Switching Purcell effect with nonlinear epsilon-near-zero media

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An optical topological transition is defined as the change in the photonic iso-frequency surface around epsilon-near-zero (ENZ) frequencies which can considerably change the spontaneous emission of a quantum emitter placed near a metamaterial slab. Here, we show that due to the strong Kerr nonlinearity at ENZ frequencies, a high-power pulse can induce a sudden transition in the topology of the iso-frequency dispersion curve, leading to a significant change in the transmission of propagating as well as evanescent waves through the metamaterial slab. This evanescent wave switch effect allows for the control of spontaneous emission through modulation of the Purcell effect. We develop a theory of the enhanced nonlinear response of ENZ media to s and p polarized inputs and show that this nonlinear effect is stronger for p polarization and is almost independent of the incident angle. We perform finite-difference time-domain (FDTD) simulations to demonstrate the transient response of the metamaterial slab to an ultrafast pulse and fast switching of the Purcell effect at the sub-picosecond scale. The Purcell factor changes at ENZ by almost a factor of three which is an order of magnitude stronger than that away from ENZ. We also show that due to the inhomogeneous spatial field distribution inside the multilayer metal-dielectric super-lattice, a unique spatial topological transition metamaterial can be achieved by the control pulse induced nonlinearity. Our work can lead to ultra-fast control of quantum phenomena in ENZ metamaterials.

Progress in quantum information processing at optical frequencies can be accelerated by developments in the control and manipulation of light-matter interaction at the nanoscale. Some recent achievements include on-chip superconducting single photon detectors, spin routing of single photons, long range dipole-dipole interactions, enhancement or suppression of vacuum fluctuations in a selected spectrum, modulation and switching of light sources, control of evanescent waves, and switching at the single photon level. However, all-optical manipulation and switching at the same time is a difficult task as light hardly interacts with the same kind of mode due to field enhancement and spatially coherent wave behavior in these media. This has led to many interesting phenomena and applications such as controlling thermal emission, observation of Ferrel-Bereman modes, super-coupling, and enhanced dipole-dipole interaction. Recently, strong nonlinearity in ENZ media has become a domain of interest because of significant change in the refractive index around the ENZ frequencies as well as the field enhancement due to boundary effects at the high contrast interfaces. Phase mismatch-free propagation, and strong nonlocality at ENZ are also important aspects. Here, we emphasize the second approach due to ease of fabrication and experimental verification. Figure 1a shows the parallel \( \epsilon_{xx} = \epsilon_{yy} \) and normal \( \epsilon_{zz} \) components of the effective dielectric tensor of an Ag/TiO\(_{2}\) multilayer metamaterial derived from Maxwell-Garnett effective medium theory (EMT). An optical topological transition from type I to type II is seen at the ENZ wavelength. HMMs show interesting characteristics due to their open dispersion surfaces and the coupling of high-k modes (evanescent waves in vacuum) to the metamaterial modes. Figure 1c shows the transmission of low-k \( (k_x < k_0) \) and high-k \( (k_x > k_0) \) modes through the multilayer using transfer matrix method. The peaks cor-

HMMs are a class of non-magnetic uniaxial metamaterials with double-sheeted (type I) or single-sheeted (type II) hyperbolic iso-frequency dispersion curves. There are two well-known approaches for practical realization of HMMs: metallic nanowires in a dielectric host matrix, and metal-dielectric multilayers. Here, we propose experiments to verify our predictions taking into account the role of dispersion, absorption, and finite unit-cell size in practical metal-dielectric nonlinear HMMs. Furthermore, to understand the response of the metamaterial to ultra-fast pulses, we carry out finite-difference time-domain (FDTD) simulations. We propose optimum gate pulse widths to open up the possibility for switching Purcell effect with sub-picosecond response times using ENZ media.

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Figure 1. Switching of Purcell effect. (a) A nonlinear gate signal controls the transmission of evanescent waves emitted by a quantum emitter above HMM around the ENZ frequencies. (b) The real part of effective permittivity of the Ag/TiO₂ multilayer metamaterial with Silver filling fraction of $\rho = 0.5$. The inset illustrates the iso-frequency dispersion curve of type I and type II HMMs. An optical topological transition is observed at ENZ. (c) Transmission of propagating and evanescent $p$-polarized waves through an Ag/TiO₂ multilayer with a total thickness of 400 nm and a periodicity of $\Lambda = 50$ nm. The transmission of high-$k$ modes in both regions is high, but in type I region, the effect is stronger, especially close to the ENZ wavelengths. Due to the finite thickness of the unit-cell, the ENZ wavelength is red-shifted in multilayer structure in comparison with EMT. (d) Purcell factor for a vertically oriented dipole above the slab modeled by EMT. A rapid change in the Purcell factor is observable at ENZ due to the optical topological transition. Other peaks in type I correspond to slow-light modes of the HMM slab.

Figure 2. Nonlinear response at ENZ. Nonlinear response of (a) $s$ polarized and (b) $p$ polarized incidences at different incident angles for the multilayer structures in Fig. 1 at $\lambda = 522$ nm which is slightly longer than the ENZ wavelength. As the input power increases, the transmission increases due to the change in effective permittivity of the multilayer slab. The power level is independent of the incident angle for the $p$-polarization. It is important to note that ENZ media show bistable behavior in the presence of low loss (inset). This bistability does not exist if we take into account nonlinear absorption i.e. imaginary part of $\chi^{(3)}$.

Metals have very large Kerr nonlinearity ($\epsilon_{NL} = \epsilon_L + 12\pi\chi^{(3)}|E|^2$) where $\epsilon_L$, $\chi^{(3)}$, and $|E|$ are linear permittivity, Kerr susceptibility, and the electric field amplitude, respectively) which is orders of magnitude larger than that of dielectrics. However, since they are strongly reflective at optical frequencies, the electric field decays very fast inside the bulk metals, so their nonlinearity is not easily accessible. Enhancing the nonlinear response of metals in inhomogeneous structures at transparent window has been proposed for applications such as optical limiting and switching of propagating waves. As illustrated in Fig. 1.(c), metal/dielectric multilayer structures are transparent in HMM type I region where $\epsilon_{xx} > 0$. Thus, the nonlinear response in that range of frequencies can considerably be enhanced. Furthermore, a small change in permittivity around the ENZ point can lead to a topological transition in HMMs from type II to type I.

To calculate the nonlinear response, we use a finite difference method to discretize the wave equation in structures which are inhomogeneous only in one direction i.e. stratified structures. Nonlinear wave propagation within a stratified medium has been analytically studied only for $s$ polarized incidences and has been approximated for $p$ polarized incidences. However, the recent observation of strong nonlinearity at the ENZ wavelengths for $p$ polarized incidences raises the need for a robust analytical formulation to study full-wave non-
linear wave propagation for $p$ polarization as well. We have modified the method proposed by Bennink et al.\textsuperscript{57} to solve the nonlinear response for $p$ polarized waves.

For an $s$ polarized plane wave propagating through a multilayer structure which is inhomogeneous only in the $z$ direction, the wave equation can be written as:

$$\frac{d^2 E_x(z)}{dz^2} + \left( k_0^2 \epsilon_{NL}(z) - k_x^2 \right) \frac{d}{dz} \left[ \frac{1}{k_0^2 \epsilon_{NL}(z) - k_x^2} \right] \frac{dE_x(z)}{dz} + \left( k_0^2 \epsilon_{NL}(z) - k_x^2 \right) E_x(z) = 0, \quad (1a)$$

$$E_z = \frac{ik_x}{k_0^2 \epsilon_{NL}(z) - k_x^2} \frac{dE_x(z)}{dz}. \quad (1b)$$

Since $\epsilon_{NL}$ depends on both $E_x$ and $E_z$, the wave equation for the $p$ polarized incidence cannot be solved explicitly as what we did for the $s$ polarization case. However, with a recursive algorithm, accurate result is achievable. The nonlinear response for the $p$ polarized case is plotted in Fig. 2.(b). It is seen that the response is almost independent of the incident angle, contrary to the $s$ polarized cases.

Figure 3.(a) displays the electric field distribution inside the EMT structure at $\lambda = 500$ nm when $I_{in} = 8$ GW/cm$^2$. Since the transmission is not significantly high, the electric field intensity is larger around the input interface. Due to the inhomogeneous field distribution, the permittivity changes inhomogeneously inside the structure. Hence, we can see a transition point at which the effective permittivity sign is flipped (Fig. 3.(b)). This type of inhomogeneous metamaterials in which the iso-frequency dispersion curve changes from type II to type I HMMs is known as “transition ENZ metamaterials”\textsuperscript{65–67}. This phase change is controlled by the electromagnetic field and is fundamentally different from the natural phase change materials like VO$_2$.\textsuperscript{68} Note that the operating wavelength of the structure is tunable by changing layers thickness or changing the layer materials.

Propagation of a nonlinear light passing through a multilayer metamaterial slab not only modifies the transmission of the propagating waves, but also changes the tunneling of the evanescent waves through the metamaterial slab. This allows us to control spontaneous emission rate of an emitter near the slab. Figure 4 compares transmission through the slab with and without the presence of the nonlinear signal near ($\lambda =-522$ nm) and away ($\lambda =450$ nm) from the ENZ wavelength. The control signal is a $p$ polarized wave at $\lambda = 522$ nm with $I_{in} = 8$ GW/cm$^2$ and $\theta = 45^\circ$. An observable change in transmission of both propagating and evanescent waves is seen around the ENZ, and it looks similar to the transmission at a shorter wavelength ($\lambda = 510$ nm) when the pump signal is off. This shows how the nonlinear response due to the control pulse effectively shifts the dispersion of the metamaterial slab around the ENZ. Away from the ENZ, the control pulse does not change the topology of the dispersion curve and hence the change in transmission is not significant for a wide range of wave vectors (Fig. 4.(b)). For the case at ENZ, the gate control signal changes the Purcell factor from 41 to around 15. This observable change allows us to control the coupling of the
and the velocity of wave propagation through the HMM slab. As we increase the pulse-width, the transmission increases gradually (Fig. 5, (a) inset) and the steady state response is reached when the pulse width is longer than 50 fs. It means that 50 fs is enough for the wave to pass and be distributed in the metamaterial slab and simultaneously change the nonlinear permittivity of the slab. Note that if we consider $\chi^{(3)}$ to be dispersive, the nonlinear dielectric response is not instantaneous, but the response to the nonlinear pulse can still occur at sub-picosecond scale$^{37,88,70}$.

Figure 5.(b) shows the switching of the Purcell effect as a function of time when the pulse width is 50 fs using the time-dependent nonlinear permittivity derived from the FDTD simulations. More than two-fold Purcell factor reduction is seen near the transition point when the gate pulse is on. The numerical simulations are in good agreement with the analytical calculations. The change in the Purcell factor far away from the transition point is less than 10%. The change in Purcell factor for isotropic media at ENZ is also negligible$^{71}$ (See Supplemental Materials). This means that switching of the Purcell effect is only possible around the ENZ wavelength.

In conclusion, we have proposed an all-optical nonlinear approach to control the transmission of propagating waves as well as the tunneling of the evanescent waves through an HMM slab around ENZ wavelengths. We have shown that the nonlinear effect is strong enough to change the topology of the HMM slab. This can lead to a significant change in the Purcell factor of a quantum emitter near the slab. We have performed full-wave FDTD simulations to show the effect of pulse-width on the nonlinear response of the slab. The switching can happen at sub-picosecond time scales. Our predicted nonlinear effect is spectrally sensitive and can lead to control of quantum and nonlinear photonic phenomena in the ENZ regime.

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