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Can We Make a Nanoscopic Laser?

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Abstract—It is argued that, by exploiting the quasi-static eigenstates in a metal/dielectric composite medium, it should be possible to construct a new type of device called a SPASER (surface plasmon amplification by stimulated emission of radiation), where a strong, coherent ac electric field is built up in a spatial region whose linear size is much smaller than the wavelength.

An essential component of most types of lasers is an optical resonator. This is needed in order to have states of the electromagnetic (EM) field which are approximate eigenstates, where the field intensity can be built up to large values as a result of many stimulated, and therefore coherent, emissions of photons. Usually, these resonators are macroscopic in size and are constructed using good mirrors for reflecting EM waves. In recent years, the phenomenon of "random lasing" was discovered and studied [1–3]. This phenomenon is due to the occurrence of small optical resonances in a collection of transparent dielectric grains. Even in this case, the resonators and resonance states cannot be smaller than the wavelength of light. This places a seemingly strong lower bound on how small a laser can be. Naturally, this lower bound is even greater in the case of infrared lasers than in the case of visible light lasers. We now argue that it should actually be possible to overcome this apparent limitation and build a nanometer sized laser where a strong coherent EM field can be built up, and eventually radiated, whose frequency is in the infrared or visible region of the optical spectrum.

The basic premise upon which our argument is based is that in heterogeneous media there exist quasi-static resonances of the EM field whose spatial smallness is almost unlimited. These resonances are eigensolutions of Maxwell's equations in the static or quasi-static regime. Such an eigenstate is represented by an electric potential field $\phi(\mathbf{r})$ that satisfies the equation

$$\nabla \cdot \hat{\varepsilon} \cdot \nabla \phi = 0$$

and vanishes over the entire external surface or boundary of the system. Here, $\hat{\varepsilon}$ is the electric permittivity, which in the general case is a position-dependent second-rank tensor with components that are complex and frequency-dependent.

The simplest case of such quasi-static eigenstates occurs in a macroscopically heterogeneous mixture of two separately uniform constituents with scalar permittivities ε_1, ε_2. In such a composite medium, there exists an infinite set of resonances or eigenstates; these are characterized by special negative values of the ratio $\varepsilon_1/\varepsilon_2$ or by special values of the related materials parameter $s \equiv \varepsilon_2/(\varepsilon_2 - \varepsilon_1)$ which lie on the real, semi-closed segment [0, 1) of the real axis [4–6]. Of course, no real composite medium can ever be found in such a resonance state, since no real material can have an electrical permittivity that is real and negative—if the real part is negative, then there must also be a nonzero imaginary part. However, if the imaginary part is small, then the system can be excited to a state that is close to such a resonance. The best case known to us is when metallic silver (Ag) inclusions are embedded in a conventional dielectric host and the frequency of the EM field is in the near infrared or visible range. In those ranges, the real part of the electric permittivity of Ag is negative, large, and frequency-dependent, while the imaginary part is as small as 0.18 [7]. An important aspect of the quasi-static resonances in such a composite medium is that they do not depend on any of the characteristic EM lengths, such as wavelength in the dielectric constituent or skin depth in the metallic constituent. In fact, they are independent of all physical properties of the constituent materials and depend only on the microgeometry of the interface between those constituents.

In numerical studies, we discovered that, in a two-constituent composite medium with a disordered microstructure, many of the quasi-static eigenstates are strongly localized—the electric field has nonnegligible values only in a limited region of space, whose size is determined only by the detailed shape of the interface [8]. Because of this, it appears that microstructures can be found where quasi-static resonances exist whose size is as small as we like, as long as it is still valid to use electric permittivity and the continuum limit for the description of the EM materials response.

These resonances have some peculiar properties. (a) Because of the quasi-static nature of the fields, the magnetic component is negligible—the field is almost exclusively an electric field. This is in marked contrast with the states in a conventional EM resonator, where

the electric and magnetic fields have comparable magnitudes. (b) The small size of the resonance is achieved by an effective dynamic screening of the electric field outside the resonance. This is done primarily by the conduction electrons in the metal, which are free to move and readjust their self-field so as to nullify the total field. This is the same physical phenomenon that is responsible for the collective excitations known as plasmons, except that now the response is strongly affected by the metal/dielectric interface. That is why these quasi-static resonances are also commonly known as "surface plasmon resonances."

These properties mean that, in contrast with conventional optical resonators, where all the action is in the EM field and everything could take place even in a vacuum, the quasi-static resonances require active and essential dynamic participation of the conduction electrons in the metallic constituent. That is why we call the process which occurs when such a resonance builds up its amplitude, by interacting with an appropriate set of two-level excited systems, by the name "surface plasmon amplification by stimulated emission of radiation (SPASER)" instead of by the better known name "light amplification by stimulated emission of radiation (LASER)" [9].

Another important point to consider is how a SPASER can interact with a regular incident EM field, and whether or how it can emit such a field. Indeed, the localized quasi-static resonances do not interact with such regular EM fields in the quasi-static approximation. However, beyond that approximation, there will be a weak interaction. This is like having a conventional LASER with optical mirrors forming the resonator that have a very low, but not zero, transmissivity. This is desirable because it allows the field intensity to build up inside the resonator before being extracted for outside use. The same can be said of the localized quasi-static resonances, except that now we also need to worry about dissipation: Because the electric permittivity of the metallic constituent has a finite imaginary part, the value attained by the physical parameter s can never be exactly equal to any resonance value s_n (recall that these values are real and satisfy $0 \le s_n < 1$). At best, the difference $s - s_n$ will be small and imaginary. Consequently, the resonance, once excited, will decay by dissipation (i.e., EM energy will disappear and reappear as internal energy and increased entropy of the composite material) with a characteristic exponential dissipative decay time τ_d given by [9]

$$\tau_d = \left[\frac{1}{\mathrm{Im}\, s(\omega)} \frac{d\,\mathrm{Re}\, s(\omega)}{d\omega}\right]_{\omega = \omega_n},$$

where ω_n is the real part of the complex frequency Ω_n defined by $s(\Omega_n) = s_n$.

In addition to this, the resonance field will also decay by radiating a real EM wave to the far-field region. Since the electric dipole moment of a dark, localized quasi-static resonance vanishes [8], this radiation can only happen due to higher multipole moments of that state—the next higher moments are magnetic dipole and electric quadrupole moments. These lead to a radiated power that is smaller in magnitude by a factor of order $(ka)^2 \ll 1$ (a is the size of the eigenstate, $k = 2\pi/\lambda$ is the wave number) compared to the electric dipole radiation if that were not zero. The radiative lifetime τ_r of the eigenstate due to these two moments will be of order

$$\tau_r \sim \frac{T}{\varepsilon_m (ka)^5},$$

where $T = 2\pi/\omega$ is the period of the EM field and ε_m is the electric permittivity of the metallic constituent. This radiative lifetime is greater by a factor of $1/(ka)^2$ than would be the lifetime due to electric dipole radiation if that did not vanish.

This weak interaction of the resonance with the EM "far field" is most useful if it is just strong enough to bring about radiative decay of the resonance in a time comparable to the dissipative decay time described above. For $T = 10^{-14}$ s ($\lambda = 3$ μm) and $ka = 0.01$, we thus get

$$\tau_r \cong \frac{10^{-4}}{\varepsilon_m} \text{ s}.$$

It will also be interesting to consider microstructures where dark localized states exist for which not only the electric dipole moment but also the magnetic dipole and electric quadrupole moments vanish. In that case, the radiative decay will be due to electric octupole or magnetic quadrupole radiation and we will have

$$\tau_r \sim \frac{T}{\varepsilon_m (ka)^7}.$$

The radiative lifetime of such a state will be much greater than previously calculated. Eigenstates with zero values for the electric dipole, magnetic dipole, and electric quadrupole moments exist in the case of an isolated spherical inclusion (see below). It will be interesting to explore their occurrence also in the case of more complicated inclusion shapes.

Another important issue is the spatial extent of the resonances and how that size depends on various microstructural features of the metal/dielectric composite. In the case of an isolated metal sphere embedded in a dielectric host, the resonance potential field or eigenfunction is always a spherical harmonic: Inside the sphere, the potential is given by the "regular spherical harmonic" $r^l Y_{lm}(\Omega)$ (r and Ω are measured relative to the sphere center), but outside the sphere it is given by the "singular spherical harmonic" $r^{-l-1} Y_{lm}(\Omega)$ [5]. For large l, this means that the spatial extent of the eigenstate is essentially equal to the size of the sphere. It is expected that, for isolated, oddly shaped metal inclusions, the grain size again determines the spatial extent of the eigenstates. This is certainly so in the case

of the higher eigenstates, i.e., those with increasingly large eigenvalues s_n and an increasingly oscillatory nature of the associated potential field $\phi_n(\mathbf{r})$.

Interestingly, numerical simulations have shown that, in more complex microstructures, there sometimes appear quasi-static eigenstates whose size is much smaller than the typical inclusion size. For example, in the case of a V-shaped metallic inclusion, there are eigenstates which are strongly localized near the tip or apex of the V shape [8]. We conjecture that what determines the size of such a highly localized eigenstate is the radius of curvature of the metal/dielectric interface in the vicinity of that eigenstate. This question needs to be studied by further calculations.

In order to build up the electric field in a quasi-static eigenstate with characteristic frequency ω, we need to have a collection of excited two-level systems with an energy gap equal to $\hbar\omega$ that can be induced to emit their energy coherently into this eigenstate. In practice, this can be done in various ways. Our suggestion is to make the dielectric host a compactified collection of semiconducting quantum dots (QDs) with an appropriate gap in their spectrum of eigenstates. These can be excited either by a strong incident uniform EM field or by an electric current flowing through the host. Of course, only those QDs that lie inside the limited spatial extent of the eigenstate will be able to interact with it and excite it. Quantitative estimates showing that this can work appeared in [9].

The ideas we described here, when they are implemented, will push down the lower limit on the size of optical or near infra-red fields to the nanometer realm. Electric field states of that size would open up the possibility of constructing an electric nanoprobe for studying nanofeatures of solid surfaces. A strong coherent electric field of that size, which would be available in a SPASER, would open up the possibility of imprinting comparably small changes onto a solid surface. For these reasons, it is important to test these ideas experimentally and find out whether they can be implemented in practice.

In summary, we believe it should be possible to construct a device similar to a LASER, called a SPASER, with a spatial size much smaller than the wavelength of the EM field, in which a high intensity coherent optical electric field can be built up. Such a device would have many interesting applications. Experiments to test this expectation, as well as some other predictions, would be very desirable.

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