Casimir light in dispersive nanophotonics

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Abstract: We present a theory of entangled photon pair emission through the dynamical Casimir effect in systems which are simultaneously dispersive and time dependent. We show surface phonon polariton emission in time-modulated polar insulators. © 2021 The Author(s)

The nonvanishing zero-point energy of quantum electrodynamics leads to a variety of observable consequences such as atomic energy shifts, forces, and non-contact friction. It is known that in time-varying systems, electromagnetic vacuum fluctuations can lead to the production of real photons. Famously, the “dynamical Casimir effect” predicts how a cavity with rapidly oscillating walls can produce entangled photon pairs through parametric down-conversion of vacuum fluctuations. Other related phenomena include photon emission from rotating bodies, spontaneous parametric down-conversion in nonlinear materials, the Unruh effect for relativistically accelerating bodies, and even Hawking radiation from black holes [1]. Despite these interests, the field notably lacks a flexible and materials focused description of these phenomena which incorporates arbitrary geometry, and materials with dispersion and dissipation [2]. Such a framework is of paramount importance if modern material and nanofabrication platforms are to be used to optimize these effects for potential applications in quantum information, spectroscopy, imaging, and sensing.

Here, we present a theoretical framework, based on macroscopic quantum electrodynamics (MQED), for computing rates of dynamical vacuum emission in arbitrary nanostructured, dispersive, and dissipative time-dependent systems. As an example, we apply our theory to a thin film of the polar insulator hexagonal boron-nitride (hBN), whose transverse optical phonon frequency is rapidly modulated in time. This system is a highly dispersive time dependent medium, which produces entangled surface phonon-polaritons. Our results are particularly relevant in the context of recent experiments with the polar insulator silicon carbide (SiC), which have observed parametric amplification of optical phonons in the presence of a strong driving field, which effectively causes the TO phonon frequency to vary in time [3]. Moreover, we find that the high density of states of surface polariton modes in planar systems leads to photon pair generation efficiencies which are orders of magnitude higher than in a bulk.

There are inherent subtleties associated with dielectrics which simultaneously exhibit dispersion and time dependence. The resolution of this difficulty comes through considering linear response theory with broken time-translation symmetry. A field breaking time translation symmetry by modulating a material in time requires that linear response of the material be described by a susceptibility \( \chi(\omega, t, t') \). Consequently, the frequency response must be characterized by a two-frequency susceptibility \( \chi(\omega_1, \omega_2) \equiv \int dt dt' \chi(t, t') e^{i \omega_1 t'} \). In this case, the polarization field \( P(r) \) is connected to the electric field \( E \) through a more general constitutive relation \( P(\omega) = \varepsilon_{bg}(\omega) E(\omega) \). In our work, we assume that a background dielectric described by \( \varepsilon_{bg}(r, \omega) \) is a given small time-dependent modulation \( \Delta \varepsilon(r, t) \), so that the whole system is described by \( \chi(r, \omega, t') = 2 \pi \delta(\omega - \omega') \chi_{bg}(r, \omega) + \Delta \chi(r, \omega, t') \).

Our results for two-photon emission are based on a Hamiltonian theory which describes quantum photon interactions in time-varying media. The interaction Hamiltonian is given as \( V(t) = -e_0 \int d^3 r dt' \Delta \varepsilon_{bg}(r, t, t') E(r, t') E(r, t) \), where \( \Delta \varepsilon \) is the change to the susceptibility, and \( E \) is the electromagnetic field operator in the interaction picture, quantized through MQED [4]. We find that when the index change is perturbatively small, the total probability \( P \) of a two-photon emission event is given as

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P = \frac{1}{2 \pi e^2} \int_0^\infty d\omega d\omega' (\omega \omega')^2 \int d^3 r d^3 r' \text{Tr} \left[ \Delta \chi(r, \omega, -\omega') \text{Im} G(r, r', \omega') \Delta \chi^\dagger(r', \omega, -\omega) \text{Im} G(r', r, \omega) \right],
\]

where \( G \) is the dyadic Green’s function of Maxwell’s equations in the background medium, which accounts for dispersion and dissipation of the background structure. Notably, the presence of \( \Delta \chi(r, \omega, -\omega') \) accounts for dispersion, even in the presence of fast time modulation.

As an application of our theory, we consider the generation of entangled phonon-polariton pairs on hexagonal boron nitride (hBN) from time modulations of the structure. In polar insulators (such as hBN), the dielectric tensor is diagonal, with components described by \( \varepsilon_{bg}(\omega) = \varepsilon_0 + \omega_{\text{ph}}^2/(\omega_p^2 - \omega^2 - i \Gamma) \), where \( \omega_\text{ph} \) is the transverse optical phonon frequency, \( \omega_p \) is the plasma frequency, and \( \Gamma \) accounts for damping. Owing to its negative permittivity over a frequency bandwidth known as the Restrahlen (RS) band, thin films of hBN have been observed to support highly confined surface phonon-polaritons, which have the potential to enable strong light-matter interactions. The dispersion relation for these modes on
Fig. 2. (a) Thin film of hBN ($d_{slab} = 100$ nm), where a thin layer ($d = 10$ nm) is modulated in time. (b) Dispersion of surface phonon-polaritons on hBN in the upper Reststrahlen band. (c-d) Differential photon emission rate per unit area for a nondispersive modulation of top layer of the structure in (a). Short and long pulses durations are shown. (e) Total emission rate per area of phonon-polariton pairs as a function of pulse duration $T$ and frequency $\Omega_0$. (f-h) Same as (c-e), except that the modulation is dispersive. a 100 nm thin film of hBN is shown in Fig. 2a. To highlight the interplay between dispersion and time dependence in two-phonon spontaneous emission, we compare two different modulations of the hBN structure. The first is a nondispersive modulation, where a layer of thickness $d$ has its index perturbed by a constant amount as $\varepsilon(t) = \varepsilon_0(1 + \delta\varepsilon f(t))$. In this case, we have $\Delta\chi(\omega, \omega') = \delta\varepsilon f(\omega - \omega')$, where $f(\omega)$ is the Fourier transform of the modulation profile. If the change in index is caused by a nonlinear layer with $\chi(2) = 100$ pm/V, then an electric field strength of $10^7$ V/m gives $\delta\varepsilon = 10^{-3}$. The second is a dispersive modulation, where over a thickness $d$, the transverse optical phonon frequency $\omega_0$ is modulated to deviate from its usual value as a function of time as $\omega(t) = \omega_0(1 + \delta\omega f(t))$. In this case, $\Delta\chi(\omega, \omega') = \delta\omega \omega_0^2 f(\omega - \omega')/(\Omega(\omega)\Omega(\omega'))$, where $\Omega(\omega) \equiv \omega_0^2 - \omega^2 - i\Omega$. From the experimental models presented in [3] for SiC, we estimate that an applied field strength of 1 GV/m gives rise to a frequency shift of the order $\delta\omega = 10^{-3}$.

We study how the thin film responds to modulations of the form $f(t) = \cos(\Omega_0 t)e^{-t^2/2\Gamma^2}$ where $\Omega_0$ is the modulation frequency, and $T$ sets the timescale, enabling pulses to be ultrashort, or nearly monochromatic. In Fig. 2, we show how the spectral rate $d\Gamma/d\omega$ normalized to the sample area $A$ evolves with pulse duration and modulation frequency. The behavior of the rate spectrum $d\Gamma/d\omega$ depends on where $\Omega_0/2$ lies in the RS band. The two photon nature of this process results in several important features. The wavy fringes seen in the emission spectra are a result of interference between many possibilities for how two polaritons can distribute themselves into many branches of the dispersion. Additionally, we note that for nearly monochromatic pulses ($T = 5$ ns), the frequency distribution of radiation is symmetric about $\Omega_0/2$, since the two polariton frequencies produced must satisfy $\omega + \omega' = \Omega_0$. Importantly, we also see a substantial difference between the spectrum of emission that occurs from modulation $\delta\varepsilon$ of a background index, and that from modulation $\delta\omega$ of the phonon frequency $\omega_0$. For the nondispersive index modulation, emission is strongest for $\delta\omega/\omega_0 = 2.15$, essentially as a result of density of states for photon pairs. For the same reason, emission at $\Omega_0/\omega_0 = 2$ is at least an order of magnitude weaker, since the bottom of the RS band has a lower density of states. For a dispersive modulation, the emission strengths are reordered entirely, and the strongest emission occurs for degenerate production around $\omega_0$ when the system is modulated at $2\omega_0$. This dramatic change in behavior is owed to the strong resonance of the dispersive $\Delta\chi$, which is not present for the simple modulation. These differences are manifest not only in the shape of the spectrum, but in the overall strength of each process. We have computed that with an applied field strength of 1 GV/m, the nondispersive modulation achieved through a thin $\chi(2)$ layer has a quantum efficiency of the order $\eta = 10^{-9}$, while at the same field strength, the dispersive modulation should have $\eta = 10^{-5}$.

The Hamiltonian MQED formalism we have presented can enable further studies of light-matter interactions in highly general time-dependent materials. For example, one could model how quantum spontaneous emission is modified in the presence of strong time-modulation. Higher order processes, such as ground-state energy level shifts in the presence of time modulation could also be investigated. This kind of formalism could provide opportunities for studying the role that parametric amplification of quasiparticles can play in exotic condensed matter effects such as light-induced superconductivity.

We anticipate that our framework will be of interest for describing classical and quantum phenomena in many timely experimental platforms featuring ultrafast optical modulation of materials.

References
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