Generalization of the effects of high Q for metamaterials

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Received March 12, 2013; revised May 13, 2013; accepted May 13, 2013; posted May 14, 2013 (Doc. ID 186906); published July 19, 2013

We revisit the electrodynamics of resonant high-Q interactions in atomic systems with a view to gaining insights into the design of meta-atoms and hence bulk metamaterials with profoundly different electromagnetic responses. The relevance of phase coherence and nonlinearity in charged systems is emphasized, as is the need to take care over defining how one specifies effective boundaries and cavities that ultimately determine light–matter interactions. Radically new material properties become apparent once one designs organized clusters of small numbers of atoms or meta-atoms for which the usually applied random phase approximation (RPA) does not apply. The RPA relies on averages in sufficiently large volumes consisting of large numbers of interacting systems, while our model assumes a small volume with averages in time, i.e., ergodicity. New meaning is given to the concept of effective and practically useful constitutive parameters, based on this very fundamental point of view, which is important to metamaterials. © 2013 Chinese Laser Press

OCIS codes: (160.3918) Metamaterials; (160.4670) Optical materials.

http://dx.doi.org/10.1364/PRJ.1.000077

1. INTRODUCTION

Our objective is to tie together several fundamental concepts that link light–atom interactions, light–nanostructure interactions, and the broader descriptions of electromagnetic interactions with small antennas and meta-atoms at any frequency. The treatment described here is predominantly classical in nature and heavily reliant on the consequences of satisfying Maxwell's equations during interactions, while recognizing that all wave–particle interactions ultimately involve a discrete exchange of energy and momentum. The concept of quantizing the energy and momentum of electromagnetic waves interacting with matter led to the correct description of the radiation spectrum emitted by a blackbody. This primal understanding of light–matter interactions has lead to tremendous advances in light sources (lasers) and detectors. It is foundational and guides how we describe interactions involving both naturally occurring atoms in materials and artificial or meta-atoms occurring in metamaterials. However, in this paper, we begin by revisiting the Lorentz model of the interaction of an atom with light and its role in the treatment of transitions between eigenstates and radiative emission, using a quantum mechanical description of interaction. We consider the concept of a high-Q region containing an atom defining an electrostatic frequency quite similar to a plasma frequency and generalize this concept to illustrate the importance of some kind of cavity, real or virtual, that dictates the electromagnetic response. A bulk response is governed by causality and Kramers–Kronig dispersion relations, but clearly the correlation between component entities plays an overriding role, leading to descriptions we typically accept to be averaged.

For a single hydrogen atom, the interaction volume can be defined in various ways. We demonstrate how, regarded as a cavity with an observed Q, this naturally suggests that cavity to be a sphere with a radius found to be comparable to its Bohr or nonradiating radius. The time required to excite a hydrogen atom to transparency, i.e., to transition to an excited state, defines Q but also directly suggests the Maxwellian interaction mechanism. The observed high gain factor is described in terms of a high value of Q resulting from a complex resonance, which can be explained in terms of the cavity defined by the set of basis functions employed to represent the incident light. Further, we describe a "triple" resonance that exists in nature and that can be applied to the development of structured engineered materials or metamaterials. It is this analysis that leads to the expectation that the most interesting and high-impact engineered materials will comprised small numbers of highly organized and specific meta-atoms.

The interaction of an isolated dipole with light has served to describe the interaction of light with dipoles in solids and metamaterials, as well as gases and liquids. However, strong interactions between neighboring elements can be expected to require a multipolar rather than dipolar analysis when those elements are very close together, i.e., separated by distances a lot less than the wavelength, λ. The dominant response from a dipole sheet at a point leads to an overall phase retardation and our acceptance of refractive indices greater than unity. Exploiting a more complex individual response without large-scale averaging can result in phase advances, but this has proved easier said than done in practice. The major difference between atoms in a real material and meta-atoms in a metamaterial is that in the former there are clearly many more atoms per unit of wavelength or per interaction volume than there can be or needs to be for a practical metamaterial. Historically, for real materials, an averaged response to an electromagnetic field has proved to be a reasonable assumption to make. This is referred to as the random phase

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approximation (RPA), which is credited with giving meaning to constitutive parameters such a material's permittivity or permeability [1]. This approximation is not necessary or necessarily appropriate for metamaterials.

Coupling effects between neighboring elements in a metamaterial are the key to unusual bulk metamaterial properties, but they are also well known to be difficult to model, being a many-body problem and making homogenization estimation of constitutive parameters difficult. Employing more densely packed subwavelength-scale meta-atoms is attractive for engineering larger electromagnetic responses, but this benefit will diminish if it is associated with increasing relative fabrication errors in the meta-atoms' critical dimensions, making the RPA reasonable once more.

Let us consider an electromagnetic wave interacting with any confined system containing charges. In treating an interface between two piecewise analytic regions, we are required to match wave functions and their derivatives at the interface, thereby creating reflections and leading to confinement. We are accustomed to doing this at the macroscopic level when dealing with many atoms, but the concept remains valid down to the realm of a single atom. This intrinsic boundary between regions provides trapping with high Q [2] in atoms, which we note is also entirely consistent with the properties of very small ($\ll\lambda$) antennas [3]. Moreover, while we stress that waves, unlike particles, do not have potentials, this in no way precludes an incident electromagnetic plane wave from being strongly localized. This is appreciated if one considers a representation of a plane wave in terms of spherical Bessel functions of the second kind. Coupling between the interaction volume and these modes of the incident wave is inevitable. In what follows, we shall use the results derived from a simple Lorentz oscillator model as a basis for how to develop boundary conditions and describe the light-system interaction. We recall the prediction of electronic band structures, in which we start with the atomic wave functions [4]. The Wigner–Seitz cell connects the points in the reciprocal space to generate a boundary surface for dividing the space into analytic regions for boundary conditions. Traditionally, trapping occurs within a volume of interaction such as a Fabry–Perot resonator for plane waves. If we consider a single atom (or meta-atom), a spherical volume with some radius defines the volume of interaction. Thus our spherical interaction volume or cavity similarly serves to define boundary conditions.

2. ELEMENTARY LIGHT–MATTER INTERACTIONS

Let us first consider a single atom. Because light is a wave and an atom is a particle, it has proven convenient to represent light as photons because the exchange of energy and momentum is then treated in quantum mechanics with units of $\hbar\omega$ and $\hbar k$. In reality atoms and electrons are spatially localized by their interaction potentials, which have singularities, but, without interaction, light as a wave cannot be localized. No position operator exists for a photon, and the most precise localization appears to be in the form of wavefronts [1], while interaction cross sections loosely relate to wavelength. There is no need to assume that a field of quantized photons exists *a priori* in the absence of matter or in the absence of an interaction in general. To explain observed light–matter phenomena, $E = \hbar\omega$ is better regarded as a property of matter rather than of light. Put differently, electromagnetic waves do not interfere with each other in the absence of entities with mass. *It therefore follows that a quantized state for light is not necessarily an objective property of light, but rather a statement about our knowledge of how light interacts with matter.*

Adopting this somewhat less fashionable view provides some important insights. The observed exchanges of electromagnetic energy between particles (atoms) appear to be in discrete units or quanta, but this is because of the nature of wave–particle interactions. Well away from resonance, there is still interaction, but it is weak, while closer to resonance the interaction is strong. Trapping and storing of light in a subwavelength-sized resonant volume is interpreted to be in the form of nonpropagating or evanescent waves resulting from resonant interactions, behaving electrostatically. Also important is the fact that these interactions take a relatively long time to occur, as the energy associated with the incident periodic electromagnetic field couples more or less strongly with the particle, depending on its frequency. Atomic transitions and the photoelectric effect are observed to have response times that are relatively long. They range from a few nanoseconds to much longer for a Rydberg atom, for example. Excitation of an electron to a higher-energy eigenstate takes time and many cycles of the incident electromagnetic wave. Appreciating the dynamics and physical limitations of this process is especially important for meta-atoms and the design of metamaterials and determining whether there is a net coherent or a net averaged response from an assembly of these elements.

We therefore draw the very close parallel between the electromagnetic response of real atoms and meta-atoms and can proceed to do so at all size scales without the need to invoke the concept of a photon. Individual unit responses result from either opportunistic or deliberate excitation by an electromagnetic field, which may or may not be close to a (cavity) resonance. Coupling between different resonant states is easier to understand when a sufficiently broadband electromagnetic field is present. This is not necessary, however, since losses and nonlinearities play a role. For an atomic system, putting a non-Hermitian term in the Schrödinger equation represents damping and or nonlinearities. If losses are high, then it is pointless to describe resonant states and propagation constants, but not for moderate values of Q. With a non-Hermitian operator, we give up eigenvalues' being constant for all time, and wave functions belong to two or more different states, thereby providing a coupling mechanism.

As electromagnetic energy is transferred from the field to the charges, our concept is that of a light-dipole or light-multipole quasi-particle, which is assumed to be uncoupled from other light-dipole quasi-particles [5], but this is not necessarily the case. Thus the overall interaction is governed by the volume density of the dipoles (or multipoles) present, but are the dynamics suggesting a correlated or uncorrelated response? There is an equivalence of events in a single cycle over a large volume and events in a very small volume but spread over a long period of time. Therefore, the main contribution to the electromagnetic response of bulk materials and metamaterials is determined by the volume density of interacting particles. We argue here that this concept of the volume density of interacting particles is applicable down

to a single atom or meta-atom in its interaction volume and that for smaller clusters of entities we can replace the RPA by statistics.

3. EXCITATION OF AN ATOM BY AN INCIDENT ELECTROMAGNETIC WAVE

Trapping of light in a cavity such as a Fabry–Perot resonator results in static oscillations between the cavity walls. In a spherical cavity, the trapping between $r = 0$, the origin, and $r = a$ defines the resonator. The cavity may be represented by an infinity at $r = 0$, requiring the wave to be zero there to avoid a singularity. For a single atom, trapping can be represented by a spherical cavity between $r = 0$ and some $r = a_0$. We can decompose an incident plane wave into a weighted sum of spherical harmonic functions; i.e., we can write a linearly polarized component of the electric field as

$$E(x) = E_0 \exp(ikx) = \sum_{m=-\infty}^{m=\infty} i^m J_m(kr) \exp(im\varphi). \quad (1)$$

These spherical harmonics carry angular momentum, and the higher-order m modes have a radial component of the field and carry exponentially less energy at the point of expansion for increasing m. Thus the interaction with the H atom is primarily with the first- or second-order modes, the first-order mode having spherical symmetry, as shown in Fig. 1(a), while the $m = 4$ and 12 modes are shown in Figs. 1(b) and 1(c).

Exponential decay of high-m modes at the center is the result of conservation of angular momentum, where $m = k_\theta r$, and k_θ is the tangential component of the wave vector and r is the distance from the origin or location of the atom. Thus we can write that $k_\theta \propto 1/r$. Given the dispersion relation

$$k_\theta^2 + k_r^2 = \varepsilon \frac{\omega^2}{c^2}, \quad (2)$$

then when the tangential wave vector increases toward $r = 0$, it follows that k_r has to eventually vanish. This defines a boundary for the interaction, as can be more clearly seen for larger m as in Fig. 1(c). Beyond this radius the incident wave will be reflected back, but inside this radius, the angular momentum states are evanescent. The point at which $k_r \sim 0$ occurs at a radius $r \sim m$, using $m = k_\theta r$. One can interpret each harmonic function associated with the incident wave as efficiently interacting with electron wave functions that have similar symmetries and hence maximum overlap. The $m = 2$ spherical harmonic maps well onto a p orbital.

4. LORENTZ FORCE

The electromagnetic response of an atom or meta-atom is thus a function of the effective boundary conditions defining the "box" in which free or bound electrons are confined. The motion of these electrons, both near and far from resonance, can be described more carefully than by simply adopting a Drude-like model [6]. The Lorentz force on the electrons associated with the illumination cannot be ignored and alters their quiescent states. The model can change from

$$d\mathbf{v}/dt + \gamma \mathbf{v} = -\frac{e}{m}\mathbf{E} \quad (3)$$

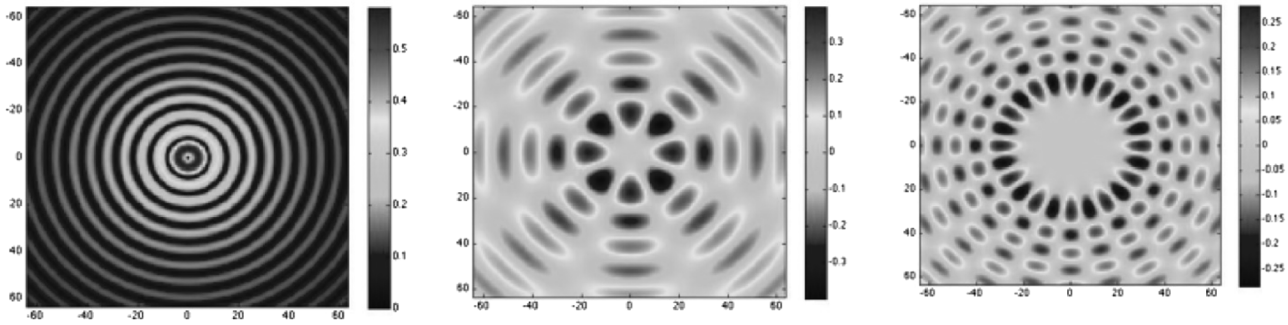
to

$$d\mathbf{v}/dt + (\nabla \cdot \nabla)\mathbf{v} + \gamma \mathbf{v} = -\frac{e}{m}(\mathbf{E} + \mathbf{v} \times \mathbf{B}) - \beta^2 \nabla \cdot n, \quad (4)$$

leading to $\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}_f + \mathbf{P}_c$, denoting contributions to the polarization from both free and core electrons. We assume that electrons are confined in a cavity or potential well, as described in the previous section. In more complex systems, such as atoms, there will be deviations from the Coulomb law we describe by shielding, which neglects the possibility of energy transfer between atomic electrons. There is an uncertainty associated with the time of transfer of energy in these systems, especially if a loss mechanism is present. This is another argument why one should be cautious about the use of the word "photon," since this implies a more definite type of particle nature than is really possessed by electromagnetic energy [7]. We also note that increased confinement in physically smaller structures restricts motion, which further necessitates that the role of electron–electron interactions be included.

5. BOUNDARY CONDITIONS

Strictly speaking, particles all have potentials having an r^{-1} singularity, but waves do not. In treating an interface between two piecewise analytic regions, we match wave functions and their derivatives at the interface, leading to reflections and confinement. We are accustomed to doing this at the macroscopic level when dealing with many atoms, but we argue here that the concept remains valid down to the realm of a single atom. Not all boundaries are smooth, and specifying boundary



(a) (b) (c)

Fig. 1. (a) $m = 1$ spherical harmonic, (b) $m = 4$, and (c) $m = 12$.

conditions without assuming RPA, for example between two media with different periodicities, requires a scattering matrix treatment, (e.g., [8]). Nevertheless, an effective boundary between regions can provide trapping with high Q [2], which we will see is consistent with the extensive studies of very small ($\ll\lambda$) antennas [3], thereby suggesting a natural and instructive Maxwellian equivalence between the two. Nothing precludes an incident electromagnetic plane wave from being strongly localized, and coupling between the interaction volume and these modes of the incident wave is inevitable.

In what follows, we use the results derived from a simple Lorentz oscillator model as a basis for how to develop boundary conditions. We need to consider how to specify boundary conditions between separated regions without using the classifications defined by constitutive equations, such as dielectric functions ε_1 and ε_2. The latter are more easily definable as averaged quantities. One is used to finding solutions to differential equations in piecewise analytic regions where waves and their derivatives are matched over the dividing lines separating these two regions. This traditional approach is applied in classical physics as well as quantum mechanics. However, at scales small on the dimensions of a wavelength, boundaries can occur between a local homogeneous medium and a potentially nonlocal metamaterial or photonic crystal [8,9].

6. CLASSICAL VERSUS QUANTIZED FIELD

A physical picture in terms of a modal decomposition or representation was given of the coupling between an incident plane wave and the permitted electron wave functions in confined volumes, such as an atom, structured potential well, conducting nanostrip, or meta-atom. This provides insight as to why some minimum interaction time is inevitable in order to drive those electrons into a response, possibly a resonant response leading to transformation to a higher energy eigenstate. It is well known that the model of a classical electromagnetic field is satisfactory in order to describe such interactions. Electromagnetic field quantization is only required [10] for Planck's law, the Compton effect, spontaneous emission, and electrodynamic level shifts, and all of these can be subsumed into a loss term added to the Green function [4]. However, Hong *et al.* [11] have demonstrated that an emitted wave-packet has energy $h\nu$ and occupies a finite extent in space and time. Thus interactions of an electromagnetic field with electrons in matter lead to observations that can be interpreted as particle-like simply because of these localized effects that take place over finite interaction times.

The emission of radiation from an atom (very much smaller than the wavelength) or meta-atom (i.e., a structure somewhat smaller than the wavelength) can be compared to that of the excitation of and reradiation from an antenna. In the near field, an electrically small antenna, such as a dipole has an extraordinarily high Q, and extremely large field enhancements (i.e., stored electromagnetic energy) are possible. The electromagnetic field is derived from a vector potential, and, under these extreme conditions, the wave vectors of the fields are nonlinear (Bloembergen [12]). This is evident from a simple logical argument based on transition times between eigenstates through to ionization, as a function of incident field amplitudes versus frequencies.

7. RESONANCE

We start with the classical Lorentz model,

$$\ddot{x} + \gamma\dot{x} + \omega_0^2 x = -eE/m \tag{5}$$

with the dipole moment

$$P = -Nex = (Ne^2/m\epsilon_0)E/[\omega_0^2 - \omega^2 - i\gamma\omega]. \tag{6}$$

It is important to repeat that our main point is to replace N by a single dipole per unit volume of $4\pi a^3/3$, with a being the radius to be determined where a response is large. The dielectric function is

$$\epsilon(\omega) = 1 + (\omega_{es}^2)/[\omega_0^2 - \omega^2 - i\gamma\omega]. \tag{7}$$

Here, ω_{es} is usually expressed in terms of the plasma frequency ω_p, with the number density determined, for example, by the density of electrons in a medium, which is given in terms of the number of atoms per unit volume in a typical solid. This description holds well for atoms or meta-atoms. An incident field of the right frequency will excite the electron(s)–nucleus system in an atom. These charged elements represent a plasma-medium in a cavity or meta-atom structure, just as a single atom (e.g., a dipole) interacting with an electromagnetic wave defines a volume of interaction.

The Q for a resonant system in most cases can be expressed by $i\omega\tau$, where τ is the lifetime. We note that the engineering definition of Q, namely $2\pi\times$ energy stored per unit volume/energy lost per cycle is only meaningful for $Q > 10$ [2]. Although this latter definition is not as widely applicable, the definition in terms of energy stored and energy lost per cycle is very useful, since it provides an estimate of the energy stored once the Q and loss per cycle are known. The energy stored can be expected to induce changes in the cavity characteristics that we label as nonlinear phenomena.

8. HYDROGEN ATOM

Consider the very special case of the $1s - 2p$ transition of a H atom. This transition involves $\omega = 1.55 \times 10^{16}$ s^{-1} and the lifetime $\tau = 1.6 \times 10^{-9}$ s, giving $Q = 2.48 \times 10^7$. In other words, it takes 25 million cycles to build up the spherical interaction volume's resonance. The remarkable feature of a single atom interacting with even very low intensities of background radiation is that it describes a resonating system with such a high Q. We argue that this provides insights in devising high-to-low-Q switchable states for systems in metamaterial devices. Switching phenomena arising from stored energy inducing an input–output hysteresis is well documented through dielectric confinement and nonlinear responses [13] and phase change mechanisms leading to Rabi-like oscillations of conductance in quantum dots [4].

In Eq. (7), we equate $\omega_{es}^2 = \omega_0^2$, with ω_0 being the transition frequency of the $1s - 2p$ levels of the H atom. Intuitively, to achieve resonance, we need to set the frequency of the incident light to be equal to the $1s - 2p$ transition frequency [14]. This is an important point in our discussion here. From Eq. (7) we obtain the real and imaginary parts of the dielectric function $\epsilon_1(\omega)$ and $\epsilon_2(\omega)$ shown below:

$$\epsilon_1(\omega) = 1 + (\omega_{es}^2)\{(\omega_0^2 - \omega^2)/[(\omega_0^2 - \omega^2)^2 + (\gamma\omega)^2]\}, \quad (8a)$$

$$\epsilon_2(\omega) = (\omega_{es}^2)\{\gamma\omega/[(\omega_0^2 - \omega^2)^2 + (\gamma\omega)^2]\}. \quad (8b)$$

At $\omega = 0$, $\epsilon_1(0) = 1 + \omega_{es}^2/\omega_0^2$, and since this is for resonance, we set $\omega_{es} = \omega_0 = \omega$, then $\epsilon_1(0) = 2$, reaching a value of $\epsilon_1(\omega_0 - \gamma/2) = 1 + \omega_{es}^2 Q/2\omega_0^2 = 1 + Q/2$; and $\epsilon_1(\omega_0) = 1$; $\epsilon_1(\omega_0 + \gamma/2) = 1 - Q/2$ and at $\omega = \infty$, $\epsilon_1 = 1$. For $\epsilon_2(\omega)$, this function starts at 0, rises to $\pm Q/2$ at $\omega_0 - \gamma/2$, and $\omega_0 + \gamma/2$ with a value of Q at resonance, $\omega = \omega_0$, falling to 0 at $\omega = \infty$.

The Q factor can be further maximized by allowing a variation of ω_{es} by adjusting the size of the interaction volume. We can expect some detuning of ω_{es} in practice due to the sustained interaction of the incident electromagnetic field with the charge(s) in the H-atomic system. The electromagnetic forces acting on the electron(s)–nucleus system are well understood from Maxwell's equations, there being components depending on the electric and magnetic multipole moments, the electric and magnetic fields, and their gradients. The motion of the bound electrons in their potential well define a cavity whose structural complexity increases with additional electrons and higher-frequency incident electromagnetic waves.

Once the value of the electric field of spontaneous emission is determined, the Q factor allows us to determine the value of the electric field after buildup at steady state. The electric field strength due to spontaneous emission has been well treated by Fitzpatrick [15], and what follows represents a few steps leading to the final conditions resulting from an initial electric field $\mathbf{E}_i$. Following Fitzpatrick, the value of γ in Eqs. (8a) and (8b) may be written as

$$\begin{aligned}\omega_{2p-1s}{}^{\text{spon}} &= \omega_{2p-1s}{}^3 d_{2p-1s}{}^2/3\pi\epsilon_0\hbar c^3 = \gamma \\ &= (2/3)^8\alpha^5(mc^2/\hbar) = 6.3\times 10^8 \text{ s}^{-1}\end{aligned} \quad (9)$$

with $\alpha = 1/137$ being the fine structure constant in γ, and the inverse of γ is the lifetime $\tau = 1.6$ ns, which is used for the trapping time. Fitzpatrick listed all the dipole matrix elements, repeated below for convenience:

$$\langle 100|x|2,1,\pm 1\rangle = \pm(2^7/3^5)a_0, \quad (10a)$$

$$\langle 100|y|2,1,\pm 1\rangle = i(2^7/3^5)a_0, \quad (10b)$$

$$\langle 100|z|2,1,0\rangle = \sqrt{2}(2^7/3^5)a_0, \quad (10c)$$

with the dipole moment $d^2 = |(10a)^2 + (10b)^2 + (10c)^2|e^2 = (2^{15}/3^{10})e^2a_0^2$, then $d = 0.632\times 10^{-29}$, and the electric field $E = \hbar\gamma/d = 1.05\times 10^4$ V/m.

Therefore, after Q factor buildup, the electric field for the dipole transition between $1s$ and $2p$ is $E_{1s-2p} = \sqrt{Q}E = 5.15\times 10^7$ V/m. This value remains significantly lower than the static electric field between the nucleus and electron inside the atom and supports perturbation-based methods being used to describe these quantum mechanical transitions; the Rydberg field is 5.14×10^{11} V/m.

9. TRIPLE RESONANCE

Traditionally we describe the plasmonic interaction of oscillating electrons with light in terms of summing the individual dipole moment per unit volume excited by an incident plane wave. The interaction volume is generally much greater than the separation of the oscillating dipoles, which, having sufficient collisions, allows us to apply the RPA to the sum, giving an average dipole moment per unit volume, excited by the incident wave. For a finite interaction volume smaller than the wavelength, one can better envisage the incident wave as represented by one or more of the spherical harmonics into which a plane wave can be expanded. We recognize that the plasma frequency can be at resonance with the frequency of the incident wave, but that in any physical situation at a nonzero (Kelvin) temperature, we should include the background radiation in our defined space of interaction. Also, as the size of the interaction volume is reduced, we reduce the number of dipoles, but as long as the average leading to the RPA is still operative, the resonance remains unchanged. However, when we are left with very few, or a single dipole, then we would take the single dipole per unit volume to describe the plasma frequency. The interaction volume in this case is determined by a sphere with an effective radius given by the relationship of the spherical harmonics of the incident wave to the dynamic response of the oscillating electron (if we assume ionization does not occur). We note the need to account for the nonzero background. This is particularly important when the excitation is very weak and comparable to the background when there is dissipation. The unaccounted interaction with the background is a factor that determines the Q, or the linewidth of this intrinsic broadening. Thus the behavior of a dipole antenna is identical to our model for the interaction of a single atom with light, once one includes the background! In other words, for the H atom, if the incident electromagnetic radiation is much stronger than the background, this incident wave would instantly drive the electron into $1s - 2p$ transition, and the interacting system would not have to wait 1.6 ns to start the Rabi oscillation. The Rabi frequency is proportional to the strength of the electromagnetic wave.

The term "triple resonance" refers to the fact that the frequency of transition $1s - 2p$ is the same as that of the incident light as well as being the same as the plasma frequency used in the Lorentz model. While the Lorentz model is classical in its origins, we point out that in any Green functions for waves, including Schrödinger equations or those for harmonic oscillators, the form is identical to the Lorentz model.

At the resonant frequency, multiple reflections in the cavity eventually lead to zero reflection and unity transmission. This matching condition is equivalent to a saturation of absorption by the atom, and dispersion relations dictate that the sharp index changes close to the resonant frequency disappear. After the 1.6 ns buildup time, when the atom becomes transparent, either the persistence of the incident field or some other perturbation induces the release of the stored energy. In the initial absence of the atom-cavity's high Q, the emission time can be much faster than the buildup time.

The resonant frequencies of a real atom correspond to the differences between energy eigenstates, but consider a meta-atom. The electromagnetic response of an isolated meta-atom can exhibit multiple resonant frequencies determined by its material properties and shape. Coupling and transitions,

including Rabi-like oscillations, between these states can occur in just the same way. Induced current densities are necessarily finite and can saturate, and the displacement field may not follow the incident field, leading to perceived nonlinearities.

We ask how we can make a better source of electromagnetic waves, such as a laser or another useful device based on meta-atoms and the relevance of the antenna model. If we take a single antenna or a single atom, our approach leads to a similar result. Should we have two or more dipoles, we would need to consider cross terms, and we should use the dyadic Green function rather than the RPA.

10. SMALL ANTENNAS

The mechanism responsible for the Q of small antenna is similar to that determining the Q of the interaction of low-intensity light with a single atom. This involves a triple resonance, in other words, the frequency of the dipole transition at resonance with the effective dipole resonance, which is defined by the interaction volume as well as the frequency of the background radiation. We have shown that our so-called triple resonance involves the basic assumption of assigning a value for N in the usual expression for the plasma frequency ω_p, by considering a single dipole per unit volume, and taking that interaction volume to be that of a sphere whose radius is adjusted to equate the frequency ω_{es} to be equal to both ω_0 and the frequency of the incident light ω. Mathematically this procedure produces the maximum trapping, or in other words the largest Q factor, because the imaginary part of the dielectric function has a maximum value equal to Q at $\omega = \omega_0$, etc. We consider here how our results compare with the Q-factor used in ultrasmall antennas. Much has been written on the fundamental limits on Q for an antenna. While some refinements have been made over the years, there is broad agreement that for very relatively small antennas, the maximum Q is well approximated by $Q_{\mathrm{Chu}} = (1/ka)^{-3}$, where the subscript Chu is in recognition of the first definitive paper on this topic [16,17]. For the H atom transition from $1s$ to $2p$, the transition frequency is $2\pi \times 0.24 \times 10^{16}$ Hz, which corresponds to a wavelength of 124 nm. If we adopt the simple Lorentz dispersive model of Eq. (7), and assume that ω_{es} is the plasma frequency, $Ne^2/\varepsilon_0 m$. We assume that for an H atom, N, the number density of dipoles, is simply one per unit volume and the volume is $4/3\pi a^3$. Equating the plasma frequency to the transition frequency, the value for the radius of the interacting sphere is $a = 0.6$ Å. Using this and knowing that $k = 2\pi/1.215 \times 10^{-5} = 3.1 \times 10^{-3}$ cm^{-1}, we obtain $Q_{\mathrm{Chu}} = 3.4 \times 10^7$. Interestingly, our $Q/Q_{\mathrm{Chu}} = 0.73$. Considering the use of widely different concepts for arriving at the value for the Q factor, having only a 30% difference is a remarkable consequence.

Thus, in summary, using the H atom as an example, we know that the lifetime (1.6 ns) of the excitation of $1s - 2p$ corresponds to a certain $Q(= 2.5 \times 10^7)$ and that for a small antenna (Chu, [16]) that same Q corresponds to an antenna circumscribed by a sphere of radius 0.6 Å, almost exactly the (Bohr) radius of the H atom. We explain this in more detail in what follows. The H atom is conceptually identical to a small antenna driven by a large current into large oscillation almost instantly. Since the ratio of the Q calculated for the single atom of the H atom is 2.48×10^7, while the Q_{chu} is 3.4×10^7, a factor of 1.36 larger, this may be accountable by considering the difference in the intensity of the background frequency of the small antenna, compared to that of the H atom, indicating that perhaps the background for microwave frequency is 1.36 times below that of the H atom being excited in the UV. The difference of 1.36 is in the right direction but hardly sufficient from blackbody background. Perhaps something unknown to us is involved.

11. ENERGY TRANSFER AND STORAGE

Energy transfer into this resonant system is governed by the Poynting vector. The rather complicated description of momentum transfer requires that one choose a suitable basis such as the spherical Bessel functions of the second kind, as previously mentioned, to represent the incident wave. It is well known that the effective radius of this cavity scales as n^2, the scattering cross section as n^4, and the system's polarizability as n^7, where n here is the principle quantum number associated with the energy eigenstate involved. These resonances are excited as a result of a finite energy transfer into the atom's plasma medium following some number (millions) of cycles of the incident field. While being excited, energy is being given up either by inelastic scattering or nonradiatively. Energy is also given up radiatively, in units reflecting the energy differences between the energy eigenstates and coupling, and induced perturbations contribute to the instability of each eigenstate. Some are relatively long lived, such as the H $2s$ state, but the fact that emission occurs from all eigenstates, broadly subject to selection rules, can be explained by the highly dynamic electromagnetic environment of a real atom in which all electromagnetic fields interact to some extent, however weakly. In other words, electromagnetic energy permeates an atom from its neighbors and other fields.

The Kramers–Kronig relations define refractive index walls around a resonant frequency, increasingly sharper the higher the Q of the system. Being a direct consequence of causality, these relations hold for dynamic and nonlinear behavior, although full use of these integral transforms might be difficult to estimate over small bandwidths and near resonances. These refractive index barriers contribute to the cavity until the absorption saturates when there is sufficient energy stored to reach a new eigenstate, at which moment they disappear and the electromagnetic energy is no longer optically trapped or confined. These interfaces require many cycles of the incident wave to build up the energy density in the interior, which is ultimately limited by the loss factor. In addition, we note that the Kramers–Kronig dispersion relationships assume analyticity, which implicitly requires that all spectral functions are defined on a continuous variable.

We must realize the meaning of such high Q with reference to response and sensitivity. Detectors need high sensitivity, favoring a high-Q system. However, in electronic applications, a Q over one million is too slow to be of any use at all. An atomic system is slow compared to a solid-state system, i.e., slow with respect to processes not under our control, such as interactions with impurities. One adopts high-Q systems in parallel to increase response time, and such systems are very useful for sensing and parametric systems.

We have described how bound electrons in an atom represent a spatially confined plasma that interacts with all electromagnetic fields in its vicinity, but strongly so when the incident frequency is close to a resonance. At resonance

the phase relationship is locked. This is equivalent to exciting a resonant cavity with highly reflecting boundaries but applies equally to the example of a single atom. Light is slowed down by the high Q of the atomic cavity as energy is stored. The Q, and hence the stored energy, as for a dipole antenna, increases dramatically as the physical size of the antenna decreases. It is therefore no surprise that it takes time to store energy in an atom or meta-atom cavities. The excited electron is thought of as a quasi-particle that is transformed between energy eigenstates over time, and the spatial distribution of energy is well modeled by a quasi-electromagnetic cavity mode that corresponds to the quantum mechanical construct of a wave function.

According to Lamb and Scully [10], energy density can grow, leading to electron emission in a solid (photoelectric emission) because of the total stored energy. Similarly, for any simple resonator, such as an atom, a combination of stored energy and applied (Lorentz) force can result in ionization. In other words, the 10^6 field oscillations required to excite the H atom could be experienced serially or in parallel. Thus, as pointed out in Section 9, excitation to any excited state (or ionization) simply requires sufficient total energy in the resonator, since mixing of modes or eigenstates occurs immediately and the probability of a transition is finite, assuming the appropriate momentum conservation and a nonperturbative solution.

Energy density and the magnitude of the (distorting) Lorentz force increase with increasing frequency. In a finite potential well in which carriers are confined (either H atom or meta-atom), their motion becomes nonlinear at increasingly high energy densities, leading either to a transition to a new eigenstate if one exists or to ionization.

A change in eigenstate leads to an abrupt change in the system's polarizability and hence interaction with the surrounding field. A consequence of this is emission (stimulated) and Rabi oscillations, whose frequencies also increase with increasing electromagnetic wave frequency:

$$\mathbf{E}(\mathbf{r},\omega) = \mathbf{E}_0(\mathbf{r},\omega) + i\omega\mu_0\mu_r(\omega)\int_V \overleftrightarrow{\mathbf{G}}(\mathbf{r},\mathbf{r}',\omega)\mathbf{j}_s(\mathbf{r}')\mathrm{d}^3r', \quad (11)$$

and for the dipole

$$\mathbf{j}_s(\mathbf{r}) = i\omega\mathbf{q}\delta(\mathbf{r}-\mathbf{r}'). \quad (12)$$

This further reinforces the direct analogy between the Q of an atom and that for a dipole or multipole based on the Chu limit. We can generalize this to more complex resonator shapes, such as heavy atoms and meta-atoms, especially in the near field. Can we use our description for the interaction of light with a H-atom as an initial model for designing a better meta-atom for metamaterials? In an exactly analogous fashion, the excitation and relaxation time of man-made resonators will be finite and can be characterized in terms of the effective L, C, R parameters. These man-made structures can have very high Qs and can be arranged in proximity to each other with relative precision, allowing responses governed by coherent phase-sensitive coupled interactions between them, rather than an averaged responses described by a RPA.

We have found that the H atom's transition from $1s$ to $2p$ involves the maximum interaction with incident light when the dipole moment has an oscillation frequency equal to the frequency of the incident light and a volume with a radius ~0.6 Å. For other transitions, this volume will change appropriately due to the Lorentz force associated with the incident electromagnetic wave. We explain and explore these concepts below.

12. ATOMS AND META-ATOMS AS ANTENNAS AND CAVITIES

A single atom interacts with light having wavelengths many orders of magnitude larger than the range of interaction. We have argued that the field enhancement, in terms of Q, is extremely large and of the order of tens of millions. As shown above, a plane wave having $\lambda \gg a$, can be expanded in a series of spherical harmonic functions and a represents a radius of significant interaction with those basis functions that turns out to be close to the Bohr radius of the atom. Light in the form of a plane wave has to be expanded in terms of Bessel functions of the first and second kind, to allow us to describe the interaction with a multipole having a singularity $\sim 1/r$, as with most potentials. As we have seen, the Q of a correspondingly very small antenna is within 20%–30% of this high Q for atom–light interaction. It is therefore very likely that there is a common underlying cause.

From a classical cavity perspective, one can interpret the transparency post-transition as the interference of the stored with the incident light, resulting in cancellation of the field at the interface specified by a. An ideal cavity is almost reflectionless for any high-Q matching network, such as a Fabry–Perot interferometer, or simply a parallel plate resonator. The atom-cavity is defined by a physical volume that is determined by the range of motion of the electron as it gains energy from the incident field. One can envisage this as a somewhat stable quasi-particle, which should include nonlinear interactions, in principle. As is well known from a purely electromagnetic perspective, certain motions (see [18]) of the electron, despite its acceleration, will not radiate. Schott considered the special case of a uniformly charged sphere of radius a and demonstrated that this sphere will produce no radiation for a radius $a = mcT/2$, where m is an integer. The period of the oscillation, T, is an integer multiple of the amount of time it takes for light to cross the sphere's diameter. This indicates that the sphere's range of motion is limited by relativity to be much less than its diameter or, in other words, that the sphere's motion is more of a wobble than an orbit (see [19]).

These nonradiating states are the bound states of the atom, which quantum mechanics demonstrates are eigenstates and perfectly stable should no ongoing excitation occur to perturb the cavity volume and/or the energy of that otherwise stable state. The volume is dictated by the efficiency of energy transfer from the incident wave. To a good approximation, given the relative scale of the wavelength to the atom's size, the incident wave is well approximated by a plane wave. Consequently only a portion of the incident electromagnetic field, as represented by the first one or two terms of its spherical harmonic expansion, will strongly couple with the H atom. It is therefore not surprising that many oscillations of the field are necessary to result in a transition. If the plane wave is viewed as a mode that is heavily populated with photons, i.e., has high power, then the excitation time could be reduced. The characteristic response time of ~1.6 ns for a H atom to transition

from $1s$ to $2p$ assumes low power, and the probability of a single photon with exactly the correct energy corresponding to the transition's energy difference effecting that transition is extremely low. The amplitude of the harmonic shown in Fig. 1(a) will dictate the transition rate and Rabi frequency. The volume of the cavity is determined by the radius at which higher angular momentum expansion functions representing the plane wave have a k_r that goes to zero. The point at which $k_r \sim 0$ occurs at a radius $r \sim m$, using $m = k_\theta r$.

13. METAMATERIALS

Almost all metamaterials consist of large numbers of repeated meta-atoms forming a lattice similar to a superlattice or photonic crystal, but with a repeat that is much greater than the atomic dimensions while simultaneously being much less than λ. Systems with repeated subwavelength scale structures that are loosely interacting resemble adding many small antennas [20]. Engineering such media to increase the interactions between elements increases the metamaterial's electromagnetic response, while simultaneously diminishing the role automatically assumed to be at play of the RPA. There is an opportunity to make a fast low-Q system or one that has a very narrow bandwidth and the high gain of a high-Q system. One can consider systems having nonlinearly interacting components that minimize the problem of nearest neighbors' losing their relative phase coherence. Such an approach calls for interactions where parametric interactions can take place, for example, by adding an idler frequency. This is tantamount to mimicking a triple resonance.

The simplest meta-atom in a metamaterial is multipole-like (split-ring resonators, S structures, etc). We think of these as resonant *LCR* circuits abstracted into a material shape. Excitation and emission (that is, radiating) is equivalent to the response of small antennas, and no RPA is necessary. Even in traditional lasers, the response is described in terms of rates because of the RPA. If we consider a mode-locked laser, mode locking is achieved by insertion of a switch or passive saturable absorber inside the cavity. When that is the case, we are essentially adding another interaction that we can control. We tend to think of a meta-atom as a resonant structure but not as a resonant cavity. However, if we make metamaterials from meta-atoms comprising natural materials in which absorption can be saturated or in which phase changes can occur (e.g., [4]), then a meta-atom can be both a resonant structure and a resonant cavity and behave like a real atom. Making a material from two or more such meta-atoms might allow us to make an artificial lasing source or parametric amplifier in which the RPA is not a constraint and rate equations are replaced by a more powerful phased array concept.

We cite, as an example, the success of the semiconductor superlattice. This is an active element and involves an applied dc as well as ac field, resulting in gain for amplifiers and oscillators [4]. It would seem only reasonably to expect that metamaterials could and should also be active and be capable of having a voltage applied. Several papers demonstrating tunable and nonlinear metamaterials have been published [e.g., 21]. However, the performance of engineered structures such as a conventional superlattice, which are ostensibly periodic structures, is dramatically compromised with even very small degrees of spatial or material disorder, such as defects (see, for example, [22]).

This observation makes it necessary to turn one's attention to components and devices that have relatively small numbers of well-defined periods without losing those desired coherent effects. An example is the coherent superposition of meta-atom resonant responses that are exploited for large field enhancements or large changes in permittivity or permeability. We have observed that increasing the number of meta-atoms in order to increase a metamaterial's volume can be counterproductive. Random processes associated with fabrication ultimately lead to an averaging of desirable properties, which one can attribute to the central limit theorem or, in condensed matter physics, attribute to the RPA. It is worth drawing lessons learned from technologically sound advances that were made with a finite array, such as the chirp radar and the Yagi antenna. Neither are periodic, allowing some very unique features to be incorporated into their functions. An analogy for our purposes would be a practical superlattice comprised of just a few periods or a small cluster of meta-atoms. This suggests that a meta-molecule may be more practical and effective as a localized or lumped element than would be a larger structure with sensitive delocalized properties. The Yagi antenna, by virtue of its outer elements, allows one to design a beam and gain profile having unique features. The chirp radar allows pulse compression and has $1/\Delta$ periods, which are far from being equal. Even the quantum cascade laser is basically a generalization of a superlattice in the category of metamaterials.

14. MAKING NANOSCALE META-ATOMS

The model for free electrons inside metallic structures fails for metals whose critical dimensions are of the order of a few nanometers. A modified description should account for atomic and subatomic interactions, as well as electron–electron repulsion. The Pauli exclusion states that two electrons cannot occupy the same state at the same time. This can be represented as a repulsive force between charge carriers. In addition to the classical Coulomb force, this quantum repulsion results in a pressure in an electron gas in response to an electromagnetic field. This electron pressure is taken into account by a hydrodynamic description of the collective motion of the electrons inside a metal [23]. The currents $\mathbf{J}$ inside a metal induced by an electric field $\mathbf{E}$ oscillating at frequency ω can be described by

$$\beta^2 \nabla(\nabla \cdot \mathbf{J}) + (\omega^2 + i\gamma\omega)\mathbf{J} = i\omega\omega_p^2 \varepsilon_0 \mathbf{E}, \tag{13}$$

where ε_0 is the vacuum permittivity, and γ and ω_p are the damping coefficient and the plasma frequency, respectively, which also appear in the usual Drude formula,

$$\varepsilon(\omega) = 1 - [\omega_p^2/(\omega^2 + i\gamma\omega)], \tag{14}$$

and β, which is approximately the speed of sound in the Fermi-degenerate plasma of conduction electrons, is proportional to the Fermi velocity v_F.

The effect of including the pressure term in the electron response is that the longitudinal dielectric function, ε_L, becomes nonlocal, depending on the propagation vector $\mathbf{k}$ in addition to the frequency, as follows:

$$\varepsilon_L(\mathbf{k},\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\gamma\omega - \beta^2|\mathbf{k}|^2}, \tag{15}$$

whereas the transverse response is unchanged (see [24]). The simple picture of a surface charge layer with infinitesimal extent is replaced with a continuous charge density, whose extent will be determined by $\beta/\omega_p \sim \lambda_{\mathrm{TF}} = v_F/\omega_p$, is the Thomas–Fermi screening length. Rather than a strict surface charge density, the nonlocality produces a volume charge density that spreads out from the surface a distance $\sim\lambda_{\mathrm{TF}}$, which is of the order of 1 Å.

Either using the Drude model or using Lorentz expressions for the plasmonic dielectric function is same as using the RPA. Therefore any interactions, such as electron–electron or electron lattice interactions, would lead to departure from the simple Drude model, which should show up as nonlinear terms. In fact, even the linear term should not be the same expression as the simple plasmonic frequency. The best way to understand the trend is to consider all the interacting terms in terms of a density matrix having all of the off-diagonal terms. When one diagonalizes the density matrix, one would find that the new frequency would not be given by the fundamental with 2× or 3× or 4×, etc.

15. IMPORTANCE OF PHASE COHERENCE

Having made a clear distinction between waves and particles, we recognize the fact that waves are represented by second-order differential equations and are characterized by their phase and amplitude. In quantum theory there is no direct physical meaning attributed to complex wave functions describing the quantum state of a particle, but its magnitude can be interpreted as a probability density. Making Schrödinger's equation invariant to wave-function-phase leads naturally to vector and scalar quantities that satisfy Maxwell's equations given for atomic systems. In describing wave interactions with matter, we also define a probability density to get rid of the phase, making the problem more closely related to that for particles. That step can be done with quantum mechanics or classical mechanics treatments of wave–particle interactions. However, once phase is removed, interference is removed; particles no longer have phase velocity. Our constitutive equations attributing properties to materials, including metamaterials, are based on this. In other words, they are represented by the RPA, namely, in dielectric functions, expansion coefficients, etc., and phase disappears from further considerations. However, we are advocating here that with metamaterials we need not do this. The way to introduce phase in constitutive relationships requires a much more complicated approach based on many-body theory and it is this very problem that was referred to earlier as homogenization, namely, reliably extracting constitutive parameters from an array of coupled meta-atoms.

As long as the RPA is considered an approximation, there is the possibility of adopting approaches used in the diagonalization of an infinite matrix into blocks, e.g., introducing phase relationships among the elements within a given block while eliminating phases among various blocks. Conversely, one could keep phase relationships among the blocks but use the RPA for the individual blocks. This last scheme is precisely what is involved with superlattices, where phase is reintroduced among various heterojunctions. To put it in a different perspective, terminating a tight binding computation of a band structure achieves some aspect of incorporating a range for the RPA, such as assuming an energy-dependent collision frequency.

Our earlier H-atom model involves very high reflectivity, up to six significant figures approaching unity, in order to produce trapping of millions of cycles of the light incident onto the atom. However we observe that typical man-made resonant systems, regarded as LCR circuits, have a Q that might only reach a few thousand mainly because materials are lossy. It appears that the type of resonance presented in this work may be imitated by a system having a resonance that need not be restricted by material media with high dielectric losses or transport losses. This suggests the use of configurations with multipole electrodes as originally expounded by Tsu [25]. The main concept involves the use of electrodes, which we could equally well refer to as meta-atoms.

16. NONLINEAR METAMATERIALS AND THE RPA

We have discussed how all-interacting systems exhibit some nonlinearities in their response. As a result, the question is whether one can engineer a metamaterial composed of designed meta-atoms that could exhibit much larger and hence more useful nonlinear responses than most naturally occurring materials. By suitable design of a meta-atom's response, highly nonlinear behavior can be realized. A recent example of this is to place a diode in the gap of a split-ring resonator. Also, since a bulk metamaterial is fabricated from large numbers of these meta-atoms, fabrication tolerances and meta-atom spacings are parameters we can control. This allows short range co-operative or phase coherent responses to build the overall number, N, of participating electrons per unit volume. Averaged responses defining the constitutive relations based on the RPA can be diminished, resulting in properties that are governed only by the central limit theorem as it applies to large numbers of the unit volumes, each of which is responsible for coherent responses.

In all cases, the time dependence of the interaction of the incident (plane) wave with the eigenstates of the atom or meta-atom, will be causal, and a (short-time) Fourier transform provides real and imaginary parts of the frequency-dependent constitutive parameters. These real and imaginary parts evolve with time but are still inevitably dependent on each other as a result of Cauchy's integral formula. Close to resonant frequencies we can predict effective parameter swings that can be large and physically significant for very high-Q systems. These Lorentzian responses, dictated by causality, are consistent with a simple Drude model for a free-electron response. Lower Q and faster responses lead to smaller net effects and correspondingly smaller nonlinearities. Increasing disorder of atoms or meta-atoms from whatever physical cause leads to lower Q, reduced responses, and the material properties we have largely come to accept. It was not until the insight of the superlattice that engineering structures with local co-operating electron responses paved the way for improved electronics and, as is now clear, metamaterials with nonnaturally occurring electromagnetic responses [26]. For example, if we are interested in making a parametric amplifier, then to achieve this, the exciting or pumping field

has to be able to impose phase coherence on the system, i.e., with no RPA [27].

It would seem that efforts should be directed to realizing material systems with $Q > 10^{4-6}$ range, and these could be used for detectors and phase modulators. There is another reason why we should be looking for such Q values. The mechanisms involved in emissions from even a dipole antenna are far more involved once near field ($<\lambda$) interactions have to be included. In ordinary quantum mechanics, the interaction of electrons and photons are introduced via the generalized momentum operator. Once two systems are coupled, forming a coupled mode, there ceases to be a start and finish. In other words, when describing an interaction between A and B, we may study how A is affected by B and we may ask how A triggers B. However, with a coupled quasi-particle A–B, we have to develop a new understanding (e.g., [28]). All interacting particles have a pole-like singularity at their origin, and any interaction approaching these singularities results in interactions that are much stronger. This is borne out by the small-antenna example given above. We are arguing that reducing Q reduces response times and that this is desirable for faster next-generation devices. However, the choice of high Q or low Q depends on applications. For example, suppose we want to use A to trigger B. The trigger must not have very high Q; otherwise we would suffer a great delay in the response of our system. Now, if the detectability is very low, we are forced to utilize high Q, and it is understood that we are doing so at the expense of response time.

We also recognize that a system does not have to be represented by a single process and hence a single physical model. We can augment particular processes by combining them either in series or in parallel, since we do not want to rule out multiple components. For example, a superlattice is nothing but components put together as a single unit consistent with the principle of heterojunctions. Within each individual section, the principle of the RPA is applied, while between the components, phase relationships are kept. This is how resonant tunneling devices were introduced. While individual sections are represented by the RPA, phase relationships are kept between these individual epitaxial layers. In our view, this opportunity is precisely what metamaterials offer, and the consequences of avoiding the RPA opens up the possibility of completely new classes of devices. To rephrase our major point, for both superlattices and metamaterials, we can exploit the common practice of having identical systems operating in parallel.

With regard to metamaterial design, consider nonlinear interactions. In the usual approach [11] we consider incident light interacting with an electron or system of electrons including those inside an atom via the term $\mathbf{p} = \mathbf{p} - (e/c)\mathbf{A}$ where $\mathbf{A}$ is the vector potential field in the kinetic energy term of the Hamiltonian. The arrangement of atoms or meta-atoms defines the potential energy term of the Hamiltonian. Interactions can be solved by diagonalizing the Hamiltonian, i.e., when we have all of the off-diagonal terms equal to zero. However, diagonalization results in new energy states, and for a frequency corresponding to $Energy = \hbar\omega$, we find that ω is no longer a constant of the interaction, and there are harmonics and subharmonics present. In dealing with extreme nonlinearities, such as the superlattice potential [26], the computed harmonics are much higher than the values given by use of constitutive equations. The physical reason for this is that the high nonlinearity is not based on a perturbed simple harmonic oscillator potential, but rather on whether one can drive electrons into regions having higher nonlinear potential. In fact we use this understanding routinely, for example, by applying a DC bias, and we do this with terahertz Bloch oscillators, microwave amplifiers, or even parametric amplifiers.

17. CONCLUDING REMARKS

Much important and insightful physics depends on a simple model. We should not be misled into thinking that the Lorentz model is only classical. If one starts from quantum mechanics, one would have developed a simple harmonic model, identical to the Lorentz model. Building on this, traditional models for dielectric functions, elastic constants, expansion coefficients, etc., basically utilize RPAs. Nevertheless, our simple model applies, and it applies remarkably well. In particular we noted the surprising observation and consistency of the triple resonance. The increase in Rabi frequency toward a resonant transition frequency leading to Rabi flopping has an analogy for meta-atoms akin to mode-locking from which higher harmonics can be generated. Our approach is in some respects just the recognition that the boundary forming the interaction of the charge response of an atom with light is no more than defining a volume for the boundary value problem, thereby defining a cavity which at high energy densities can lead to even stronger interactions. Under these conditions, a high Q need not be slow, since a background field sustains the response and allows extreme changes in permittivity and permeability, as dictated by Kramers–Kronig dispersion relations, to occur near resonant frequencies with minimal losses. The boundary for an atom was defined by a sphere allowing the interaction with the series expansion of the incident wave, and it gives rise to the value of the measured linewidth of the atomic transitions. The specific line shape of a meta-atom lends itself to an entirely equivalent analysis.

A molecule, quantum dot, or meta-atom has many electrons participating. We expect the shape and size of this box to be important and to determine the resonant frequencies or eigenstates. For an atom, eigenstates are a function of energy ($\hbar\omega$) and momentum ($\hbar k$, i.e., shape). Shape may be more important in determining the electromagnetic response for larger structures containing many carriers, since there are more degrees of freedom to induce complex magnetic as well as electronic interactions. This is especially the case at higher (e.g., optical) frequencies at which magnetic responses are typically ignored. As for an atom, we might expect an excitation to take time to build up depending on the Q of the box, but that time will depend on the rate of transfer of energy into the box. Transitions between eigenstates or modes will again require sufficiently high frequencies for the interacting electromagnetic wave for certain eigenstate transitions to occur, but now momentum states may be more important because of meta-atom shapes.

For either real or meta-atoms, below their ionization frequency or the required energy density to produce ionization, electron motion is confined to the cavity or box described earlier. This confinement must accommodate nonlinearity with increasing electromagnetic energy densities. The smaller the box, then the easier it might be to induce a nonlinear response: hence our hope to be able to design small

(on the scale of the wavelength) meta-atoms. However, larger nonlinearities are customarily slower. This is consistent not with the reduced N (weaker effect) but the correspondingly larger Q as we approach the Chu limit.

Based on this, we make the following observations with regard to the design of metamaterials:

1. Meta-atoms need not necessarily be spaced equally, and might preferably be in a compact almost-periodic system.
2. Meta-atoms may contain switchable features such as many orders of magnitude change in the conductivity of the elements due to a phase change, for example, allowing one to be able to change their functionality by external voltage, light, or a magnetic field; a phase change such as the $1s - 2p$ transition is always ultimately an electronic process.
3. Nonlinearities and diversity of meta-atom shape and electromagnetic response allow for a completely new class of materials that are not bound by the restrictions of symmetries and averaging (RPA) that naturally occurring materials are subject to.
4. Generally, with the size of individual elements or meta-atoms are much less than the wavelength of light, but fabrication realities demand metamaterial designs involving smaller numbers of elements that need not necessarily be $\ll\lambda$, but merely $<\lambda$. Chemical as well as dimensional stability is important for small meta-atoms.
5. Higher-power devices and faster response times typically call for one to maximize or minimize certain features. Metamaterials are man-made engineered materials that, as we understand the underlying wave–metamatter interactions more deeply, should lead to unlimited design opportunities in the future.

Developing improved meta-atom shapes, including exploiting symmetries, suggests a simple set of building blocks that are both general purpose and manufacturable. Research has been reported over the last couple of years examining the effects of symmetry and proximity of meta-atoms [e.g., 29] to better explain overall electromagnetic responses at a fundamental level.

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