in the magnetic form $U_M = \mu^2\mu_0^2H^2V/2$. Their ratio is

$$\frac{U_M}{U_E} \sim \frac{\mu_0\mu|H|^2}{\varepsilon_0\varepsilon_r|E|^2} \sim \left(\frac{\pi n a}{\lambda}\right)^2\left(1 + \frac{\varepsilon_i^2}{\varepsilon_r^2}\right) \sim \left(\frac{\pi n a}{\lambda}\right)^2\left(1 + \frac{\gamma^2}{\omega^2}\right), \quad (2)$$

where n is the refractive index. It is easy to see from here that, as long as $\varepsilon_i \ll \varepsilon_r(\omega \gg \gamma)$ and the confinement scale $a \ll \lambda/n$, the magnetic energy is but a tiny fraction of the electro-static energy (the well-known static limit) and, from an energy conservation point of view, it follows that half of the time practically all the energy is stored in the kinetic

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motion of electrons where it is inevitably lost at the rate 2γ. Hence, for truly sub-λ confinement, the rate of energy loss near resonance is always close to γ as is indeed confirmed by calculations for spherical nanoparticles near surface plasmon (SP) resonance.[10] The electrostatic limit, however, is no longer valid at lower frequencies $\omega \sim \gamma$ and less, as conduction rather than displacement current becomes dominant and H is no longer inversely proportional to λ. Thus, a smaller fraction of energy is contained in the kinetic motion of electrons and, rather ironically, as the Q_m decreases below unity, the Q of the whole device increases.

To confirm and quantify these predictions, we first consider an example of an SRR shown in Fig. 1(a) that consists of a ring of radius a made of a cylindrical wire with radius r and one or more gap capacitors C that are treated as adjustable to provide a required resonant frequency $\omega \sim 1/\sqrt{LC}$. When current flows through the wire energy is stored in two forms—magnetic energy $U_M = L_M I^2/2$, where $L_M \approx \mu_0 a \ln(8a/r)$ is the conventional (magnetic) inductance and the kinetic energy of the electrons $U_K = L_K I^2/2$, where the kinetic inductance is $L_K = 2\pi a/A\varepsilon_0\omega_p^2$,[8] and A, the area over which the current flows subject to skin effect, can be found as $A(\omega) \approx \pi r^2 \left(1 + \pi r/\lambda_p \sqrt{1 + 2/Q_m(\omega)}\right)^{-1}$. At resonance, the total energy alternates between potential (electro-static energy in the capacitor) and kinetic, i.e., $U_M + U_K$. Therefore the dissipation rate of the whole structure is $\gamma_{SRR} = \gamma L_K/(L_K + L_M)$. The λ-depended Q is then

$$Q_{SRR}(\lambda_\gamma) = \frac{1}{\lambda_\gamma\gamma'} + \frac{2\pi^2}{\gamma'}\ln\frac{8}{\beta}\beta a_\lambda Q_{m0}$$
$$\times \left[\frac{1}{\beta a_\lambda \lambda_\gamma Q_{m0}} + \frac{\pi}{\sqrt{1+2\lambda_\gamma\gamma'}}\right]^{-1}, \quad (3)$$

where $\beta = r/a$ is the diameter/thickness ratio, $a_\lambda = a/\lambda$ is the "sub-wavelength" parameter of the structure, $Q_{m0} = \omega_p/\gamma$ is the maximum Q of metal in the DM (of course in real metal, this Q is never achieved since near plasma frequency losses increase dramatically—it is used just as a parameter here), and $\lambda_\gamma = \lambda/\lambda_b = Q_m^{-1}(\lambda)$ is the ratio of λ to the "border wavelength" $\lambda_b = 2\pi c/\gamma$ at which $\omega = \gamma$, i.e., what we can loosely define as a border between electronics and optics. Parameters for the DM of gold[11] ($\omega_p = 13.06 \times 10^{15}\,s^{-1}$, $\gamma = 12.3 \times 10^{13}\,s^{-1}$) yield $Q_{m0} = 110$ and $\lambda_b \sim 15\,\mu m$. Finally, the factor $\gamma' = \gamma(\omega)/\gamma$ accounts for the discrepancy between the actual metal permittivity[11] and the DM.

In Figs. 2(a) and 2(b), the results for Q_{SRR} and the ratio of the effective loss rate $\gamma_{eff}(\lambda) = \omega/Q_{SRR}(\lambda)$ to the DM value of
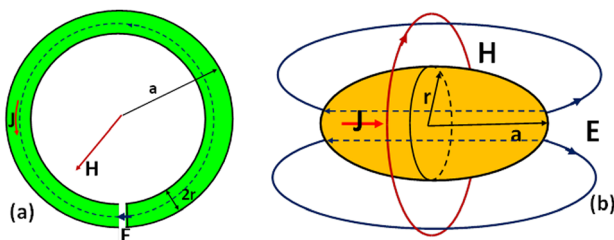
the loss in metal are shown as functions of wavelength for a rather thick wire $\beta = 0.25$ and four different values of a_λ. The dashed lines are obtained using the DM while the solid lines represent results obtained using experimental values of gold permittivity.[11] As one can see, the initial sharp increase in Q_{SRR} as λ moves from visible to near IR is associated solely with the intrinsic reduction of metal loss as photon energy becomes insufficient to cause the interband transitions in the metal. But once the metal loss settles at a constant DM value, the improvement in Q_{SRR} becomes less dramatic for larger SRR's ($2a = \lambda/4$) and is reversed for smaller, truly sub-λ SRR's. The Q_{SRR} starts recovering only in the vicinity of the "border wavelength" and from there increases roughly as $\lambda^{1/2}$ as the very last term in Eq. (3) becomes dominant. The behavior of $\gamma_{eff}(\lambda)$ plotted in Fig. 2(b) is similar—the initial rapid decrease is followed by an essentially flat region when $\gamma_{eff}(\lambda) \approx \gamma$, exactly as predicted above, and only when wavelength exceeds λ_b, i.e., when one is essentially in electronic rather than optical domain, the loss decreases as roughly $\lambda^{-3/2}$.

Next we consider the losses in the prolate ENP (Fig. 1(b)), used extensively for local field enhancement,[1] with large half-axis a and two equal small half-axes r. Unlike the SRR where ω can be tuned over a wide range by adjusting the gap(s), in the ENP, the resonant frequency is determined only by the eccentricity. As Fig. 3 shows for all values of $2a/\lambda$, the effective loss rate after the initial dive quickly settles at exactly γ and the Q factor accordingly falls as λ^{-1}. This is simply the consequence of the fact that in order to shift resonance to a long λ, one has to reduce the ellipsoid cross-section to very low values and thus increase L_K. For field enhancement, clearly ENP's work best at the short λ's that are just long enough to avoid the band-to-band absorption in the metal, e.g., close to $1\,\mu m$ in gold. At these λ's, their Q_{ENP} can be as large as or a bit larger than Q_{SRR}, which makes ENP ideally suited for sensing. At longer λ's, it is preferable to use SRR or some structures in between, for example, nano-antennas with gaps between them to provide adjustable C to tune the resonance into the red. Nevertheless, it is fair to say that at long wavelengths, their Q would not exceed than that of an SRR. Hence, the key conclusion of our work remains valid and states that in true

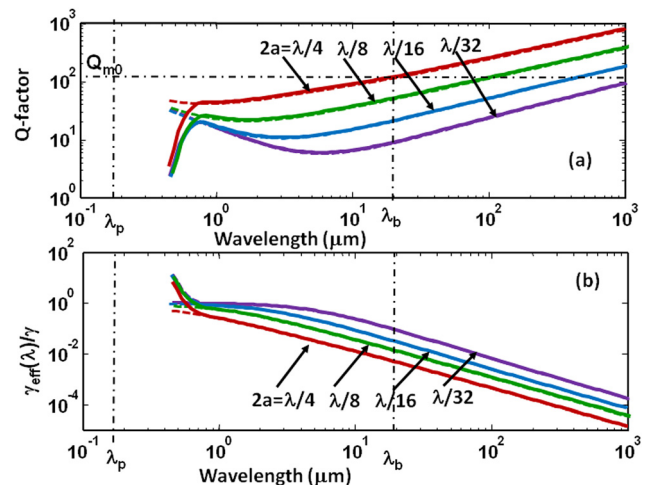FIG. 1. (Color online) Geometries and fields for (a) split ring resonator and (b) elliptical nanoparticles.

FIG. 2. (Color online) Q-factor (a) and effective loss rate (b) in the SRR with $r = a/4$ and different diameters $2a$. Solid lines: exact values of ε_M, dashed lines: Drude model.

sub-wavelength metal-dielectric structures, the loss cannot be reduced significantly below the one half of the metal loss for as long as the frequency is larger than metal scattering rate.

One can extend these results to propagating SP polaritons (SPP) on the metal-dielectric boundaries, where confinement in the interface plane is measured by the SPP's wavelength that near resonance becomes very short. Unfortunately, once that wavelength becomes just a few times shorter than λ/n, the loss will inevitably become equal to precisely one half of the metal loss, i.e., γ.[12] Once again, it is essential to state that our results are applicable only to the case of sub-wavelength confinement in all three dimensions (3D)—the moment the mode becomes comparable to $\lambda/2n$ in just one direction[13–17] H-field becomes large enough to suck the energy out of the kinetic motion of electrons and the loss is greatly reduced, in fact to the point where it can be compensated by the gain in the dielectric and lasing can be attained in small volumes, less than $(\lambda/2n)^3$.[13,14,17] Yet if one examines the reports of sub-λ lasing, one cannot avoid noticing that, at least in one dimension, the mode size always remains larger than $(\lambda/2n)$.

Although SP "spasing"[18,19] is not the main subject of this work, one can still make a simple order-of magnitude estimate of what it would take to compensate the projected modal loss $\gamma_{eff} \sim 10^{14} \mathrm{s}^{-1}$ in a true sub-λ mode. In a semiconductor gain medium, obviously, a very high injection density of the order of $10^{19} \mathrm{cm}^{-3}$ would be required which is high, but not-unattainable provided the spontaneous recombination rate (RR) is reasonably slow. But this, regrettably, goes against the physics of sub-λ cavities where spontaneous RR is greatly enhanced by the Purcell effect[20] (along with the stimulated emission rate). Hence, the injection rate (current) required to maintain carrier density becomes prohibitively high. In fact, simple rate equation for the number of SPP's in the mode (similar to laser rate equation[21]), $\dot{n}_{SPP} = g_{eff}(n_{SPP} + 1) - \gamma_{eff} n_{SPP}$, shows that when modal gain g_{eff} is close to compensating the loss, the spontaneous RR approaches $\gamma_{eff} \sim 10^{14} \mathrm{s}^{-1}$, meaning that injection current $i_{inj} \sim e\gamma_{eff} \sim 10\ \mu\mathrm{A}$ must flow into the very small modal volume. For the 50 nm mode that corresponds to more than 1MA/cm^2, more than two orders of magnitude higher than in a typical diode laser.[22] Thus, we cannot perceive how the

true sub-λ in all 3D "spaser"[18,19] can be feasible at optical λ's, at least when it comes to its most practical electrically pumped implementation. The corresponding power densities of 10^7–$10^8\ \mathrm{W/cm}^2$ can only be achieved in pulsed regime of high power lasers as in Ref. 23, but these technique does not easily lend itself to miniaturization.

In conclusion, we have shown that when one operates with sub-λ confinement $a \ll \lambda$ in 3D and at frequencies that are higher than the scattering rate in the metal ($\omega \gg \gamma$), i.e., in what is referred to as "plasmonics" rather than "optics" ($a \geq \lambda$) or "electronics" ($\omega \leq \gamma$) regime, no amount of clever engineering can reduce the modal loss significantly below γ. In hindsight, this conclusion appears obvious, as in order to reap all the benefits of "plasmonics" a significant fraction of energy must constantly flow in and out of "plasma" (i.e., the inherently lossy motion of free electrons in the metal). Yet this fact is not commonly recognized, so we hope that our work will show the limits of engineering the shape of NP and MM structures. Furthermore, we have pointed out that since the recombination time in sub-λ structures is shortened by the Purcell, the pump rates required to compensate the loss with gain appear to be impractically high. That leaves finding a better material with negative ε the only viable alternative to bring losses in the plasmonics down. In the absence of such material, NP and MM's still can find use in selective niches such as sensing, where loss is not the key factor.
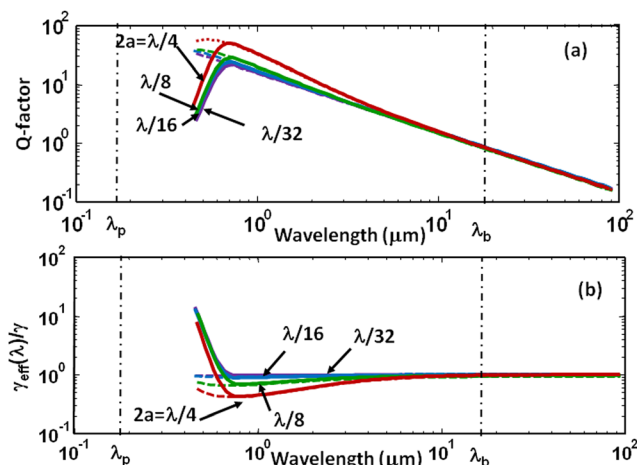
FIG. 3. (Color online) Q-factor (a) and effective loss rate (b) in the ENP of different lengths 2a. Solid lines: exact values of ε_N, dashed lines: Drude model.

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