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**Exploring nonlinear optical properties of graphene from microwave to mid-infrared frequencies**

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<b>14. ABSTRACT</b> Six projects supported by the award were completed. They resulted in seven published papers and one more paper is submitted as an invited paper to APL-Photonics. (1) A rigorous QM theory of the second-order nonlinear response of two-dimensional (2D) massless Dirac fermions in graphene and topological insulators has been developed. The resulting nonlinear susceptibility tensor satisfies all symmetry and permutation properties with respect to its indices and arguments, is gauge-invariant, and has a correct quasiclassical limit for low frequencies. The theory has been applied to a number of nonlinear optical processes which form the basis of ultracompact, high-performance optoelectronic devices for ultrafast secure communications, remote sensing, and imaging. (2) A new optical mechanism of spin-valley entanglement of excitons in transition metal dichalcogenide monolayers has been proposed which can be utilized in future systems for quantum communications and information processing. (3) A theory of the third order nonlinearity of 2D electron gas in coupled quantum well systems has been developed and applied to the four-wave mixing and phase locking of laser modes in quantum cascade lasers (QCLs). The collaborators at Sheffield experimentally demonstrated active mode locking of mid-infrared QCLs, which is a major milestone in laser science. (4) In the presence of a magnetic field, a nonperturbative theory for 2D magnetoexcitons interacting with terahertz radiation has been developed and applied to explain the striking stability of 2D magnetoexcitons against dissociation (the absence of the Mott transition) at high magnetic fields, observed by experimental collaborators at Rice University. (5) A strong enhancement of both spontaneous emission and nonlinear optical response of graphene and other 2D materials in subwavelength 2D cavities has been predicted.			
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**Exploring nonlinear optical properties of graphene from microwave to mid-infrared frequencies**

Six projects supported by the award were completed. They resulted in seven published papers, including such high-profile journals as Nature Communications and Physical Review Letters [1–8]. One more paper is submitted as an invited paper to APL-Photonics. (1) A rigorous quantum-mechanical theory of the second-order nonlinear response of two-dimensional (2D) massless Dirac fermions in graphene and topological insulators has been developed. The resulting nonlinear susceptibility tensor satisfies all symmetry and permutation properties with respect to its indices and arguments, is gauge-invariant, and has a correct quasiclassical limit for low frequencies. The theory has been applied to a number of nonlinear optical processes which form the basis of ultracompact, high-performance optoelectronic devices for ultrafast secure communications, remote sensing, and imaging. (2) A new optical mechanism of spin-valley entanglement of excitons in transition metal dichalcogenide monolayers has been proposed which can be utilized in future systems for quantum communications and information processing. (3) A theory of the third-order nonlinearity of 2D electron gas in coupled quantum well systems has been developed and applied to the four-wave mixing and phase locking of laser modes in quantum cascade lasers (QCLs). The collaborators at Sheffield experimentally demonstrated active mode locking of mid-infrared QCLs, which is a major milestone in laser science. (4) In the presence of a magnetic field, a nonperturbative theory for 2D magnetoexcitons interacting with terahertz radiation has been developed and applied to explain the striking stability of 2D magnetoexcitons against dissociation (the absence of the Mott transition) at high magnetic fields, observed by experimental collaborators at Rice University. (5) A strong enhancement of both spontaneous emission and nonlinear optical response of graphene and other 2D materials in subwavelength 2D cavities has been predicted and applied to the design of nonlinear photonic circuits. (6) The optical response and electrodynamic properties of massless Dirac fermions in Dirac and Weyl semimetals in a magnetic field have been investigated. The results indicate that Weyl semimetals can serve as efficient polarizers, light modulators, and natural hyperbolic materials in the terahertz (THz) and infrared spectral ranges, with applications in ultracompact photonic chips.

Projects 1 through 4 have been described in detail in the previous two reports. Here we focus on projects 5 and 6, which were the subject of investigation during the last year.

# Enhancement of the spontaneous emission in subwavelength quasi-two-dimensional waveguides and resonators

We consider a quantum-electrodynamic problem of the spontaneous emission from a two-dimensional (2D) emitter, such as graphene or a 2D semiconductor, placed in a quasi-2D waveguide or cavity with subwavelength confinement in one direction. We apply the Heisenberg-Langevin approach which includes dissipation and fluctuations in the electron ensemble and in the electromagnetic field of a cavity on equal footing. The Langevin noise operators that we introduce do not depend on any particular model of dissipative reservoir and can be applied to any dissipation mechanism. Moreover, our approach is applicable to nonequilibrium electron systems, e.g. in the presence of pumping, beyond the applicability of the standard fluctuation-dissipation theorem. We derive analytic results for simple but practically important geometries: strip lines and rectangular cavities. Our results show that a significant enhancement of the spontaneous emission, by a factor of order 100 or higher, is possible for 2D emitters in a subwavelength cavity.

## I. INTRODUCTION

Enhancement of the radiative processes due to the localization of emitters in a subwavelength cavity (so-called Purcell enhancement [9]) is a fundamental cavity-quantum electrodynamics (QED) effect which finds an increasingly broad range of applications in the areas as diverse as nanophotonics, plasmonics, linear and nonlinear optical sensing, and high-speed communications, to name a few. It has been studied theoretically and experimentally so many times that it is hard to believe that any further development is needed. However, there seems to be a significant gap in the formalism for the situations typically encountered in quantum optoelectronic devices, when the electron ensemble is out of equilibrium and there is strong dissipation both in the optical dipole oscillations in a macroscopic ensemble of fermionic emitters (e.g. electrons and holes in a semiconductor quantum well or a layer of quantum dots, or a 2D semiconductor such as MoS<sub>2</sub>, or monolayer graphene) and for the electromagnetic (EM) field in a cavity. Examples include subwavelength semiconductor lasers [10–14] and other devices or circuits with subwavelength confinement in one or more dimensions e.g. [15–17]. In this case using a simple Purcell-type factor  $\sim Q\lambda^3/V$ , where  $Q$  is a quality factor of EM modes in a cavity of volume  $V$  and  $\lambda$  is the emission wavelength, can drastically overestimate the cavity enhancement of the spontaneous emission. Although this fact is well known, a consistent QED theory including dissipation and fluctuations is usually replaced by a more phenomenological rate equations approach [11]. Recent theoretical analysis of subwavelength lasers [10] did include QED Heisenberg-Langevin equations for the EM cavity modes, but not for the dynamics of the active medium.

Here we use a consistent Heisenberg-Langevin approach [18, 19] which includes dissipation and fluctuations in the fermionic ensemble and in the EM field of a subwavelength cavity on equal footing. Note that the description of dissipation and noise of a quantum field due to its interaction with an active nonequilibrium medium require a completely different approach as compared to the effects of Ohmic losses or radiation losses in an electrodynamic system. The latter effects can be analyzed within a standard fluctuation-dissipation theorem [20]. The standard description would also work for a popular model of a dissipative reservoir as an equilibrium ensemble of oscillators [21]. This approach would only allow one to describe thermal emission from a medium in thermal equilibrium. In contrast, our formalism allows us to treat radiation effects and obtain a correct expression for the current operator including fluctuations due to nonequilibrium electron systems, e.g. in the presence of pumping, beyond the applicability of the standard fluctuation-dissipation theorem. The Langevin noise operators that we introduce are not based on a model of a thermal reservoir. Instead, they are derived directly from the condition of preserving the commutation relations and generalized Einstein relations [21]. Therefore, they can be applied to any dissipation/fluctuation mechanism.

We apply the general formalism to the problem of spontaneous emission in a quasi-2D waveguide or cavity with subwavelength confinement in one direction. Remarkably, we are able to derive closed-form analytic results for all relevant quantities such as spontaneous emission power for simple but practically important geometries: strip lines and rectangular cavities. Our results provide general framework and convenient formulas for the evaluation of enhancement of radiative processes in such systems. Our results also indicate that a significant enhancement of the spontaneous emission, by a factor of order 100 or higher, is possible for QWs and other 2D emitters sandwiched between metal plates in a subwavelength cavity.

Section II describes the spatial structure of the EM field in a subwavelength quasi-2D electrodynamic structure and develops the quantization procedure. Section III introduces coupling to the fermionic system. Section IV derives and solves Heisenberg-Langevin equations for the density operator of quasiparticles and EM field operators. It derives the expression for the spontaneous emission power and its useful limiting cases.

## II. ELECTROMAGNETIC FIELD OF A SUBWAVELENGTH CAVITY

### A. Spatial structure of the EM field modes

Consider a very thin layer of quantum dipole emitters (which we will call a quantum well (QW) for brevity, although it can be any fermionic system), placed inside a strip line or a cavity formed by two metallic planes at  $z = \pm L_z/2$  where  $L_z \ll c/\sqrt{\bar{\epsilon}}\omega$ , where  $\bar{\epsilon}$  is a typical (average) value of the dielectric constant  $\epsilon = \epsilon(z)$  of the filling; see Fig. 1.

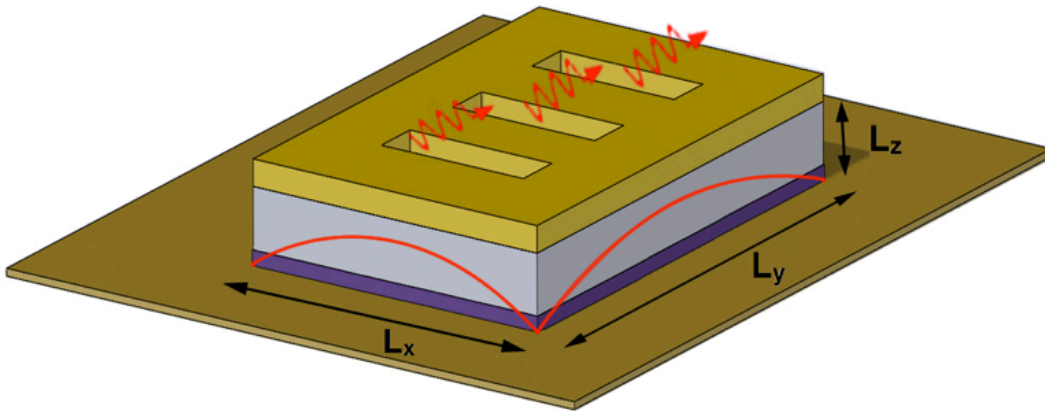


FIG. 1. A sketch of a nanocavity with thickness  $L_z$  much smaller than wavelength. An active layer of 2D emitters is shown in dark blue. The profile of the electric field of the fundamental  $TE_{011}$  mode is sketched on the sides. The radiation can be outcoupled through the gratings or cavity edges.

A TM-polarized EM field is described by the following components of the electric field, magnetic field and electric induction:

$$(\mathbf{E}_{x,z}, \mathbf{B}_y, \mathbf{D}_{x,z}) = \text{Re} \left[ \left( \tilde{\mathbf{E}}_{x,z}(z), \tilde{\mathbf{B}}_y(z), \tilde{\mathbf{D}}_{x,z}(z) \right) e^{-i\omega t + iqx} \right] \quad (1)$$

Where we assumed that the strip line is oriented along  $x$ . From Maxwell's equations,

$$\nabla \cdot \mathbf{D} = 0, \quad \nabla \times \mathbf{B} = \frac{\dot{\mathbf{D}}}{c}, \quad \nabla \times \mathbf{E} = -\frac{\dot{\mathbf{B}}}{c} \quad (2)$$

together with the material equation,

$$\mathbf{D} = \epsilon(z)\mathbf{E} \quad (3)$$

we obtain:

$$\frac{\partial \tilde{D}_z}{\partial z} = -iq\tilde{D}_x, \quad iq\tilde{B}_y = -\frac{i\omega\tilde{D}_z}{c}, \quad \frac{\partial \tilde{E}_x}{\partial z} = i\frac{\omega}{c}\tilde{B}_y(z) + i\frac{q}{\epsilon(z)}\tilde{D}_z \quad (4)$$

The first equation in (4) yields

$$\tilde{D}_z = \tilde{D}_z \left( -\frac{L_z}{2} \right) - iq \int_{-\frac{L_z}{2}}^z \tilde{D}_x dz'$$

For subwavelength thickness  $L_z q \ll 1$  the previous equation gives  $\tilde{D}_z \approx \text{const}$ , which corresponds to the quasi-electrostatic structure of the field in the  $(y, z)$  cross section of the strip line. From the second and third equations in Eq. (4) we can obtain

$$\frac{\partial \tilde{E}_x}{\partial z} = -i \frac{\omega^2 \tilde{D}_z}{qc^2} + i \frac{q}{\varepsilon(z)} \tilde{D}_z \quad (5)$$

Next we integrate Eq. (5) as  $\int_{-\frac{L_z}{2}}^{\frac{L_z}{2}} dz \dots$ , taking into account  $\tilde{D}_z \approx \text{const}$  and the boundary conditions on the metal planes:  $\tilde{E}_x(+\frac{L_z}{2}) = \tilde{E}_x(-\frac{L_z}{2}) = 0$ . As a result, we obtain the dispersion relation:

$$\frac{\omega^2}{q^2 c^2} = \frac{1}{L_z} \int_{-\frac{L_z}{2}}^{\frac{L_z}{2}} \frac{dz}{\varepsilon(z)}. \quad (6)$$

Since the direction of  $x$ -axis was arbitrary, we can represent the electric field vector as

$$\mathbf{E} = D_q \mathbf{F}_q(\mathbf{r}) e^{-i\omega_q t} + \text{C.C.}, \quad (7)$$

where the factor  $\mathbf{F}_q(\mathbf{r})$  determines the spatial structure of the field:

$$\mathbf{F}_q(\mathbf{r}) = \mathbf{z}_o \frac{e^{i\mathbf{q}\mathbf{r}}}{\varepsilon(z)}, \quad (8)$$

vector  $\mathbf{q}$  is in the  $(x, y)$  plane,  $D_q$  is a constant which in this case corresponds to a  $z$ -independent amplitude of the electric induction. According to the Brillouin concept, one can use the waves defined by Eqs. (6)-(8) to construct any waveguide and cavity modes. They have quasi-TEM polarization. In particular, if the sides  $y = \pm L_y/2$  are also metal-coated, consider the lowest order (01) waveguide mode:

$$\mathbf{E} = D_{q_x} \mathbf{F}_{q_x}(\mathbf{r}) e^{-i\omega_{q_x} t} + \text{C.C.}, \quad q_x^2 + \left( \frac{\pi}{L_y} \right)^2 = \frac{\omega^2}{c^2} \frac{L_z}{\int_{-\frac{L_z}{2}}^{\frac{L_z}{2}} \varepsilon(z)^{-1} dz} \quad (9)$$

where the explicit form to the factor  $\mathbf{F}_{q_x}(\mathbf{r}) \propto e^{-iq_x x}$  is given below. If the facets  $x = \pm L_x/2$  are metal-coated as well, the waveguide becomes a resonator and the lowest order modes are  $TE_{01N}$ :

$$\mathbf{E} = D_N \mathbf{F}_N(\mathbf{r}) e^{-i\omega_N t} + \text{C.C.}, \quad \left( \frac{N\pi}{L_x} \right)^2 + \left( \frac{\pi}{L_y} \right)^2 = \frac{\omega^2}{c^2} \frac{L_z}{\int_{-\frac{L_z}{2}}^{\frac{L_z}{2}} \varepsilon(z)^{-1} dz} \quad (10)$$

In Eqs. (9) and (10) the factors  $D_{q_x}$  and  $D_N$  are coordinate-independent amplitudes of the electric induction. The factors  $\mathbf{F}_{q, q_x, N}(\mathbf{r})$  in Eqs. (7), (9), (10) can be written in the same form using the index  $\nu = \mathbf{q}, q_x, N$  to denote a corresponding spatial structure:

$$\mathbf{F}_\nu(\mathbf{r}) = \mathbf{z}_o \frac{\zeta_\nu(x, y)}{\varepsilon(z)}, \quad \zeta_{\mathbf{q}} = e^{i\mathbf{q}\mathbf{r}}, \quad \zeta_{q_x} = \cos\left(\frac{\pi y}{L_y}\right) e^{iq_x x}, \quad \zeta_N = \cos\left(\frac{\pi y}{L_y}\right) \times \begin{cases} \cos\left(\frac{N_{\text{odd}}\pi x}{L_x}\right) \\ \sin\left(\frac{N_{\text{even}}\pi x}{L_x}\right) \end{cases} \quad (11)$$

where  $\int_S \zeta_\nu \zeta_{\nu'}^* d^2r \propto \delta_{\nu\nu'}$ . For a particular case of a uniform dielectric constant, Eqs. (6)-(11) are exact. Similar equations can be derived if one simply utilizes jumps of the dielectric constants on the sides instead of metal coating. Even without any jump in the dielectric constants, an open end of a thin waveguide with vertical size much smaller than wavelength is a good reflector and therefore any radiation losses through the facets are small and are not affecting the mode spatial structure significantly.

## B. Field quantization in a subwavelength waveguide/cavity

Here we consider field quantization in a volume  $V = L_z S$ , where  $S = L_x \times L_y$ . The field operator can be represented in a standard form [22, 23]:

$$\hat{\mathbf{E}} = \sum_{\nu} [\mathbf{E}(\mathbf{r})_{\nu} \hat{c}_{\nu} + \mathbf{E}^*(\mathbf{r})_{\nu} \hat{c}_{\nu}^{\dagger}] \quad (12)$$

where  $\hat{c}_{\nu}$  and  $\hat{c}_{\nu}^{\dagger}$  are boson annihilation and creation operators,  $\mathbf{E}_{\nu}(\mathbf{r}) = \mathbf{z}_{\mathbf{o}} \frac{\zeta_{\nu}(x, y)}{\varepsilon(z)} D_{\nu}$ , and  $D_{\nu}$  is the normalization constant corresponding to the  $z$ -independent amplitude of the electric induction. The value of  $D_{\nu}$  needs to be chosen in such a way that the commutation relation for boson operators  $\hat{c}_{\nu}$  and  $\hat{c}_{\nu}^{\dagger}$  have a standard form  $[\hat{c}_{\nu}, \hat{c}_{\nu'}^{\dagger}] = \delta_{\nu\nu'}$ . In this case the field Hamiltonian will also be standard:

$$\hat{H}_f = \sum_{\nu} \hbar\omega_{\nu} (\hat{c}_{\nu}^{\dagger} \hat{c}_{\nu} + \frac{1}{2}) \quad (13)$$

To find the explicit expression for  $D_{\nu}$  we apply the phenomenological procedure of field quantization in a medium [22, 24] which was justified in [25] based on a rigorous quantum electrodynamics theory. According to this approach, the normalization is determined by the requirement that the classical energy density  $W$  of the EM field  $\mathbf{E} = \mathbf{E}_{\nu}(\mathbf{r})e^{-i\omega_{\nu}t} + \text{C.C.}$ ,  $\mathbf{B} = \mathbf{B}_{\nu}(\mathbf{r})e^{-i\omega_{\nu}t} + \text{C.C}$  give the total energy of  $\int_V W d^3r = \hbar\omega_{\nu}$ . For our strip line this procedure yields the following expression for the normalization constant:

$$|D_{\nu}|^2 = \frac{2\pi\hbar\omega_{\nu}}{\int_S \zeta_{\nu} \zeta_{\nu}^* d^2r \times \int_{-\frac{L_z}{2}}^{\frac{L_z}{2}} \frac{1}{2\varepsilon^2(\omega_{\nu}, z)\omega_{\nu}} \left[ \frac{\partial(\omega^2\varepsilon(\omega, z)}{\partial\omega} \right]_{\omega=\omega_{\nu}} dz} \quad (14)$$

where  $\int_S \zeta_q \zeta_q^* d^2r = S$ ,  $\int_S \zeta_{qx} \zeta_{qx}^* d^2r = S/2$  and  $\int_S \zeta_N \zeta_N^* d^2r = S/4$ . In the limiting case of plane waves in a homogeneous medium Eq. (14) corresponds to a standard normalization of the electric field [22, 24, 25]; indeed, taking into account that in a homogeneous medium  $D_{\nu} = E_{\nu}\varepsilon(\omega_{\nu})$ , Eq. (14) gives  $|E_{\nu}|^2 = \frac{2\pi\hbar\omega_{\nu}}{\frac{V}{2\omega_{\nu}} \left[ \frac{\partial(\omega^2\varepsilon(\omega, z)}{\partial\omega} \right]_{\omega=\omega_{\nu}}}$ , where

$V = L_z S$  is the quantization volume.

## III. NON-DISSIPATIVE DYNAMICS OF A COUPLED SYSTEM OF PHOTONS AND ELECTRONS

### A. General formalism

We will denote a quantum state of an electron in a QW or any other 2D nanostructure by a band index  $m$  which may include also the subband, spin, and valley index as needed, and the 2D quasimomentum  $\mathbf{k}$  corresponding to the motion in  $(x, y)$  plane. The second-quantized energy of a system of such quasiparticles is

$$\hat{H}_e = \sum_{m\mathbf{k}} W_{m\mathbf{k}} \hat{a}_{m\mathbf{k}}^{\dagger} \hat{a}_{m\mathbf{k}} \quad (15)$$

where  $\hat{a}_{m\mathbf{k}}^{\dagger}$ ,  $\hat{a}_{m\mathbf{k}}$  are creation and annihilation operators of fermions,  $W_{m\mathbf{k}} \equiv W_{mm\mathbf{k}\mathbf{k}}$  are the diagonal matrix elements of the energy operator of a quasiparticle. The eigenfunctions can be written as

$$|m, \mathbf{k}\rangle = \frac{e^{i\mathbf{k}\mathbf{r}}}{\sqrt{S}} \psi_m(z) \quad (16)$$

where  $\int_S e^{i(\mathbf{k}-\mathbf{k}')\mathbf{r}} d^2r = S\delta_{\mathbf{k}\mathbf{k}'}$ ,  $\int_{-\frac{l}{2}}^{\frac{l}{2}} \psi_m(z)\psi_n^*(z)dz = \delta_{mn}$ . Here we assume that a 2D nanostructure occupies a region  $-l/2 \leq z \leq l/2$ ,  $l \leq L_z$ . The total Hamiltonian of a coupled system of photons and electrons is

$$\hat{H} = \hat{H}_f + \hat{H}_e + \hat{V} \quad (17)$$

where the operators  $\hat{H}_f$  and  $\hat{H}_e$  are given by Eqs. (13) and (15), and  $\hat{V}$  is the interaction Hamiltonian, which can also be written in the second-quantized form:

$$\hat{V} = \sum_{mn\mathbf{k}\mathbf{k}'} \hat{V}_{nm\mathbf{k}'\mathbf{k}} \hat{\rho}_{nm\mathbf{k}\mathbf{k}'} \quad (18)$$

where  $\hat{\rho}_{nm\mathbf{k}\mathbf{k}'} = \hat{a}_{n\mathbf{k}'}^\dagger \hat{a}_{m\mathbf{k}}$  is the density operator. Matrix elements  $\hat{V}_{nm\mathbf{k}'\mathbf{k}}$  in Eq. (18) are operators since they depend on the quantum field.

Taking into account the quasi-electrostatic structure of the electric field in the transverse cross-section of a strip line, we can write the interaction Hamiltonian in the electric potential approximation:

$$\hat{V} = e \int_{-l/2}^z \hat{E}_z dz \quad (19)$$

Using Eq. (12) for the field operator, the matrix elements of the interaction Hamiltonian are

$$\hat{V}_{nm\mathbf{k}'\mathbf{k}} = -\tilde{d}_{nm} \sum_{\nu} (D_{\nu} \hat{c}_{\nu} \zeta_{\mathbf{k}'\mathbf{k}}^{(\nu)} + D_{\nu}^* \hat{c}_{\nu}^{\dagger} \zeta_{\mathbf{k}'\mathbf{k}}^{(\nu)\dagger}) \quad (20)$$

where  $\tilde{d}_{nm}$  is the effective dipole moment of the optical transition:

$$\tilde{d}_{nm} = -e \int_{-l/2}^{l/2} \left[ \psi_n^*(z) \left( \int_{-l/2}^z \frac{dz'}{\varepsilon(z')} \right) \psi_m(z) \right] dz \quad (21)$$

$$\zeta_{\mathbf{k}'\mathbf{k}}^{(\nu)} = \frac{1}{S} \int_S e^{-i\mathbf{k}'r} \zeta_{\nu}(x, y) e^{i\mathbf{k}r} d^2r, \zeta_{\mathbf{k}'\mathbf{k}}^{(\nu)\dagger} = (\zeta_{\mathbf{k}'\mathbf{k}}^{(\nu)})^* \quad (22)$$

For a homogeneous medium, in which  $E_{\nu} = D_{\nu}/\varepsilon$ , Eq. (20) will contain a standard expression  $\tilde{d}_{nm} D_{\nu} = -e \langle n|z|m \rangle E_{\nu}$ . The Hamiltonian Eq. (17) gives rise to the Heisenberg equations for photon operators:

$$\begin{aligned} \dot{\hat{c}}_{\nu} &= \frac{i}{\hbar} [\hat{H}, \hat{c}_{\nu}] = -i\omega_{\nu} \hat{c}_{\nu} + \frac{i}{\hbar} D_{\nu}^* \sum_{mn\mathbf{k}\mathbf{k}'} \tilde{d}_{nm} \zeta_{\mathbf{k}'\mathbf{k}}^{(\nu)\dagger} \hat{\rho}_{mn\mathbf{k}\mathbf{k}'}, \\ \dot{\hat{c}}_{\nu}^{\dagger} &= \frac{i}{\hbar} [\hat{H}, \hat{c}_{\nu}^{\dagger}] = i\omega_{\nu} \hat{c}_{\nu}^{\dagger} - \frac{i}{\hbar} D_{\nu} \sum_{mn\mathbf{k}\mathbf{k}'} \tilde{d}_{nm} \zeta_{\mathbf{k}'\mathbf{k}}^{(\nu)} \hat{\rho}_{mn\mathbf{k}\mathbf{k}'} \end{aligned} \quad (23)$$

We write a similar equation for the density operator using a shortcut notation  $|m, \mathbf{k}\rangle = |\mu\rangle$  for brevity. Using the fundamental commutation relation [25–27]

$$[\hat{\rho}_{\mu'\eta'}, \hat{\rho}_{\mu\eta}] = (\delta_{\mu'\eta} \hat{\rho}_{\mu\eta'} - \delta_{\mu\eta'} \hat{\rho}_{\mu'\eta}) \quad (24)$$

which is valid whether the creation and annihilation operators  $\hat{a}_{\eta}^{\dagger}$  and  $\hat{a}_{\mu}$  satisfy the commutation relations for bosons or fermions, we obtain:

$$\dot{\hat{\rho}}_{\mu\eta} = \frac{i}{\hbar} [\hat{H}, \hat{\rho}_{\mu\eta}] = -\frac{i}{\hbar} \sum_{\mu'} (\hat{H}_{\mu\mu'} \hat{\rho}_{\mu'\eta} - \hat{\rho}_{\mu\mu'} \hat{H}_{\mu'\eta}) \quad (25)$$

The resulting equation for the density operator has the same form as the von Neumann equation, although the original Heisenberg equation had an opposite sign in front of the commutator [25–27]. This is to be expected, because for time-dependent Heisenberg operators  $\hat{a}_{\eta}^{\dagger}$  and  $\hat{a}_{\mu}$  the average of dyadics  $\hat{\rho}_{\mu\eta} = \hat{a}_{\eta}^{\dagger} \hat{a}_{\mu}$  over the initial quantum state should correspond to a usual density matrix.

## B. Matrix elements of the interaction Hamiltonian

The form of the interaction Hamiltonian for the fields with different spatial structure depends on the matrix elements  $\zeta_{\mathbf{k}'\mathbf{k}}^{(\nu)}$  defined in Eq. (22). In particular, for plane waves we obtain  $\zeta_{\mathbf{k}'\mathbf{k}}^{(q)} = \delta_{\mathbf{k}'; \mathbf{k}+\mathbf{q}}$ .

If we take into account that the de Broglie wavelength of electrons is typically much smaller than the spatial scale of the EM field, i.e.  $k \gg |\mathbf{q}|, q_x, \frac{\pi N}{L_x}, \frac{\pi N}{L_y}$ , the expressions for matrix elements are simplified. Indeed, in this case we can assume that the optical transitions are direct in momentum space and take  $\zeta_{\mathbf{k}'\mathbf{k}}^{(\nu)} \approx \alpha_\nu \delta_{\mathbf{k}'\mathbf{k}}$ . The factor in front of the delta-function is one for plane waves; for a waveguide or a cavity one should choose  $\alpha_\nu = \sqrt{\sum_{\mathbf{k}'} \zeta_{\mathbf{k}'\mathbf{k}}^{(\nu)} \zeta_{\mathbf{k}\mathbf{k}'}^{(\nu)\dagger}}$ . With this choice, a resonance line which is smeared in the quasimomentum space can be reduced to the delta-function  $\alpha_\nu \delta_{\mathbf{k}'\mathbf{k}}$  while conserving the sum of intensities of all transitions within the line. The Parseval theorem then gives  $\sum_{\mathbf{k}'} \zeta_{\mathbf{k}'\mathbf{k}}^{(\nu)} \zeta_{\mathbf{k}\mathbf{k}'}^{(\nu)\dagger} = S^{-1} \int_S \zeta_\nu \zeta_\nu^* d^2r$ . As a result the matrix element can be written in the same form for plane waves, in a waveguide, and in a cavity:

$$\hat{V}_{nm\mathbf{k}\mathbf{k}'} \approx -\tilde{d}_{nm} \sum_\nu (\tilde{D}_\nu \hat{c}_\nu + \tilde{D}_\nu^* \hat{c}_\nu^\dagger) \delta_{\mathbf{k}'\mathbf{k}} \quad (26)$$

where

$$|\tilde{D}_\nu|^2 = \frac{2\pi\hbar\omega_\nu}{SG(L_z, \omega_\nu)} \quad (27)$$

$$G(L_z, \omega_\nu) = \int_{-\frac{L_z}{2}}^{\frac{L_z}{2}} \frac{1}{2\varepsilon^2(\omega_\nu, z)\omega_\nu} \left[ \frac{\partial(\omega^2\varepsilon(\omega, z))}{\partial\omega} \right]_{\omega=\omega_\nu} dz \quad (28)$$

Note that in a uniform nondispersive medium  $\tilde{d}_{mn} = d_{mn}/\varepsilon$  and  $G = L_z/\varepsilon$ .

### C. The probability of the spontaneous emission

Consider a spontaneous radiative transition  $m \rightarrow n$  for a quasiparticle in an open electrodynamic system, e.g. in the space between two conducting planes or in a waveguide. The transition probability is usually calculated using Fermi's golden rule [28]:

$$A_{m \rightarrow n} = \frac{2\pi}{\hbar^2} \int d\Pi_f |V_{fi}|^2 \delta\left(\frac{W_i}{\hbar} - \frac{W_f}{\hbar} - \omega_\nu\right) \quad (29)$$

where the integration  $\int d\Pi_f$  is taken over all final states of a system labeled by  $f$ . The matrix element  $V_{fi}$  in this case is equal to  $\langle 1_\nu | \hat{V}_{nm\mathbf{k}'\mathbf{k}} | 0_\nu \rangle$ , where  $|n_\nu\rangle$  is a Fock state of photons. Using Eqs. (20) and (26)-(28) we obtain

$$V_{fi} = -\tilde{d}_{nm} D_\nu^* \zeta_{\mathbf{k}'\mathbf{k}}^{(\nu)\dagger} \approx -\tilde{d}_{nm} \tilde{D}_\nu^* \delta_{\mathbf{k}'\mathbf{k}} \quad (30)$$

Taking into account the photon density of states, one can get for the radiation emitted into space between two conducting planes

$$d\Pi_f = \frac{S|\mathbf{q}|d\theta d\omega_q}{(2\pi)^2 |\partial\omega_q/\partial\mathbf{q}|}$$

where  $\theta$  determines the direction of vector  $\mathbf{q}$  in the  $(x, y)$  plane. For the radiation emitted into a waveguide,

$$d\Pi_f = \frac{L_x d\omega_{q_x}}{2\pi |\partial\omega_{q_x}/\partial q_x|}$$

The resulting expressions for the spontaneous emission probabilities are

$$A_{m \rightarrow n}^{(\mathbf{q})} = \frac{2\pi |\tilde{d}_{mn}|^2 \omega_{mn} |\mathbf{q}|}{\hbar G(L_z, \omega_{mn}) |\partial\omega_q/\partial\mathbf{q}|_{\omega_q=\omega_{mn}}} \quad (31)$$

$$A_{m \rightarrow n}^{(q_x)} = \frac{2\pi |\tilde{d}_{mn}|^2 \omega_{mn}}{\hbar L_y G(L_z, \omega_{mn}) |\partial\omega_{q_x}/\partial q_x|_{\omega_{q_x}=\omega_{mn}}} \quad (32)$$

where  $\omega_{mn}$  is the transition frequency.

In order to use Fermi's golden rule in a cavity, one has to formally introduce the density of states assuming that the modal spectrum is spread near the resonance frequency  $\omega_{mn}$  by the linewidth  $\Delta\omega$  :

$$d\Pi_f = \frac{(\Delta\omega/2\pi)}{(\omega_{mn} - \omega_N)^2 + (\Delta\omega/2)^2} d\omega \quad (33)$$

which results in

$$A_{m \rightarrow n}^{(N)}(\Delta\omega) = \frac{2\pi |\tilde{d}_{mn}|^2 \left( \frac{4\omega_{mn}}{\Delta\omega} \right)}{\hbar L_x L_y G(L_z, \omega_{mn})} \quad (34)$$

Eq. (34) is also valid for a waveguide at a critical frequency, i.e. for  $|\partial\omega_{q_x}/\partial q_x| = 0$ , because such a system is effectively a cavity. In a homogeneous medium, expressions (31), (32) and (34) can be simplified. In this case Eqs. (21) and (28) lead to

$$\frac{|\tilde{d}_{mn}|^2}{G(L_z, \omega_{mn})} = \frac{|d_{mn}|^2}{\frac{L_z}{2\omega_\nu} \left[ \frac{\partial(\omega^2 \varepsilon)}{\partial \omega} \right]_{\omega=\omega_{mn}}}$$

Finally we compare the spontaneous emission probability in a cavity with that in free space. The latter is equal to  $A^{(0)} = \frac{4\omega^3 |d_{mn}|^2 \sqrt{\varepsilon}}{3\hbar c^3}$ . Their ratio is

$$\frac{A_{m \rightarrow n}^{(N)}}{A^{(0)}} \approx \frac{3\pi}{2} \frac{(c/\omega\sqrt{\varepsilon})^3}{L_x L_y L_z} \left( \frac{4\omega_{21}}{\Delta\omega} \right) \quad (35)$$

Note that in Eq. (35) the minimal lateral sizes of an electrodynamic system we consider are  $L_{x,y} = \pi c/\omega\sqrt{\varepsilon}$ , whereas the value of  $L_z$  can be much smaller.

Up to a numerical factor which depends on geometry, Eq. (35) is a widely used expression for the Purcell enhancement of the spontaneous emission. However, Eqs. (31), (32), and (34) do not include the effects of nonradiative relaxation in an ensemble of fermions. Moreover, the above approach does not allow one to determine the line broadening in a cavity in a consistent way. To include all dissipation processes consistently, we use the Heisenberg-Langevin formalism.

## IV. DISSIPATIVE DYNAMICS IN AN ENSEMBLE OF PHOTONS AND ELECTRONS

### A. Heisenberg-Langevin equations for the quasiparticle density operator

Dissipative effects in an open quantum system can be taken into account by adding the relaxation operator  $\hat{R}_{\mu\eta}$  and corresponding Langevin noise operator  $\hat{F}_{\mu\eta}$  to the right-hand side of Eq. (25)[18, 19, 23, 26, 27, 29]. One cannot add dissipation phenomenologically, without including Langevin sources, because this would violate the fundamental commutation relation Eq. (24) [18, 19, 26, 27]. For the simplest model of ‘‘transverse’’ relaxation, when

$$\hat{R}_{\mu \neq \eta} = -\gamma_{\mu\eta} \hat{\rho}_{\mu\eta}. \quad (36)$$

Refs. [18, 19] derived the following expressions for the commutator and correlator of the Langevin noise (for a particular case of a two-level system):

$$\begin{aligned} [\hat{F}_{\mu\eta}(t'), \hat{F}_{\mu\eta}^\dagger(t)] &= (-\gamma_{\mu\eta}(\hat{\rho}_{\eta\eta} - \hat{\rho}_{\mu\mu}) + \hat{R}_{\eta\eta} - \hat{R}_{\mu\mu})\delta(t' - t) \\ \langle \hat{F}_{\mu\eta}^\dagger(t), \hat{F}_{\mu\eta}(t') \rangle &= (2\gamma_{\mu\eta} \langle \hat{\rho}_{\mu\mu} \rangle + \langle \hat{R}_{\mu\mu} \rangle)\delta(t' - t) \end{aligned} \quad (37)$$

where  $\hat{F}_{\mu\eta}^\dagger = \hat{F}_{\eta\mu}$  and the symbol  $\langle \dots \rangle$  means in this case the averaging over both the initial quantum state and the statistics of a dissipative reservoir. The dissipation operator in its simplest form of Eq. (36) implies the absence of any inertia in a dissipative subsystem; that is why the noise operator turns out to be delta-correlated in time. Note

that for degenerate fermion distributions Eqs. (37) are valid if the evolution equation for the density operator includes exchange effects which take care of Pauli blocking.

The nonzero value of the relaxation operator for populations,  $\hat{R}_{\mu\mu} \neq 0$  in Eq. (37) corresponds to the nonequilibrium distribution. A steady-state distribution can be nonequilibrium because of an external pumping. An incoherent pumping generally redistributes populations over many subbands; therefore within the model taking into account a limited number of subbands such a pumping is convenient to introduce as a source  $\hat{J}_{\mu\eta}$  in the evolution equation for the density operator. This way we can assume that there is a generalized relaxation operator  $\hat{R}_{\mu\eta} = \hat{R}_{\mu\eta} + \hat{J}_{\mu\eta}$  on the right-hand side of Eq. (25), and the steady-state (but not necessarily equilibrium) distribution corresponds to the condition  $\langle \hat{R}_{\mu\mu} \rangle = 0$  for all  $\mu$ . Of course, the modification of the relaxation operator causes the noise operator to change. However, within the simplest model of Eq. (36) this does not affect the general form of Eqs. (37). One just needs to keep in mind that the relaxation constants  $\gamma_{\mu\eta}$  and operators  $\hat{R}_{\mu\mu}$  in Eq. (36), (37) contain the contribution from incoherent pumping.

The equation for the density operator can be further simplified if we (i) include only two subbands, i.e.  $m, n = 1, 2$ ; (ii) assume that optical transitions in the interaction Hamiltonian are direct; see Eq. (26). In this case the equation for the off-diagonal density operator elements includes only the elements  $\hat{\rho}_{21\mathbf{k}\mathbf{k}}$  and  $\hat{\rho}_{12\mathbf{k}\mathbf{k}} = \hat{\rho}_{21\mathbf{k}\mathbf{k}}^\dagger$ . Finally, (iii) we assume populations to satisfy  $\hat{R}_{11\mathbf{k}\mathbf{k}} = \hat{R}_{22\mathbf{k}\mathbf{k}} = 0$ . This gives

$$\dot{\hat{\rho}}_{21\mathbf{k}\mathbf{k}} + i\omega_{21}(\mathbf{k})\hat{\rho}_{21\mathbf{k}\mathbf{k}} + \gamma_{21\mathbf{k}\mathbf{k}}\hat{\rho}_{21\mathbf{k}\mathbf{k}} = \frac{i\tilde{d}_{21}}{\hbar} \left( \sum_{\nu} \tilde{D}_{\nu} \hat{c}_{\nu} \right) \cdot (\hat{\rho}_{11\mathbf{k}\mathbf{k}} - \hat{\rho}_{22\mathbf{k}\mathbf{k}}) + \hat{F}_{21\mathbf{k}\mathbf{k}}, \quad (38)$$

where  $\omega_{21}(\mathbf{k}) = \frac{W_{2\mathbf{k}} - W_{1\mathbf{k}}}{\hbar}$ .

As usual, the properties of the Langevin source  $\hat{F}_{21\mathbf{k}\mathbf{k}}(t)$  in Eq. (38) are convenient to express through the properties of its spectral components:  $\hat{F}_{21\mathbf{k}\mathbf{k}}(t) = \int_{-\infty}^{\infty} \hat{F}_{\omega;21\mathbf{k}\mathbf{k}} e^{i\omega t} d\omega$ ,  $\hat{F}_{-\omega;12\mathbf{k}\mathbf{k}} = \hat{F}_{\omega;21\mathbf{k}\mathbf{k}}^\dagger$ . Taking into account that  $\hat{R}_{11\mathbf{k}\mathbf{k}} = \hat{R}_{22\mathbf{k}\mathbf{k}} = 0$ , we can get from Eq. (37) (see also [18, 27])

$$\langle \hat{F}_{\omega;21\mathbf{k}\mathbf{k}}^\dagger \hat{F}_{\omega';21\mathbf{k}\mathbf{k}} \rangle = \frac{\gamma_{21\mathbf{k}\mathbf{k}}}{\pi} n_{2\mathbf{k}} \delta(\omega - \omega'), \quad \langle \hat{F}_{\omega;21\mathbf{k}\mathbf{k}} \hat{F}_{\omega';21\mathbf{k}\mathbf{k}}^\dagger \rangle = \frac{\gamma_{21\mathbf{k}\mathbf{k}}}{\pi} n_{1\mathbf{k}} \delta(\omega - \omega'), \quad (39)$$

where  $n_{1\mathbf{k}} = \langle \hat{\rho}_{11\mathbf{k}\mathbf{k}} \rangle$  and  $n_{2\mathbf{k}} = \langle \hat{\rho}_{22\mathbf{k}\mathbf{k}} \rangle$  are constant populations supported by pumping.

## B. Heisenberg-Langevin equations for field operators

Similarly to relaxation in the medium, relaxation of the EM field gives rise to the noise sources in the equations for field operators [23]. When field absorption by fermions is included, the noise term for the EM field appears due to Langevin noise terms in the density operator equations [18, 19, 27, 30]. Including any additional field absorption unrelated to absorption in the medium should be accompanied by adding Langevin noise terms directly to field equations. We take into account this additional absorption for the  $\nu$ th mode of the field by including phenomenological dissipative operators  $-\Gamma\hat{c}_{\nu}$  and  $-\Gamma\hat{c}_{\nu}^\dagger$  to the right-hand side of the field equations (23). To preserve the commutation relation  $[\hat{c}_{\nu}, \hat{c}_{\nu}^\dagger]$  we need to add the Langevin noise operator  $\hat{L}(t)$ , satisfying the commutation relation  $[\hat{L}(t'), \hat{L}^\dagger(t)] = 2\Gamma\delta(t - t')$ . Its correlator is equal to  $\langle \hat{L}^\dagger(t') \hat{L}(t) \rangle = \Xi \cdot 2\Gamma\delta(t - t')$ , where the parameter  $\Xi$  is determined by a state of a dissipative reservoir. When the latter is in equilibrium, we obtain [23]  $\Xi = (e^{\hbar\omega_{\nu}/T} - 1)^{-1}$ .

Next we take into account that the dissipation of a given  $\nu$ th mode of the EM field could also be due to absorption in metal walls and bulk material unrelated to the active medium. In this case we add the dissipative operators to the right-hand side of Eq. (23),  $-(\Gamma_r + \Gamma_{\sigma})\hat{c}_{\nu}$  and  $-(\Gamma_r + \Gamma_{\sigma})\hat{c}_{\nu}^\dagger$ , together with corresponding Langevin noise terms,  $\hat{L}_r^{(\nu)}$  and  $\hat{L}_{\sigma}^{(\nu)}$ . Here the factor  $\Gamma_r$  describes radiative and diffraction losses out from the cavity and  $\Gamma_{\sigma}$  describes Ohmic losses. Taking into account Eq. (26) for the interaction Hamiltonian, we obtain

$$\dot{\hat{c}}_{\nu} + (i\omega_{\nu} + \Gamma_r + \Gamma_{\sigma}) \cdot \hat{c}_{\nu} = \frac{i\tilde{d}_{12}\tilde{D}_{\nu}^*}{\hbar} \sum_{\mathbf{k}} \hat{\rho}_{21\mathbf{k}\mathbf{k}} + \hat{L}_r^{(\nu)} + \hat{L}_{\sigma}^{(\nu)}. \quad (40)$$

Here the Langevin sources can again be defined through the properties of their spectral components:

$$\begin{aligned}\hat{L}_{r,\sigma}^{(\nu)} &= \int_{-\infty}^{\infty} \hat{L}_{r,\sigma;\omega}^{(\nu)} e^{-i\omega t} d\omega, & \hat{L}_{r,\sigma;-\omega}^{(\nu)} &= \hat{L}_{r,\sigma;\omega}^{(\nu)\dagger}; \\ \langle \hat{L}_{r,\sigma';\omega'}^{(\nu)\dagger} \hat{L}_{r,\sigma;\omega}^{(\nu)} \rangle &= n_{T_{r,\sigma}}(\omega_\nu) \frac{\Gamma_{r,\sigma} \delta_{\nu\nu'}}{\pi} \delta(\omega - \omega'), \\ \langle \hat{L}_{r,\sigma;\omega}^{(\nu)} \hat{L}_{r,\sigma';\omega'}^{(\nu)\dagger} \rangle &= [n_{T_{r,\sigma}}(\omega_\nu) + 1] \frac{\Gamma_{r,\sigma} \delta_{\nu\nu'}}{\pi} \delta(\omega - \omega'),\end{aligned}\quad (41)$$

where  $n_{T_{r,\sigma}}(\omega_\nu) = \frac{1}{e^{\hbar\omega_\nu/T_{r,\sigma}} - 1}$  and  $T_{r,\sigma}$  are the temperature of the ambient space which controls radiative losses and the bulk material inside the cavity. The presence  $\delta_{\nu\nu'}$  in Eq. (41) corresponds to the Langevin sources that are  $\delta$ -correlated not only in space but also in time [18, 27].

### C. Spontaneous emission from an ensemble of nonequilibrium fermions in a single-mode cavity

If we assume the populations to be given, the Heisenberg equations for the off-diagonal elements of the density operator can be averaged over the original state of quasiparticles. After averaging, the off-diagonal elements will depend on the field operators, noise operators, and populations  $n_{m\mathbf{k}}$ . The operators of populations  $\hat{\rho}_{mm\mathbf{k}\mathbf{k}}$  in Eq. (38) will be replaced by c-numbers:  $\hat{\rho}_{mm\mathbf{k}\mathbf{k}} \implies n_{m\mathbf{k}}$ ; see [25, 27].

The structure of Eqs. (38) and (40) suggests the substitution  $\hat{c}_\nu = \hat{c}_{0\nu}(t)e^{-i\omega_\nu t}$ ,  $\hat{c}_\nu^\dagger = \hat{c}_{0\nu}^\dagger(t)e^{+i\omega_\nu t}$ . Here  $\hat{c}_{0\nu}(t)$  and  $\hat{c}_{0\nu}^\dagger(t)$  are ‘‘slow’’ amplitudes in the following sense:  $\langle \dot{\hat{c}}_{0\nu} \rangle \ll \omega_\nu \langle \hat{c}_{0\nu} \rangle$ ; see [25]. Neglecting any inhomogeneous broadening of the resonance line, a steady-state solution of Eq. (38) for a single-mode cavity is

$$\hat{\rho}_{21\mathbf{k}\mathbf{k}} \approx \frac{id_{21}\tilde{D}_\nu}{\hbar} \frac{\hat{c}_{0\nu} e^{-i\omega_\nu t} (n_{1\mathbf{k}} - n_{2\mathbf{k}})}{i(\omega_{21} - \omega_\nu) + \gamma_{21\mathbf{k}\mathbf{k}}} + \int_{-\infty}^{\infty} \frac{\hat{F}_{\omega;21\mathbf{k}\mathbf{k}} e^{-i\omega t} d\omega}{i(\omega_{21} - \omega) + \gamma_{21\mathbf{k}\mathbf{k}}}. \quad (42)$$

Substituting Eq. (42) into Eq. (40) we obtain

$$\begin{aligned}\dot{\hat{c}}_{0\nu} + (\Gamma_r + \Gamma_\sigma + i\delta\omega + \gamma)\hat{c}_{0\nu} &= \frac{id_{21}\tilde{D}_\nu^*}{\hbar} \sum_{\mathbf{k}} \int_{-\infty}^{\infty} \frac{\hat{F}_{\omega;21\mathbf{k}\mathbf{k}} e^{-i(\omega - \omega_\nu)t} d\omega}{i(\omega_{21} - \omega) + \gamma_{21\mathbf{k}\mathbf{k}}} \\ &+ \int_{-\infty}^{\infty} \hat{L}_{r\omega'}^{(\nu)} e^{-i(\omega' - \omega_\nu)t} d\omega' + \int_{-\infty}^{\infty} \hat{L}_{\sigma\omega''}^{(\nu)} e^{-i(\omega'' - \omega_\nu)t} d\omega''\end{aligned}\quad (43)$$

where

$$\delta\omega = \Omega^2 \text{Re} \sum_{\mathbf{k}} \frac{n_{1\mathbf{k}} - n_{2\mathbf{k}}}{(\omega_{21} - \omega_\nu) - i\gamma_{21\mathbf{k}\mathbf{k}}}, \quad \gamma = \Omega^2 \text{Im} \sum_{\mathbf{k}} \frac{n_{1\mathbf{k}} - n_{2\mathbf{k}}}{(\omega_{21} - \omega_\nu) - i\gamma_{21\mathbf{k}\mathbf{k}}}, \quad (44)$$

$$\Omega^2 = \frac{|\tilde{d}_{21}|^2 |\tilde{D}_\nu|^2}{\hbar^2} = \frac{|\tilde{d}_{21}|^2 2\pi\omega_\nu}{\hbar L_x L_y G(L_z, \omega_\nu)}. \quad (45)$$

The frequency shift  $\delta\omega$  of the ‘‘cold’’ cavity mode is due to the optical transitions between electron states in a QW. We can redefine the cavity mode frequency assuming that the effect of electrons has been included in  $\omega_\nu$  from the very beginning (a ‘‘hot’’ cavity mode). The decay rate  $\gamma$  describes absorption by electrons; the population inversion corresponds to  $\gamma < 0$ . If  $(\gamma + \Gamma_r + \Gamma_\sigma) < 0$  the instability develops and the field grows with time; we don’t consider this case here.

The steady-state solution of Eq. (43) has the form

$$\begin{aligned}\hat{c}_{0\nu} &= \frac{id_{21}\tilde{D}_\nu^*}{\hbar} \sum_{\mathbf{k}} \int_{-\infty}^{\infty} \frac{\hat{F}_{\omega;21\mathbf{k}\mathbf{k}} e^{-i(\omega - \omega_\nu)t} d\omega}{[i(\omega_\nu - \omega) + \Gamma_r + \Gamma_\sigma + \gamma] \times [i(\omega_{21} - \omega) + \gamma_{21\mathbf{k}\mathbf{k}}]} \\ &+ \int_{-\infty}^{\infty} \frac{\hat{L}_{r\omega'}^{(\nu)} e^{-i(\omega' - \omega_\nu)t} d\omega'}{[i(\omega_\nu - \omega') + \Gamma_r + \Gamma_\sigma + \gamma]} + \int_{-\infty}^{\infty} \frac{\hat{L}_{\sigma\omega''}^{(\nu)} e^{-i(\omega'' - \omega_\nu)t} d\omega''}{[i(\omega_\nu - \omega'') + \Gamma_r + \Gamma_\sigma + \gamma]}.\end{aligned}\quad (46)$$

Next, we use the Hermitian conjugate of Eq. (46) to find the value of  $\langle \hat{c}_{0\nu}^\dagger \hat{c}_{0\nu} \rangle$ , assuming that the statistics of noise operators  $\hat{F}_{21\mathbf{k}\mathbf{k}}(t)$ ,  $\hat{L}_r^{(\nu)}(t)$  and  $\hat{L}_\sigma^{(\nu)}(t)$  are independent from each other. Using Eqs. (39) and (41) we obtain

$$\langle \hat{c}_{0\nu}^\dagger \hat{c}_{0\nu} \rangle = \Omega^2 \sum_{\mathbf{k}} \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \frac{\gamma_{21\mathbf{k}\mathbf{k}} n_{2\mathbf{k}}}{[(\omega_\nu - \omega)^2 + (\Gamma_r + \Gamma_\sigma + \gamma)^2] \times [(\omega_{21} - \omega)^2 + \gamma_{21\mathbf{k}\mathbf{k}}^2]} + \frac{\Gamma_r}{\Gamma_r + \Gamma_\sigma + \gamma} n_{T_r}(\omega_\nu) + \frac{\Gamma_\sigma}{\Gamma_r + \Gamma_\sigma + \gamma} n_{T_\sigma}(\omega_\nu). \quad (47)$$

For simplicity, we neglect the last two terms in Eq. (47) which describe the contribution of the EM background of a surrounding medium and thermal radiation of the material inside a cavity. The power emitted by electrons into the outside space is  $P = 2\Gamma_r \times \hbar\omega_\nu \times \langle \hat{c}_{0\nu}^\dagger \hat{c}_{0\nu} \rangle$ :

$$P = \hbar\omega_\nu \Omega^2 \sum_{\mathbf{k}} \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \frac{2\Gamma_r \gamma_{21\mathbf{k}\mathbf{k}} n_{2\mathbf{k}}}{[(\omega_\nu - \omega)^2 + (\Gamma_r + \Gamma_\sigma + \gamma)^2] \times [(\omega_{21} - \omega)^2 + \gamma_{21\mathbf{k}\mathbf{k}}^2]}. \quad (48)$$

Equation (48) for the spontaneous emission power is the main result of this section. It has two obvious limiting cases:

(i) The transition line is much narrower than the cavity resonance:  $\Gamma_r + \Gamma_\sigma + \gamma \gg \gamma_{21\mathbf{k}\mathbf{k}}$ . In this case we can get from Eq. (48)

$$P = \hbar\omega_\nu [A_{2 \rightarrow 1}^{(N)}(\Delta\omega)_{\Delta\omega = \Delta\omega_{eff}^{(1)}}] \cdot \frac{\Gamma_r}{\Gamma_r + \Gamma_\sigma + \gamma} \cdot \frac{\Gamma_r + \Gamma_\sigma + \gamma}{(\omega_\nu - \omega_{21})^2 + (\Gamma_r + \Gamma_\sigma + \gamma)^2} \cdot \sum_{\mathbf{k}} n_{2\mathbf{k}}, \quad (49)$$

where  $A_{2 \rightarrow 1}^{(N)}(\Delta\omega)$  is the probability of the spontaneous emission in a cavity given by Eq. (34) and  $\Delta\omega_{eff}^{(1)} = 2(\Gamma_r + \Gamma_\sigma + \gamma)$ . The second factor in Eq. (49) determines the fraction of the radiation which escaped outside. The third factor is due to a position of the narrow transition line within a broader cavity mode line. The last factor is a number of radiating particles:  $\sum_{\mathbf{k}} n_{2\mathbf{k}} \Rightarrow \frac{S}{(2\pi)^2} \int n_{2\mathbf{k}} d^2\mathbf{k}$ .

(ii) The transition line is much wider than the cavity resonance:  $\Gamma_r + \Gamma_\sigma + \gamma \ll \gamma_{21\mathbf{k}\mathbf{k}}$ . In this case

$$P = \hbar\omega_\nu [A_{2 \rightarrow 1}^{(N)}(\Delta\omega)_{\Delta\omega = \Delta\omega_{eff}^{(2)}}] \cdot \frac{\Gamma_r}{\Gamma_r + \Gamma_\sigma + \gamma} \cdot \sum_{\mathbf{k}} \frac{\langle \gamma_{21} \rangle \gamma_{21\mathbf{k}\mathbf{k}} n_{2\mathbf{k}}}{(\omega_\nu - \omega_{21})^2 + \gamma_{21\mathbf{k}\mathbf{k}}^2}. \quad (50)$$

Instead of the cavity linewidth  $2(\Gamma_r + \Gamma_\sigma + \gamma)$  Eq. (50) contains the homogeneous linewidth  $\Delta\omega_{eff}^{(2)} = \langle \gamma_{21} \rangle$  where the right-hand side is an average value of  $\gamma_{21\mathbf{k}\mathbf{k}}$ . Now the third factor is due to a position of the narrow cavity mode line within a broader transition line. Therefore, the effective quality factor is determined by greater of the two values,  $\Gamma_r + \Gamma_\sigma + \gamma$  or  $\langle \gamma_{21} \rangle$ . The spontaneous emission efficiency is proportional to the factor  $\frac{\Gamma_r}{\Gamma_r + \Gamma_\sigma + \gamma}$ , where  $\gamma$  is the decay rate of the field due to absorption by electrons. Since  $\gamma$  depends on the electron density, the spontaneous emission efficiency per particle also depends on their density.

One can further simplify Eq. (48) if relaxation constants  $\gamma_{21\mathbf{k}\mathbf{k}}$  do not depend on  $\mathbf{k}$ , i.e.  $\gamma_{21\mathbf{k}\mathbf{k}} \equiv \gamma_{21}$ :

$$P = \hbar\omega_\nu \Omega^2 \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \frac{2\Gamma_r \gamma_{21}}{[(\omega_\nu - \omega)^2 + (\Gamma_r + \Gamma_\sigma + \gamma)^2] \times [(\omega_{21} - \omega)^2 + \gamma_{21}^2]} \sum_{\mathbf{k}} n_{2\mathbf{k}}. \quad (51)$$

Here  $\gamma$  is defined by Eq. (44); for  $\gamma_{21\mathbf{k}\mathbf{k}} \equiv \gamma_{21}$  it becomes

$$\gamma = \Omega^2 \frac{\gamma_{21}}{(\omega_{21} - \omega_\nu)^2 + \gamma_{21}^2} \sum_{\mathbf{k}} (n_{1\mathbf{k}} - n_{2\mathbf{k}}), \quad (52)$$

where  $\Omega^2$  is given by Eq. (45). Using Eq. (34), one can rewrite Eq. (51) as

$$P = \hbar\omega_\nu A_{2 \rightarrow 1}^{(N)} \sum_{\mathbf{k}} n_{2\mathbf{k}}, \quad (53)$$

where

$$A_{2 \rightarrow 1}^{(N)} = \frac{2\pi |\tilde{d}_{21}|^2 \left( \frac{4\omega_{21}}{\Delta\omega_{eff}} \right)}{\hbar L_x L_y G(L_z, \omega_\nu)} \quad (54)$$

and

$$\frac{1}{\Delta\omega_{eff}} = \int_{-\infty}^{\infty} \frac{d\omega}{4\pi} \frac{2\Gamma_r\gamma_{21}}{[(\omega_\nu - \omega)^2 + (\Gamma_r + \Gamma_\sigma + \gamma)^2] \times [(\omega_{21} - \omega)^2 + \gamma_{21}^2]}. \quad (55)$$

For a cavity filled with a uniform and dispersionless medium with dielectric constant  $\varepsilon$  one can further simplify Eq. (53) as

$$P = \left[ \hbar\omega_\nu A^{(0)} \sum_{\mathbf{k}} n_{2\mathbf{k}} \right] \left[ \frac{6}{\pi^2} \frac{(\lambda/2\sqrt{\varepsilon})^3}{L_x L_y L_z} \right] Q_{eff}, \quad (56)$$

Where  $A^{(0)} = \frac{4\omega^3 |d_{21}|^2 \sqrt{\varepsilon}}{3\hbar c^3}$  is the spontaneous emission rate into free space filled with dielectric medium  $\varepsilon$  and  $Q_{eff} = \frac{\omega_{21}}{\Delta\omega_{eff}}$  is the effective quality factor. The term in the first brackets on the rhs of Eq. (56) is the power of spontaneous emission into free space; the term in the second brackets is the geometric enhancement due to a subwavelength cavity.

The integral in Eq. (55) is a product of two Lorentzians which can be easily evaluated analytically but is a bit cumbersome. Assuming for simplicity exact resonance between the transition frequency and the cavity resonance,  $\omega_\nu = \omega_{21}$ , we obtain

$$Q_{eff} = \frac{\omega_{21}\Gamma_r}{2(\Gamma_r + \Gamma_\sigma + \gamma)(\gamma_{21} + \Gamma_r + \Gamma_\sigma + \gamma)} \rightarrow \frac{\omega_{21}}{2(\gamma_{21} + \Gamma_r)}, \quad (57)$$

where the last expression is in the limit  $\Gamma_r \gg \Gamma_\sigma + \gamma$ .

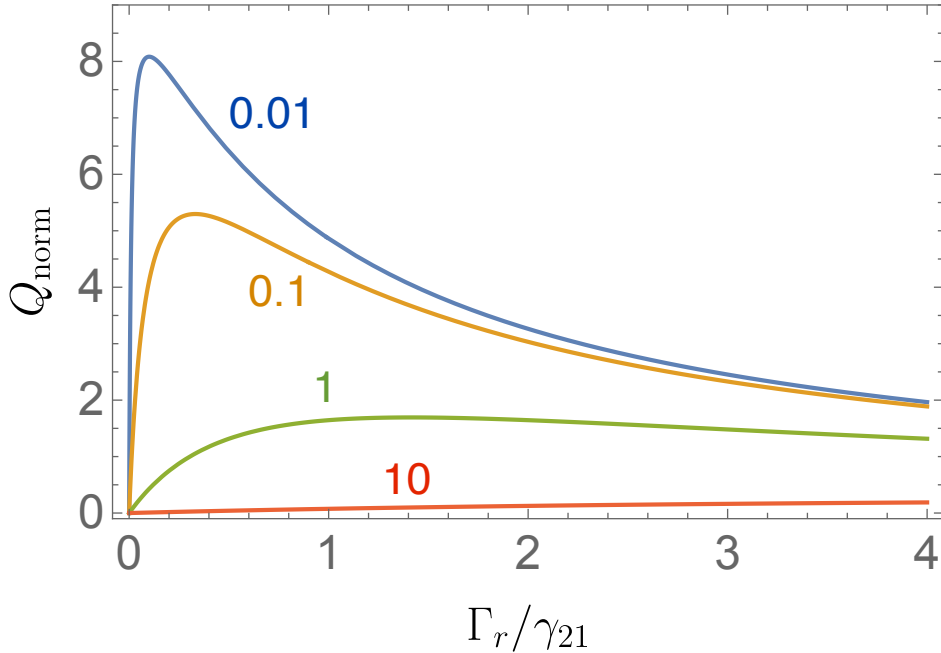


FIG. 2. The normalized effective Q-factor as a function of the normalized cavity linewidth  $\Gamma_r/\gamma_{21}$  at exact resonance  $\omega_{21} = \omega_\nu$ . Four curves correspond to four different values of the total intracavity absorption rate  $(\Gamma_\sigma + \gamma)/\gamma_{21}$ : 0.01, 0.1, 1, and 10, from top to bottom curve.

For a fixed transition linewidth  $\gamma_{21}$  we normalize  $Q_{eff}$  by the Q-factor of the radiative transition  $\frac{\omega_{21}}{2\gamma_{21}}$  and plot the normalized Q-factor  $Q_{norm} = \frac{2\gamma_{21}}{\Delta\omega_{eff}}$  as a function of the cavity linewidth  $\Gamma_r$  for different values of the total normalized intracavity absorption rate  $(\Gamma_\sigma + \gamma)/\gamma_{21}$ ; see Fig. 2. As shown in Fig. 2, the total magnitude of the intracavity absorption rate should be kept below the total linewidth  $\gamma_{21}$  of the emission line. Figure 2 also shows that

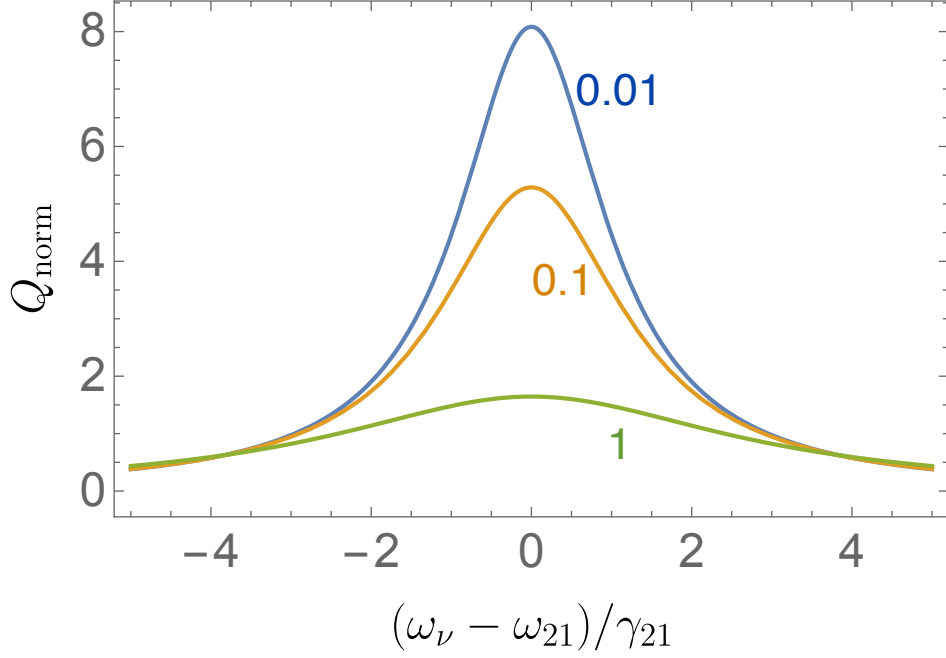


FIG. 3. The normalized effective Q-factor as a function of frequency detuning at  $\Gamma_r = \gamma_{21}$ . Three curves correspond to three different values of the total intracavity absorption rate  $(\Gamma_\sigma + \gamma)/\gamma_{21}$ : 0.01, 0.1, and 1, from top to bottom curve. They are plotted for the value of the normalized cavity linewidth  $\Gamma_r/\gamma_{21} = 0.1, 0.3,$  and 1, respectively, which correspond to the maximum  $Q_{eff}$  in Fig. 2.

it makes no sense to increase the Q-factor of the cavity mode  $\frac{\omega_\nu}{2\Gamma_r}$  beyond the value of  $\Gamma_r$  corresponding to the peak value of the effective Q-factor  $Q_{eff}$ . For smaller values of  $\Gamma_r$  the intracavity quantum efficiency will stay roughly the same, limited by the dissipation rate  $\gamma_{21}$  of the optical polarization, whereas the radiation power outcoupled from the cavity reduces  $\propto \Gamma_r$ . The effective Q-factor quickly drops down with detuning of the cavity mode from the emission line; see Fig. 3.

For mid-infrared intersubband transitions in multiple QW nanocavities at  $\hbar\omega_{21} \sim 100 - 200$  meV and full linewidth  $2\gamma_{21} = 10$  meV [31] the maximum  $Q_{eff} \sim 50 - 100$  and the geometric enhancement in Eq. (56) can add another factor of 10 – 100. For THz intersubband transitions  $Q_{eff}$  is similar whereas the geometric enhancement is a factor of 10 higher. For a near-infrared interband transition in semiconductor QWs the frequency is  $\sim 5 - 10$  times higher, but the linewidth is 2 – 3 times higher as well, so  $Q_{eff}$  can be about 100-300. This example also suggests that an optimal radiative loss from a cavity (or a cavity mode linewidth) for semiconductor 2D emitters should be of the order of 5-10 meV. 2D semiconductors such as MoS<sub>2</sub> have excitonic emission lines that are quite broad, up to 50-100 meV. They are ideally suited for integration with plasmonic nanocavities which have a relatively low Q-factor but a very small effective mode volume. As an example, Ref. [34] reports a 2000-fold enhancement in the photoluminescence intensity from MoS<sub>2</sub> monolayer in a plasmonic nanocavity formed by a gold substrate and a patch silver nanoantenna. Since the emission line of MoS<sub>2</sub> was so broad (about 30 nm at 660 nm wavelength), according to Eqs. (56), (57) the authors made an optimal choice of using a plasmonic nanocavity with strong radiative outcoupling and comparably broad nanocavity modes, but with an ultras-small effective mode volume of  $\sim 10^{-3}(\lambda/\sqrt{\epsilon})^3$ .

In conclusion, using consistent Heisenberg-Langevin approach we derived general analytic formulas describing the spontaneous emission of 2D emitters placed in plane-parallel subwavelength cavities or waveguides. We found that a significant enhancement of the outcoupled spontaneous emission and quantum efficiency of semiconductor quantum devices can be achieved for realistic device parameters. The present formalism can be extended to the nonlinear optical processes in 2D subwavelength cavities [7].

## Purcell enhancement of the photon parametric decay in two-dimensional nonlinear materials

Enhancement of the radiative processes due to the localization of emitters in a subwavelength cavity (so-called Purcell enhancement [9]) is a fundamental cavity quantum electrodynamics (QED) effect of great importance for numerous applications. The bulk of the research has been focused on exploring the enhancement of spontaneous emission in various compact radiation sources from single quantum emitters to LEDs and nanolasers. The nonlinear optics has received relatively less attention; however, recent advancements in strong light localization using sub-wavelength cavities, photonic crystals, metamaterials, and metasurfaces enabled the nonlinear optics in ultrasmall volumes and at relatively low power levels; see e.g. [31, 35–42] and references therein. The rise of 2D materials with atomic monolayer thickness and excellent nonlinear optical properties, such as graphene [43–46] and transition metal dichalcogenide monolayers [47, 48] has enabled quasi-2D cavities and waveguides only a few nm thick [49, 50]. These advances create new exciting opportunities for ultracompact nonlinear optical devices, but also raise important issues of the correct description of quantum fields in systems with strong dissipation both in a macroscopic ensemble of fermionic emitters (e.g. a 2D semiconductor or monolayer graphene, or a 2D electron gas in a quantum well) and for the electromagnetic (EM) field in a cavity.

One important application for Purcell-enhanced nonlinear optics is compact systems for generation of squeezed and entangled photon states as a result of parametric decay. Such systems are inevitably lossy. The general approach to introducing dissipation and corresponding fluctuations has been known for a long time and is based on the Heisenberg-Langevin formalism; e.g. [19, 23, 27, 29, 51]. Its generalization to systems far from equilibrium, with arbitrary dissipation mechanisms and arbitrary photon density of states is nontrivial; see e.g. [18, 26]. Recent work [42] considered the process of spontaneous parametric down-conversion in hyperbolic metamaterials, in which the EM field dissipation and fluctuations are due to an equilibrium thermal reservoir. In the present paper we consider both spontaneous and stimulated parametric decay (parametric instability) in a generic quasi-2D subwavelength cavity, taking into account dissipation and fluctuations both due to absorption in the intracavity material and due to in/outcoupling of the intracavity EM field with the outside world.

We generalize the properties of Langevin noise sources known for a single mode of a quantized field (e.g. [6, 23, 51]) to an ensemble of coupled field oscillators. We derive the properties of the Langevin sources needed to conserve their commutation relations and show that they are not affected by a more complicated dynamics of coupled Heisenberg field operators; moreover, this statement does not depend on any specific microscopic model of a dissipative reservoir. We are able to derive closed-form analytic results for the spontaneous parametric signal, the parametric gain, and the threshold for parametric instability. These expressions include the contributions from all relevant dissipation and fluctuation effects such as absorption and radiation losses, interaction with thermal and zero-point fluctuations, parametric amplification of thermal noise and seed photons at the signal frequency, etc.; see e.g. Eqs. (89), (92) below.

### V. PARAMETRIC DECAY IN A CONSERVATIVE SYSTEM

Consider three cavity modes with frequencies related by the energy conservation in the parametric decay process:

$$\omega_p = \omega_s + \omega_i. \quad (58)$$

Here the pumping at frequency  $\omega_p$  will be considered a classical coherent field,

$$\mathbf{E}_p = \mathbf{E}_p(\mathbf{r})e^{-i\omega_p t} + C.C. \quad (59)$$

The field at signal and idler frequencies,  $\omega_s$  and  $\omega_i$ , will be the quantum field described by the operator

$$\hat{\mathbf{E}} = \sum_{\nu=s,i} [\mathbf{E}_\nu(\mathbf{r})\hat{c}_\nu + \mathbf{E}_\nu^*(\mathbf{r})\hat{c}_\nu^\dagger], \quad (60)$$

where  $\hat{c}_\nu$  and  $\hat{c}_\nu^\dagger$  are boson annihilation and creation operators. The functions  $\mathbf{E}_{p,s,i}(\mathbf{r})$  in Eqs. (59) and (60) are determined by the spatial structure of the cavity modes. The normalization of functions  $\mathbf{E}_\nu(\mathbf{r})$  needs to be chosen in such a way that the commutation relation for boson operators  $\hat{c}_\nu$  and  $\hat{c}_\nu^\dagger$  have a standard form  $[\hat{c}_\nu, \hat{c}_{\nu'}^\dagger] = \delta_{\nu\nu'}$ . Following [22, 25, 33], one can obtain

$$\int_V \mathbf{E}_{\nu j}^*(\mathbf{r}) \frac{1}{2\omega_\nu} \left[ \frac{\partial (\omega^2 \varepsilon_{jk}(\omega, \mathbf{r}))}{\partial \omega} \right]_{\omega=\omega_\nu} \mathbf{E}_{\nu k}(\mathbf{r}) d^3r = 2\pi\hbar\omega_\nu, \quad (61)$$

where  $\omega_\nu$  is the eigenfrequency of a cavity mode,  $E_{\nu j}(\mathbf{r})$  is a Cartesian component of the vector field  $\mathbf{E}_\nu(\mathbf{r})$ ,  $\varepsilon_{jk}(\omega, \mathbf{r})$  is the dielectric tensor, and  $V$  is a cavity volume (a quantization volume).

Consider a 3D cavity filled with an isotropic dielectric medium, as sketched in Fig. 4. The cavity thickness in  $z$ -direction is much smaller than wavelength:  $L_z \ll c/\sqrt{\bar{\varepsilon}}\omega$ , where  $\bar{\varepsilon}$  is a typical (average) value of the dielectric constant of the filling. As was shown in [6], if the dielectric filling consists of plane-parallel layers, i.e.  $\varepsilon = \varepsilon(z)$ , the structure of the cavity eigenmodes is quasi-electrostatic along the  $z$ -axis, i.e.  $E_z\varepsilon(z) \approx \text{const}$ ,  $E_{x,y} \ll E_z$ . In this case the field of a cavity mode can be written as

$$\mathbf{E}_{p,s,i}(\mathbf{r}) \approx z_0 D_{p,s,i} \frac{\zeta_{p,s,i}(x,y)}{\varepsilon(\omega_{p,s,i}, z)}, \quad (62)$$

where the constants  $D_{p,s,i}$  are coordinate-independent amplitudes of the electric induction. To find the functions  $\zeta_{p,s,i}(x,y)$  we solve the wave equation

$$\nabla \cdot (\nabla \cdot \mathbf{E}_{p,s,i}) - \nabla^2 \mathbf{E}_{p,s,i} - \frac{\omega_{p,s,i}^2}{c^2} \varepsilon(\omega_{p,s,i}, z) \mathbf{E}_{p,s,i} = 0.$$

Consider a  $z$ -component of this equation,

$$\frac{\partial}{\partial z} \left( \frac{\partial E_{(p,s,i)x}}{\partial x} + \frac{\partial E_{(p,s,i)y}}{\partial y} \right) = \left[ \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\omega_{p,s,i}^2}{c^2} \varepsilon(\omega_{p,s,i}, z) \right] E_{(p,s,i)z}. \quad (63)$$

Following the procedure described in [6], we integrate Eq. (63) over  $\int_{-L_z/2}^{L_z/2} dz$  taking into account the boundary conditions on the metal planes of the cavity,  $E_{x,y}(\pm L_z/2) = 0$ . Then we substitute Eq. (62) into the result of integration, which gives

$$\left[ \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\omega_{p,s,i}^2}{c^2 \frac{1}{L_z} \int_{-L_z/2}^{L_z/2} \frac{1}{\varepsilon(\omega_{p,s,i}, z)} dz} \right] \zeta_{p,s,i} = 0. \quad (64)$$

The solution to Eq. (64) with zero boundary conditions at the edges of the cavity determines eigenfrequencies and the structure of the eigenmodes for a quasi-2D cavity with an arbitrary shape in the  $(x,y)$ -plane.

Similar equations can be derived if one simply utilizes jumps of the dielectric constants on the sides instead of metal coating. Even without any jump in the dielectric constants, an open end of a thin waveguide with vertical size much smaller than wavelength is a good reflector and therefore any radiation losses through the facets are small and are not affecting the mode spatial structure significantly.

The expression for the constants  $D_{p,s,i}$  for quantized fields can be obtained from the general equation Eq. (61) (see also [6]),

$$|D_\nu|^2 = \frac{2\pi\hbar\omega_\nu}{\int_S |\zeta_\nu(x,y)|^2 d^2r \frac{1}{2\omega_\nu} \int_{-L_z/2}^{L_z/2} \left[ \frac{\partial(\omega^2\varepsilon(\omega,z))}{\partial\omega} \right]_{\omega=\omega_\nu} dz}, \quad (65)$$

where  $\nu = p, s, i$ . For a simple case of a rectangular-shaped cavity, when  $S = L_x \times L_y$ , where  $L_x$  and  $L_y$  are the cavity dimensions along  $x$  and  $y$  directions, it is easy to obtain useful analytic expressions for the modal spatial structure and frequencies. For eigenmodes with one half-wavelength along the  $y$ -axis and  $N$  half-wavelengths along the  $x$ -axis, we obtain the modal profile

$$\zeta_\nu = \cos\left(\frac{\pi y}{L_y}\right) \times \begin{cases} \cos\left(\frac{N_{\text{odd}}^{(\nu)}\pi x}{L_x}\right) \\ \sin\left(\frac{N_{\text{even}}^{(\nu)}\pi x}{L_x}\right) \end{cases}, \quad (66)$$

$$\int_S |\zeta_\nu(x,y)|^2 d^2r = \frac{S}{4}. \quad (67)$$

The eigenfrequencies are given by

$$\left(\frac{N_\nu\pi}{L_x}\right)^2 + \left(\frac{\pi}{L_y}\right)^2 = \frac{\omega_\nu^2}{c^2} \frac{L_z}{\int_{-L_z/2}^{L_z/2} \frac{1}{\varepsilon(\omega_\nu, z)} dz}. \quad (68)$$

For a particular case of a uniform dielectric constant, Eqs. (66) and (68) are exact, i.e. they do not require an assumption of a quasielectrostatic field structure.

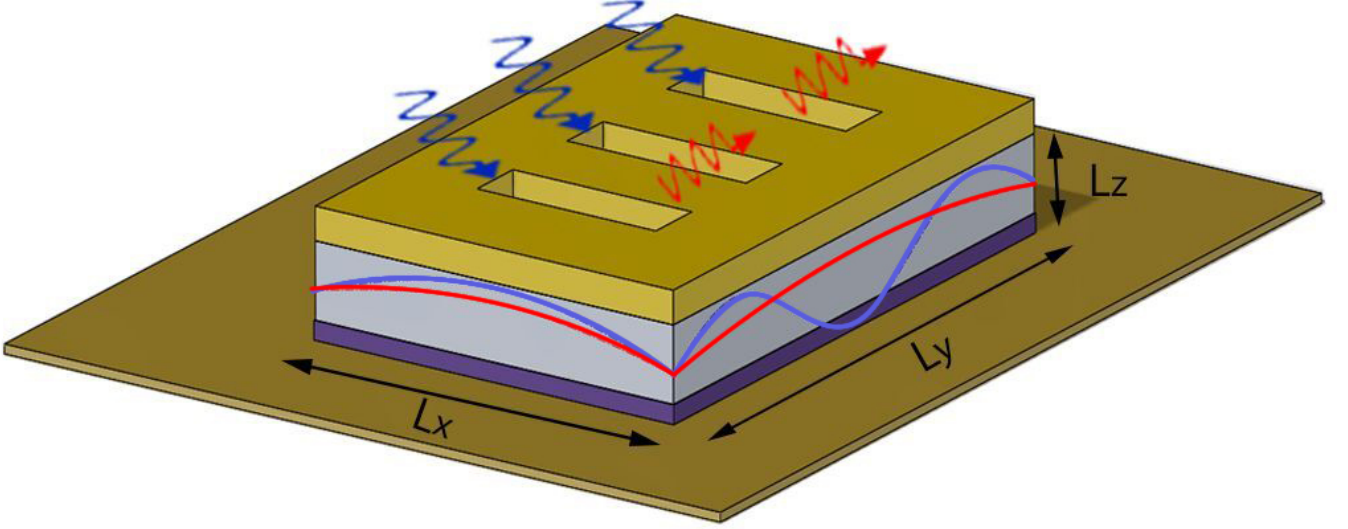


FIG. 4. A sketch of a nanocavity with thickness  $L_z$  much smaller than wavelength. The profiles of the electric fields of the TE<sub>013</sub> pump mode (blue) and TE<sub>011</sub> signal and idler modes (red) are sketched on the sides. Dark blue layer indicates a 2D nonlinear material; light blue layer is a cavity filling. Top and bottom gold layers are metal plates. The radiation can be in/outcoupled through the gratings or cavity edges.

We assume that a 2D electron system with the second-order nonlinear susceptibility is positioned in the cavity. The material can be a quantum well (QW), a 2D semiconductor, or graphene, which has a strong second-order nonlinearity beyond electric-dipole approximation, despite being centrosymmetric [1, 2]. The second-order nonlinearity gives rise to the nonlinear polarization at signal and idler frequencies. The excitation equations for the cavity modes derived from the operator-valued Maxwell's equations [22] take the form

$$\hat{c}_\nu + i\omega_\nu \hat{c}_\nu = -\frac{i}{\hbar\omega_\nu^2} \int_V \ddot{\hat{\mathbf{P}}}_{NL}(\mathbf{r}, t) \mathbf{E}_\nu^*(\mathbf{r}) d^3r. \quad (69)$$

The nonlinear polarization  $\hat{\mathbf{P}}_{NL}(\mathbf{r}, t)$  should be determined for a given electron system; in general, it has a nonuniform distribution over the cavity cross-section. However, it is obvious from Eq. (69) that only the integral over the nonlinear polarization matters. Therefore it is convenient to consider a nonlinear layer with uniform dielectric constant  $\varepsilon_{QW}(\omega_{p,s,i}) = \varepsilon_{p,s,i}$  and uniform second-order nonlinear susceptibilities  $\chi^{(2)}(\omega_s = \omega_p - \omega_i) = \chi_s^{(2)}$ ,  $\chi^{(2)}(\omega_i = \omega_p - \omega_s) = \chi_i^{(2)}$ ,  $\chi^{(2)}(\omega_p = \omega_s + \omega_i) = \chi_p^{(2)}$ . For a general case of a nonuniform layer the above quantities can be considered as parameters obtained as a result of integration in Eq. (69).

For a uniform layer the nonlinear polarization can be expressed as

$$\hat{\mathbf{P}}_{NL} = \mathbf{z}_0 \zeta_p(x, y) [\zeta_i(x, y) \chi_s^{(2)} E_p E_i^* e^{-i\omega_p t} \hat{c}_i^\dagger + \zeta_s(x, y) \chi_i^{(2)} E_p E_s^* e^{-i\omega_p t} \hat{c}_s^\dagger] + H.C.; \quad (70)$$

where  $\hat{c}_s^\dagger \propto e^{i\omega_s t}$ ,  $\hat{c}_i^\dagger \propto e^{i\omega_i t}$ ,

$$|E_{s,i}|^2 = \frac{8\pi\hbar\omega_{s,i}}{L_x L_y L_z \frac{1}{2\omega_{s,i}} \left[ \frac{\partial(\omega^2 \epsilon)}{\partial\omega} \right]_{\omega=\omega_{s,i}}}, \quad (71)$$

and  $E_p = \text{const.}$  In Eq. (71) we assumed a rectangular cavity shape for simplicity.

If the nonlinearity is non-dissipative, the nonlinear susceptibilities satisfy the symmetry properties which ensure that Manley-Rowe relationships are satisfied in a conservative system [52, 53].

$$\chi_s^{(2)} = \chi_i^{(2)} = \chi_p^{(2)*} = \chi^{(2)}. \quad (72)$$

Using the rotating wave approximation, Eqs. (69) and (70) give the following parametrically coupled equations for a given classical pumping:

$$\dot{\hat{c}}_s + i\omega_s \hat{c}_s + \varsigma e^{-i\omega_p t} \hat{c}_i^\dagger = 0, \quad \dot{\hat{c}}_i^\dagger - i\omega_i \hat{c}_i^\dagger + \varsigma^* e^{+i\omega_p t} \hat{c}_s = 0 \quad (73)$$

where

$$\varsigma = -\frac{i}{\hbar} \chi^{(2)} l E_p E_i^* E_s^* \times J, \quad (74)$$

$$J = \int_{L_x \times L_y} \zeta_p(x, y) \zeta_i(x, y) \zeta_s(x, y) d^2 r \quad (75)$$

is a modal overlap factor and  $l$  is the thickness of the nonlinear layer in  $z$  direction.

Equations (73) are Heisenberg equations for the effective Hamiltonian

$$\hat{H} = \hbar\omega_s \left( \hat{c}_s^\dagger \hat{c}_s + \frac{1}{2} \right) + \hbar\omega_i \left( \hat{c}_i^\dagger \hat{c}_i + \frac{1}{2} \right) - i\hbar\varsigma e^{-i\omega_p t} \hat{c}_s^\dagger \hat{c}_i^\dagger + i\hbar\varsigma^* e^{i\omega_p t} \hat{c}_s \hat{c}_i. \quad (76)$$

For a parametric decay of a pump photon into two identical photons, when

$$\omega_p = 2\omega_s \quad (77)$$

we arrive at the Hamiltonian

$$\hat{H} = \hbar\omega_s \left( \hat{c}_s^\dagger \hat{c}_s + \frac{1}{2} \right) - i\hbar \frac{\varsigma}{2} e^{-i\omega_p t} \hat{c}_s^\dagger \hat{c}_s^\dagger + i\hbar \frac{\varsigma^*}{2} e^{i\omega_p t} \hat{c}_s \hat{c}_s. \quad (78)$$

The condition  $J \neq 0$  is similar to the three-wave phase matching condition for the wave vectors of modes participating in the parametric decay. The phase matching needs to be satisfied together with frequency matching in Eq. (58), which could be highly nontrivial in a 3D geometry and in a dispersive medium. An important advantage of a subwavelength cavity is that these conditions are straightforward to satisfy by adjusting the cavity geometry. Indeed, consider the decay of the pump into two lowest-order TE<sub>011</sub> modes satisfying Eq. (77); in this case  $\zeta_s(x, y) = \cos\left(\frac{\pi y}{L_y}\right) \cos\left(\frac{\pi x}{L_x}\right)$ . For a pumping mode of TE<sub>01N</sub> type with N even,  $J = 0$ ; however for N odd we

get  $J \neq 0$ . For example, for a TE<sub>013</sub> pumping mode (see Fig. 1), when  $\zeta_p(x, y) = \cos\left(\frac{\pi y}{L_y}\right) \cos\left(\frac{3\pi x}{L_x}\right)$ , we obtain:

$J = \frac{L_x L_y}{\pi^2} \times \frac{16}{45}$ . In this case from Eq. (77) and mode frequencies given by Eq. (68) one can get a condition for cavity sizes:

$$\frac{L_x}{L_y} = \sqrt{\frac{9 - 4 \frac{\epsilon(2\omega_s)}{\epsilon(\omega_s)}}{4 \frac{\epsilon(2\omega_s)}{\epsilon(\omega_s)} - 1}}$$

**VI. EQUATIONS FOR PARAMETRIC DECAY IN A DISSIPATIVE SYSTEM.  
HEISENBERG-LANGEVIN APPROACH**

Here we take into account absorption and radiative losses within the Heisenberg-Langevin formalism. We remind the reader that this approach assumes that the coupling of a dynamic subsystem to a dissipative reservoir is sufficiently weak. If this is not the case and the coupling is strong, the process of energy loss by a given EM mode should be described within a closed Hamiltonian system (e.g. as a coupling to other EM modes, phonons etc.). In this case one does not need any Langevin sources, because in a Hamiltonian system proper commutation relations are satisfied automatically. Whenever the energy exchange of a dynamical subsystem with a reservoir is relatively weak and can be considered within a phenomenological approach, the “dissipation + the Langevin noise” model should be valid for any mechanism of dissipation.

For example, we assume here that the spatial mode structure corresponds to the one in an ideal cavity, whereas diffraction losses of the field out of a cavity can be described by an effective loss rate. This assumption obviously works as long as losses do not affect the mode structure significantly. If the latter is not true, one would have to solve a rigorous diffraction problem which couples the field modes in the cavity and outside. For such a rigorous problem all commutation relations would be satisfied automatically.

Introducing operators of slowly varying field amplitudes, namely  $\hat{c}_{s,i} = \hat{c}_{0s,i}(t)e^{-i\omega_s t}$ ,  $\hat{c}_{s,i}^\dagger = \hat{c}_{0s,i}^\dagger(t)e^{+i\omega_s t}$ , we obtain from Eqs. (73) the following equations:

$$\left. \begin{aligned} \dot{\hat{c}}_{0s} + \Gamma_s \hat{c}_{0s} + \varsigma \hat{c}_{0i}^\dagger &= \hat{L}_s \\ \dot{\hat{c}}_{0i} + \Gamma_i \hat{c}_{0i} + \varsigma \hat{c}_{0s}^\dagger &= \hat{L}_i \end{aligned} \right\}, \quad (79)$$

where  $\Gamma_{s,i} = \Gamma_{r(s,i)} + \Gamma_{\sigma(s,i)}$ , the coefficients  $\Gamma_{r(s,i)}$  and  $\Gamma_{\sigma(s,i)}$  denote, respectively, radiative losses due to the outcoupling of radiation from the cavity and absorption losses due to intracavity absorption.  $\hat{L}_{s,i}$  are the Langevin noise operators. To preserve commutation relations  $[\hat{c}_{0i}, \hat{c}_{0i}^\dagger] = [\hat{c}_{0s}, \hat{c}_{0s}^\dagger] = 1$  at  $\Gamma_{s,i} \neq 0$  the noise operators in the right-hand side of Eqs. (79) should satisfy the same commutation relations as in the case of one quantum oscillator [6, 23, 51] and they should also commute with each other:

$$\left[ \hat{L}_s(t'), \hat{L}_s^\dagger(t) \right] = 2\Gamma_s \delta(t-t'), \quad \left[ \hat{L}_i(t'), \hat{L}_i^\dagger(t) \right] = 2\Gamma_i \delta(t-t'), \quad \left[ \hat{L}_s(t'), \hat{L}_i^\dagger(t) \right] = 0. \quad (80)$$

The fact that commutation relations are the same for one quantum oscillator and for two (or more) interacting oscillators is expected, since the processes within the Hamiltonian dynamics do not affect the commutators; this can be easily checked, for example for the system described by the Hamiltonian Eq. (76). Noise correlators can be defined by generalizing the simplest expression in [23] to the case of two absorbers with different temperatures:

$$\left. \begin{aligned} \langle \hat{L}_s^\dagger(t) \hat{L}_s(t') \rangle &= 2 [\Gamma_{\sigma s} n_{T_\sigma}(\omega_s) + \Gamma_{r s} n_{T_r}(\omega_s)] \delta(t-t') \\ \langle \hat{L}_i^\dagger(t) \hat{L}_i(t') \rangle &= 2 [\Gamma_{\sigma i} n_{T_\sigma}(\omega_i) + \Gamma_{r i} n_{T_r}(\omega_i)] \delta(t-t') \end{aligned} \right\} \quad (81)$$

where  $n_{T_{r,\sigma}}$  is the average number of thermal photons at temperature  $T_{r,\sigma}$ ;  $T_r$  and  $T_\sigma$  denote the temperature outside and inside the cavity, respectively. Expressions (81) imply that dissipative and radiative noises are not correlated.

The solution to Eqs. (79) can be represented as [19, 27]

$$\begin{pmatrix} \hat{c}_{0s} \\ \hat{c}_{0s}^\dagger \end{pmatrix} = \begin{pmatrix} 1 \\ K_1 \end{pmatrix} e^{-\lambda_1 t} \left( \hat{c}_1 + \int_0^t e^{\lambda_1 t'} \hat{L}_1(t') dt' \right) + \begin{pmatrix} 1 \\ K_2 \end{pmatrix} e^{-\lambda_2 t} \left( \hat{c}_2 + \int_0^t e^{\lambda_2 t'} \hat{L}_2(t') dt' \right), \quad (82)$$

where  $\lambda_{1,2}$  and  $\begin{pmatrix} 1 \\ K_1 \end{pmatrix}$  are eigenvalues and eigenvectors of the  $2 \times 2$  matrix:

$$\begin{pmatrix} \Gamma_s & \varsigma \\ \varsigma^* & \Gamma_i \end{pmatrix} \times \begin{pmatrix} 1 \\ K_{1,2} \end{pmatrix} = \lambda_{1,2} \begin{pmatrix} 1 \\ K_{1,2} \end{pmatrix}, \quad (83)$$

$$\left. \begin{aligned} \hat{c}_1 &= \frac{K_2 \hat{c}_s(0) - \hat{c}_i^\dagger(0)}{K_2 - K_1}, & \hat{c}_2 &= -\frac{K_1 \hat{c}_s(0) - \hat{c}_i^\dagger(0)}{K_2 - K_1} \\ \hat{L}_1 &= \frac{K_2 \hat{L}_s - \hat{L}_i^\dagger}{K_2 - K_1}, & \hat{L}_2 &= -\frac{K_1 \hat{L}_s - \hat{L}_i^\dagger}{K_2 - K_1} \end{aligned} \right\}, \quad (84)$$

$\hat{c}_s(0)$  and  $\hat{c}_i^\dagger(0)$  are initial conditions.

From the solution (82)-(83) one can derive a standard-looking condition for the parametric instability (see e.g. [54]):

$$|\varsigma|^2 > \Gamma_s \Gamma_i. \quad (85)$$

Consider the inequality (85) in more detail, neglecting for clarity the frequency dispersion of the dielectric filling of a cavity. Taking into account Eq. (71), one can derive from Eq. (74) that

$$|\varsigma| = |\chi^{(2)} E_p| \sqrt{\omega_s \omega_i} \frac{8\pi l}{L_z \varepsilon} \frac{J}{L_x L_y}, \quad (86)$$

where the dimensionless factor  $J/(L_x L_y)$  depends only on the spatial structure of the modes with frequencies  $\omega_{p,s,i}$ . From Eqs. (85), (86) the instability condition takes the form

$$\frac{256\pi^2}{\varepsilon^2 L_z^2} (\chi^{(2)} l)^2 |E_p|^2 \left( \frac{\omega_s}{\Delta\omega_s} \right) \left( \frac{\omega_i}{\Delta\omega_i} \right) \left( \frac{J}{L_x L_y} \right)^2 > 1, \quad (87)$$

where  $\Delta\omega_{s,i} = 2\Gamma_{s,i}$  are the linewidths for the signal and idler modes.

To avoid cumbersome expressions, consider the decay of pumping into identical quanta as in Eq. (77). In this case the instability condition is  $|\varsigma| > \Gamma_s$ . It is convenient to choose the phase of the pump mode so that the value of  $\varsigma$  is real and positive. Then Eqs. (82)-(84) yield

$$\begin{aligned} \hat{c}_{0s} = e^{-\Gamma_s t} [\hat{c}_s(0) \cosh(\varsigma t) - \hat{c}_s^\dagger(0) \sinh(\varsigma t)] + \int_0^t e^{(-\varsigma + \Gamma_s)(t-t')} \frac{\hat{L}_s(t') - \hat{L}_s^\dagger(t')}{2} dt' + \\ + \int_0^t e^{(\varsigma + \Gamma_s)(t-t')} \frac{\hat{L}_s(t') + \hat{L}_s^\dagger(t')}{2} dt'. \quad (88) \end{aligned}$$

Taking into account the properties of Langevin operators in Eq. (79) and taking  $\langle \hat{c}_s^\dagger(0) \hat{c}_s^\dagger(0) \rangle = \langle \hat{c}_s(0) \hat{c}_s(0) \rangle = 0$  as an initial state, one can derive from Eq. (88) the average photon numbers for signal modes  $n_s = \langle \hat{c}_s^\dagger \hat{c}_s \rangle = \langle \hat{c}_{0s}^\dagger \hat{c}_{0s} \rangle$ :

$$\begin{aligned} n_s = e^{-2\Gamma_s t} \{ n_s(0) [\cosh^2(\varsigma t) + \sinh^2(\varsigma t)] + \sinh^2(\varsigma t) \} \\ + [\Gamma_{\sigma s} n_{T_\sigma}(\omega_s) + \Gamma_{rs} n_{T_r}(\omega_s)] \times \left( \frac{1 - e^{2(\varsigma - \Gamma_s)t}}{2(-\varsigma + \Gamma_s)} + \frac{1 - e^{2(-\varsigma - \Gamma_s)t}}{2(\varsigma + \Gamma_s)} \right) \\ + \Gamma_s \left( \frac{1 - e^{2(\varsigma - \Gamma_s)t}}{4(-\varsigma + \Gamma_s)} + \frac{1 - e^{2(-\varsigma - \Gamma_s)t}}{4(\varsigma + \Gamma_s)} - \frac{1 - e^{-2\Gamma_s t}}{2\Gamma_s} \right), \quad (89) \end{aligned}$$

where  $\Gamma_s = \Gamma_{\sigma s} + \Gamma_{rs}$ . When the parametric amplification starts from the level of vacuum fluctuations, one should put  $n_s(0) = 0$  in Eq. (89).

In the limit of zero pumping intensity, Eq. (89) gives an expression which describes how the initial perturbation of a photon number approaches equilibrium:

$$n_s = e^{-2\Gamma_s t} n_s(0) + \frac{\Gamma_{\sigma s} n_{T_\sigma}(\omega_s) + \Gamma_{rs} n_{T_r}(\omega_s)}{\Gamma_s} \times (1 - e^{-2\Gamma_s t}). \quad (90)$$

Above the instability threshold, when  $\varsigma \gg \Gamma_s$ , it is enough to keep only exponentially growing terms in Eq. (89). We can also assume that an average number of thermal photons in an ambient space  $n_{T_r}(\omega_s)$  is negligible. This gives an expression for the parametric signal power  $P_s \approx 2\varsigma \hbar \omega_s n_s$ :

$$P_s \approx \varsigma \hbar \omega_s e^{2\varsigma t} \left[ n_s(0) + \frac{\Gamma_{\sigma s}}{\varsigma} n_{T_\sigma}(\omega_s) + \frac{1}{2} \right]. \quad (91)$$

Obviously this expression is valid only at the initial linear stage. The subsequent evolution is governed by the nonlinear pump depletion and nonlinear modification of phasematching conditions and losses. An order-of magnitude estimate of the maximum steady-state power can be obtained from Manley-Rowe relations as shown below for a specific example.

The fractions of the power emitted outside and absorbed inside a cavity are  $P_{rs} \approx \Gamma_{rs} P_s / \varsigma$  and  $P_{\sigma s} \approx \Gamma_{\sigma s} P_s / \varsigma$  respectively; most of the power is accumulated in a cavity. From Eq. (91) one can see that the amplification of intrinsic thermal noise of a QW layer with temperature  $T_\sigma$  can be ignored if  $\frac{\Gamma_{\sigma s}}{\varsigma} \cdot \frac{2}{\exp(\hbar\omega_s/T_\sigma) - 1} \ll 1$ .

When the parametric growth rate is lower than losses,  $\zeta < \Gamma_s$ , the general solution Eq. (89) describes the regime of spontaneous parametric down-conversion. In the stationary limit, when  $(\Gamma_s - \zeta)t \rightarrow \infty$ , the radiated signal power  $P_s \approx 2\Gamma_{rs}\hbar\omega_s n_s$  becomes

$$P_s = \hbar\omega_s \frac{2\Gamma_{rs}\Gamma_s}{\Gamma_s^2 - \zeta^2} [\Gamma_{\sigma s} n_{T_\sigma}(\omega_s) + \Gamma_{rs} n_{T_r}(\omega_s)] + \hbar\omega_s \frac{\Gamma_{rs}\zeta^2}{\Gamma_s^2 - \zeta^2}. \quad (92)$$

The first term in the right-hand side of Eq. (92) is due to the thermal emission modified by the parametric decay of the pumping photons. The second term originates from the parametric decay of pumping photons under the action of vacuum fluctuations of the intracavity field; this is a purely quantum effect. Thermal effects can be neglected if

$$\frac{\Gamma_s \Gamma_{(\sigma,r)s}}{\zeta^2} \frac{2}{\exp(\hbar\omega_s/(k_B T_{\sigma,r})) - 1} \ll 1.$$

The results in Eqs. (89)-(92) provide the dependence of the parametric signal from all relevant dissipation and fluctuation effects. In addition to dissipation and thermal fluctuations due to absorption in the cavity walls and a semiconductor heterostructure, they take into account outcoupling of a signal into the ambient space and coupling to thermal photons from the environment. Eq. (92) determines the spontaneous parametric signal emitted from a cavity against the background of noise created by both thermal radiation from a cavity and reemission of thermal photons coupled into a cavity from the outside. The background noise depends from the cavity temperature and the environment temperature. In addition to the spontaneous decay process we take into account the modification of background noise by pumping.

For a numerical example, consider a nanocavity filled with multiple quantum wells, excited with a mid-infrared pump, as reported in [31]. Using Eq. (87) for their values of intersubband nonlinearity  $|\chi^{(2)}| \sim 3 \times 10^{-7}$  m/V,  $\omega_{s,i}/\Delta\omega_{s,i} \sim 20$  and  $J/L_x L_y \sim 0.3$  we obtain the intracavity pump field at the instability threshold to be  $E_p \simeq 8$  kV/cm, which is achievable and is lower than the saturation field for the intersubband nonlinearity. Above the threshold, the signal and idler fields start growing inside the cavity until they get limited by the Manley-Rowe relations [32], i.e. the intracavity signal field reaches  $|E_s| \simeq |E_p|/\sqrt{2}$ . The maximum output signal power per one nanocavity described in [31] is about  $8 \times 10^{-7}$  W for the photon leakage rate  $\Gamma_{rs} = 10^{12}$  s<sup>-1</sup>.

Far below the instability threshold, when  $\zeta \ll \Gamma_s$ , the spontaneous rate of parametric decay scales as  $(\Gamma_{rs}/\Gamma_s^2)\zeta^2$ . For the parameters from the above numerical example, when  $\zeta = \Gamma_s/2$  the emission rate of signal photons is around  $3 \times 10^{11}$  s<sup>-1</sup> and the power is 6 nW.

A very high second-order nonlinear surface conductivity for graphene was reported in [46], corresponding to the effective bulk susceptibility  $|\chi^{(2)}| \sim 10^{-3}$  m/V per monolayer in the THz range. This large susceptibility is partially offset by a small factor  $l/L_z$  in Eq. (86) where  $l$  is a thickness of the graphene layer. However, for hBN-encapsulated graphene utilized to fabricate low-disorder graphene samples [55] the total cavity thickness  $L_z$  can be as small as several nm, so the factor  $l/L_z$  can be as large as 0.1. Even factoring in enhanced cavity losses, this can yield a lower parametric instability threshold as compared to semiconductor quantum well samples.

## VII. COMPARING PARAMETRIC INSTABILITY IN A SUBWAVELENGTH CAVITY AND IN A FABRY-PEROT CAVITY

Compare the parametric instability in a subwavelength cavity with similar instability of modes in a Fabry-Perot (FP) cavity with all three dimensions larger than wavelength, which we will call a quasi-optical cavity. Consider a planar quasi-2D cavity of the surface area  $L_x L_y$ , in which the waves are propagating along the nonlinear layer of thickness  $l$  much smaller than the cavity thickness  $L_{FP}$  transverse to the nonlinear layer, so the cavity volume is  $L_{FP} L_x L_y$ . The dielectric constant of a cavity filling is  $\epsilon$ . In this case the parametric decay is still described by Eqs. (79), (74), and (75), in which the relaxation constants  $\Gamma_{s,i}$  and the overlap integral are determined by the FP cavity Q-factor and the corresponding spatial structure of the modes. For the normalization constants of the quantum fields entering Eq. (74) we use standard expressions for a two-mirror FP cavity:

$$|E_{s,i}|^2 = \frac{4\pi\hbar\omega_{s,i}}{L_x L_y L_{FP} \epsilon}. \quad (93)$$

The resulting parametric instability threshold is

$$\frac{64\pi^2}{\epsilon^2 L_{FP}^2} (\chi^{(2)} l)^2 |E_p|^2 \left(\frac{\omega_s}{\Delta\omega_s}\right) \left(\frac{\omega_i}{\Delta\omega_i}\right) \left(\frac{J}{L_x L_y}\right)^2 > 1, \quad (94)$$

where  $\Delta\omega_{s,i} \approx 2\Gamma_{s,i}$ .

As we already pointed out, in a cavity with all three dimensions larger than the wavelength the phase matching condition for a three-wave mixing may be more difficult to satisfy. Even if we assume that phase matching is somehow arranged and the geometric factor  $J/L_x L_y$  is of the same order as in a subwavelength cavity, the latter is expected to have a lower parametric threshold. Indeed the ratio of the threshold pump intensity  $|E_p|^2$  in a subwavelength cavity to that in a quasi-optical cavity scales as  $\sim \left(\frac{L_z}{L_{FP}}\right)^2 \left(\frac{\Delta\omega_{eff}}{\Delta\omega_{FP}}\right)^2$ , where  $\Delta\omega_{eff}$  and  $\Delta\omega_{FP} \approx \Delta\omega_{s,i}$  are typical linewidths of the subwavelength cavity and FP cavity modes, respectively. This ratio can be much smaller than 1, which indicates that a much lower pumping is needed to reach the parametric instability threshold in a subwavelength cavity, even if the FP cavity has a higher Q-factor as compared to the subwavelength cavity,  $\Delta\omega_{FP} < \Delta\omega_{eff}$ .

A plane-parallel quasi-2D subwavelength cavity geometry considered in this paper is the most natural choice for integration with 2D nonlinear materials. However, other geometries are also possible, for example plasmonic or grating structures supporting surface modes. To get an order of magnitude estimate of the parametric threshold, one can use our results in Eqs. (87), (94) after replacing  $L_z$  or  $L_{FP}$  with a mode size transversely to the nonlinear layer.

Ultracompact subwavelength electrodynamic structures utilizing 2D materials are promising for applications in integrated photonic circuits, whenever one needs a compact planar architecture. At the same time, due to strong dissipation they are unlikely to outperform conventional nonlinear devices made of bulk transparent nonlinear materials when it comes to the nonlinear conversion efficiency and power. For example, in [56] the authors realized low-threshold mode-matched parametric generation in whispering gallery mode resonators made entirely of bulk lithium niobate. In this case the bulk nonlinear material occupies all modal volume. The lower nonlinearity and the loss of Purcell enhancement are compensated by lower dissipation and increased interaction volume.

In conclusion, we applied a consistent Heisenberg-Langevin formalism to the process of nonlinear parametric decay of cavity modes in planar subwavelength cavities containing 2D nonlinear materials. We derived general analytic formulas for the spontaneous parametric signal and threshold of stimulated parametric decay of a pump cavity mode into the signal and idler modes. We found that a significant reduction in the parametric instability threshold can be achieved for realistic materials and cavity parameters due to Purcell enhancement.

## Magneto-optics of massless fermions in Weyl semimetals

Exotic topological and transport properties of Weyl semimetals generated a lot of excitement in the condensed matter community. Here we show that Weyl semimetals in a strong magnetic field are highly unusual optical materials. The propagation of electromagnetic waves is affected by an interplay between plasmonic response of chiral Weyl fermions and extreme anisotropy induced by a magnetic field. The resulting magneto-polaritons possess a number of peculiar properties, such as hyperbolic dispersion, photonic stop bands, coupling-induced transparency, and broadband polarization conversion. These effects can be used for optical spectroscopy of these materials including detection of the chiral anomaly, or for broadband terahertz/infrared applications.

Weyl semimetals (WSMs) have unusual electronic and transport properties originating from the nontrivial topology of the Brillouin zone [58–60]. They have been studied experimentally, mostly with angle-resolved photoemission spectroscopy; e.g. [61–63]. The most intensely studied phenomena include topologically protected surface states known as Fermi arcs, the chiral anomaly, or the non-conservation of the chiral charge in parallel electric and magnetic fields, and the resulting anomalous magnetoresistance [64–67]. Optics of WSMs received relatively less attention so far. Far-infrared optical spectroscopy studies of TaAs without the magnetic field have been recently reported [68]. The conductivity, magnetoplasmons, and polaritons in a magnetic field were calculated recently in quasiclassical approximation [69–75]. The strong-field optical conductivity was calculated in [76]. Here we concentrate on the wave propagation in WSMs in a strong magnetic field, when the electron motion transverse to the field becomes quantized. We show that hybridization of magnetoplasmons with electromagnetic (EM) waves in WSMs leads to fascinating optical phenomena involving magnetopolaritons: hyperbolic dispersion, the absence of Landau damping for strongly localized excitations, photonic stop bands, coupling-induced transparency, efficient polarization conversion, and pulse compression, to name a few. We show that optical spectroscopic techniques provide a straightforward and “clean” way of detecting topological properties of low-energy electron states and in particular the chiral anomaly. Moreover, WSMs show strong promise for future photonic chips enabling a wide array of broadband optoelectronic applications, such as polarizers, modulators, switches, and pulse shapers for mid-infrared through terahertz wavelengths.

### VIII. DIELECTRIC TENSOR FOR A WSM IN A MAGNETIC FIELD

We consider the material which has only one pair of Weyl nodes for simplicity, with low-energy excitations around each node described by the Weyl Hamiltonian,

$$H = \chi \hbar v_F \boldsymbol{\sigma} \mathbf{k}. \quad (95)$$

Here  $\chi = \pm 1$  is the chirality index,  $\boldsymbol{\sigma}$  is a 3D vector of Pauli matrices,  $\mathbf{k}$  is the 3D quasimomentum of electrons counted from the Weyl node, and we assumed an isotropic electron dispersion (scalar constant  $v_F$ ). In a strong magnetic field oriented along  $z$  axis the 3D conical spectrum of quasiparticles near each node is split into Landau-level (LL) subbands  $W_n$  labeled by the quantum number  $n$ :

$$W_n = \text{sgn}(n) \hbar v_F \sqrt{\frac{2|n|}{l_b^2} + k_z^2} \text{ for } n \neq 0, \quad (96)$$

$$W_0^{(\chi)} = -\chi \hbar v_F k_z \quad (97)$$

where  $l_b = \sqrt{\frac{\hbar c}{eB}}$  is the magnetic length. We assume that the field is strong enough so that  $W_1 - W_0$  at  $k_z = 0$  is much larger than the LL broadening determined by disorder.

The salient feature of the electron spectrum is the presence of chiral electron states with 1D linear dispersion at  $n = 0$  LL. The  $n = 0$  electrons near each node are able to move only in one direction, depending on the sign of  $\chi$  and neglecting internode scattering. The majority of peculiar optical properties of WSMs originates from the response of these electron states and its interplay with inter-LL transitions.

The dielectric tensor for chiral fermions in WSMs has a general structure typical for a magnetized electron-hole plasma:

$$\epsilon_{ij} = \begin{pmatrix} \epsilon_{\perp} & ig & 0 \\ -ig & \epsilon_{\perp} & 0 \\ 0 & 0 & \epsilon_{zz} \end{pmatrix} \quad (98)$$

where  $i, j = x, y, z$ . However, the expressions for its components and the resulting optical response are far from typical. Consider first the longitudinal component  $\epsilon_{zz} = \epsilon_b + 4\pi i \sigma_{zz} / \omega$  where  $\epsilon_b$  is the background dielectric constant

of a crystal. The conductivity  $\sigma_{zz}$  can be found by calculating the linear response to the longitudinal field  $E_z = \text{Re}[\mathcal{E}e^{iq_z z - i\omega t}]$ . It is convenient to define the optical field through the scalar potential  $\phi = \text{Re}[\Phi e^{iq_z z - i\omega t}]$  as  $\mathcal{E} = -iq_z \Phi$ . We will assume for simplicity that the Fermi energy for each chirality is between  $n = -1$  and  $n = 1$  and the temperature is low enough so that the states with  $n \neq 0$  are either completely filled or empty. Note also that for the longitudinal field  $\mathbf{E} \parallel \mathbf{B}$  the transitions between the Landau levels are forbidden in the electric dipole approximation. The resulting linearized density matrix equation for the density matrix elements  $\rho_{kk'}^{(\chi)}$  for each chirality is

$$-i\omega\rho_{k_z, k'_z}^{(\chi)} + i\frac{W_0^{(\chi)}(k_z) - W_0^{(\chi)}(k'_z)}{\hbar}\rho_{k_z, k'_z}^{(\chi)} = -\frac{i}{\hbar}e\Phi \langle n=0, k_z | e^{iq_z z} | n=0, k'_z \rangle \left[ f_0^{(\chi)}(k_z) - f_0^{(\chi)}(k'_z) \right], \quad (99)$$

where  $f_0^{(\chi)}(k_z)$  are populations at  $n = 0$  unperturbed by the optical field and we neglected relaxation, which will be added later. The matrix element in Eq. (99) is calculated using the electron states in a magnetic field; it is equal to the delta-function  $\delta_{k_z - q_z, k'_z}$ . The solution of Eq. (99) in the limit  $k_z \gg q_z$  is

$$\rho_{k_z, k_z - q_z}^{(\chi)} = \frac{ie\mathcal{E}}{\omega - \chi q_z v_F} \frac{\partial f_0^{(\chi)}(k_z)}{\hbar \partial k_z}. \quad (100)$$

The complex amplitude of the Fourier component of the electric current  $j_z = \text{Re}\tilde{J}e^{iq_z z - i\omega t}$  is given by

$$\tilde{J} = \sum_{k_z, \chi} (j_z)_{k_z - q_z, k_z}^{(\chi)} \rho_{k_z, k_z - q_z}^{(\chi)}, \quad (101)$$

where the matrix element of the spatial Fourier component of the current is

$$(j_z)_{k'_z, k_z}^{(\chi)} = -e \langle n=0, k'_z | e^{-iq_z z} \chi v_F \sigma_z | n=0, k_z \rangle \quad (102)$$

and the sum can be replaced by integration. The resulting longitudinal component of the conductivity tensor is

$$\sigma_{zz} = \frac{ie^3 B v_F \omega}{2\pi^2 \hbar^2 c} \frac{1}{\omega^2 - q_z^2 v_F^2}, \quad (103)$$

where the  $B$ -dependence appeared due to the density of states in a quantizing magnetic field. The longitudinal dielectric tensor component therefore takes the form

$$\epsilon_{zz} = \epsilon_b - \omega_p^2 / (\omega^2 - q_z^2 v_F^2). \quad (104)$$

This result can be also obtained from the kinetic equation. This expression has several peculiar features. First, since the electrons at  $n = 0$  can move only in one direction with the same velocity  $v_F$ , they cannot bunch in the velocity space and there is no Landau damping. Mathematically, the Landau damping emerges due to contribution from the pole in the integral over electron momenta in the linear conductivity. However, in our case there is no pole in the integral in Eq. (101) since the denominator in Eq. (100) does not depend on the electron momentum.

Second, the effective plasma frequency in Eq. (104) does not depend on the electron density:  $\omega_p^2 = \frac{2\alpha}{\pi} \frac{eBv_F}{\hbar}$ , where  $\alpha = \frac{e^2}{\hbar c}$ ; see also [66, 69]. In the limit of a uniform electric field  $q_z = 0$  Eq. (103) immediately gives the chiral anomaly. Indeed, if only  $n = 0$  electrons are involved, the chiral current  $j_{\text{chir}} = \partial[N^{(\chi=+1)} - N^{(\chi=-1)}] / \partial t$  is related to the charge current in a uniform but time-dependent field  $\mathbf{E} \parallel \mathbf{B}$  as  $\partial j_z / \partial t = -e v_F j_{\text{chir}}$ . This gives the chiral anomaly current  $j_{\text{chir}} = -e^2 \mathbf{E} \mathbf{B} / (2\pi^2 \hbar^2 c)$ , in agreement with previous results; see e.g. [59, 60] for review.

## IX. THE DISPERSION EQUATION

EM waves incident on a magnetized WSM propagate as eigenmodes that can be called magneto-polaritons. They are the solutions of Maxwell's equations for plane waves with the dielectric tensor from Eq. (98). For the photon wave vector  $\mathbf{q}$  in the  $(xz)$ -plane making an angle  $\theta$  with the magnetic field direction along  $z$ -axis, they can be written as

$$\begin{pmatrix} \epsilon_+ - \frac{1}{2}\mu^2(1 + \cos^2\theta) & \frac{1}{2}\mu^2\sin^2\theta & \frac{1}{\sqrt{2}}\mu^2\sin\theta\cos\theta \\ \frac{1}{2}\mu^2\sin^2\theta & \epsilon_- - \frac{1}{2}\mu^2(1 + \cos^2\theta) & \frac{1}{\sqrt{2}}\mu^2\sin\theta\cos\theta \\ \frac{1}{\sqrt{2}}\mu^2\sin\theta\cos\theta & \frac{1}{\sqrt{2}}\mu^2\sin\theta\cos\theta & \epsilon_{zz} - \mu^2\sin^2\theta \end{pmatrix} \begin{pmatrix} E_+ \\ E_- \\ E_z \end{pmatrix} = 0, \quad (105)$$

where  $\mu^2 = \frac{c^2 q^2}{\omega^2}$ ,  $\epsilon_{\pm} = \epsilon_{\perp} \pm g$ , and  $E_{\pm} = \frac{1}{\sqrt{2}}(E_x \pm iE_y)$ .

## X. PROPERTIES OF NORMAL ELECTROMAGNETIC MODES IN WEYL SEMIMETALS

### A. Longitudinal propagation

For the waves propagating strictly along the magnetic field, i.e.  $\theta = 0$ , the solution to Eqs. (105) consists of two eigenmodes with transverse polarization (“photons”),

$$\mu_{L,R}^2 = \epsilon_{\pm}, \quad \mathbf{E}_{L,R} = \frac{1}{\sqrt{2}}E_{\pm}(\mathbf{x}_o \pm i\mathbf{y}_o), \quad (106)$$

and the wave with the longitudinal polarization  $\mathbf{E} = E_z \mathbf{z}_o$  and dispersion equation  $\epsilon_{zz} = 0$  (“plasmon”). The plasmon dispersion is

$$\omega^2 = \frac{\omega_p^2}{\epsilon_b} + v_F^2 q^2. \quad (107)$$

We emphasize again that, in contrast to “usual” plasmons, there is no cutoff in Eq. (107) due to Landau damping at large wave vectors  $q > \omega/v_F$ . Therefore, a much stronger plasmon localization is allowed, with propagation only limited by absorption due to scattering on impurities etc.

### B. Oblique propagation

For oblique propagation, even at very small angles  $\theta$ , the plasmons and transverse waves are coupled to form hybrid plasmon-polaritons. To determine general trends and obtain analytic results, we neglect the spatial dispersion ( $q_z$ -dependence) of  $\epsilon_{zz}$  in Eq. (104), which is possible as long as  $\mu^2 \sin^2\theta \ll c^2/v_F^2$ . This is not so restrictive since  $c/v_F > 100$ . We also neglect any spatial dispersion in  $\epsilon_{\pm}$  in the dipole approximation.

It is instructive first to consider the case when the Fermi level is exactly at the Weyl point for both chiralities. In this case, due to electron-hole symmetry the off-diagonal terms in Eq. (98) vanish and the dielectric tensor looks like the one for a uniaxial anisotropic medium. The dispersion equation for the extraordinary wave, i.e. the one polarized in the  $(x, z)$  plane, can be written as

$$\frac{\mu_x^2}{\epsilon_{zz}} + \frac{\mu_z^2}{\epsilon_{\perp}} = 1. \quad (108)$$

The transverse components of the dielectric tensor are always positive, whereas the  $\epsilon_{zz}$  component becomes negative for frequencies below the plasmon resonance,  $\omega^2 < \omega_p^2/\epsilon_b$ . In this case Eq. (108) becomes hyperbolic and its isofrequency lines are hyperbolae. Therefore, a magnetized WSM is a natural hyperbolic material at low enough frequencies. Another natural hyperbolic material is hexagonal boron nitride, where the hyperbolic dispersion exists in two narrow spectral ranges near the phonon bands [78]. Otherwise, hyperbolic dispersion is achieved in the effective medium approximation in metal/dielectric metamaterials prepared by nanofabrication [79]. It is promising for numerous applications from superlenses and nanoimaging to photonic integrated circuits. The plasmon resonance frequency  $\omega = \omega_p/\sqrt{\epsilon_b}$  in WSMs which determines the upper bound for hyperbolic dispersion is in the THz to far-infrared range for a magnetic field of 1-10 Tesla,  $\epsilon_b \sim 10$  and  $v_F \sim 10^8$  cm/s. It is lower than the inter-LL absorption edge for all magnetic fields, so the only loss mechanism is due to scattering on impurities which depends on the material quality.

The ordinary wave in this limit is linearly polarized along  $y$ -axis and has a standard dispersion  $\mu^2 = \epsilon_{\perp}$ .

Fig. 5 shows the dispersion (real part of  $\mu$ ) for the extraordinary waves for several different propagation angles  $\theta$ . Far from inter-LL transitions, we can neglect any dispersion in the transverse part of the dielectric tensor, assuming

$\epsilon_{\perp} = \epsilon_b \sim 10$ . We also added the scattering rate as an imaginary part of frequency ( $\omega + i\gamma$ ) in the first term of Eq. (99) and took  $\gamma$  to be 0.01 of the plasmon resonance frequency  $\omega_{\text{res}} = \omega_p/\epsilon_b^{1/2}$ . For longitudinal propagation  $\theta = 0$  the photon dispersion is trivial:  $\mu = \sqrt{\epsilon_b}$ . For any nonzero angle, plasmons and photons hybridize. At the hybrid plasmon-polariton resonance  $\mu$  diverges in the absence of dissipation. The stop band appears between the hybrid resonance and plasmon resonance. It is defined by the condition  $\mu^2 < 0$  so that  $\text{Re}[\mu] = 0$  and the wave cannot propagate. At the boundaries of the stop band  $\text{Re}[\mu]$  goes through the value of 1 with a large derivative, leading to a small group velocity  $v_{gr} \ll c$ . This means that a layer of WSM is able to compress a pulse incident from vacuum by a factor  $c/v_{gr}$ . All spectral features are tunable by varying the magnetic field or the angle  $\theta$ .

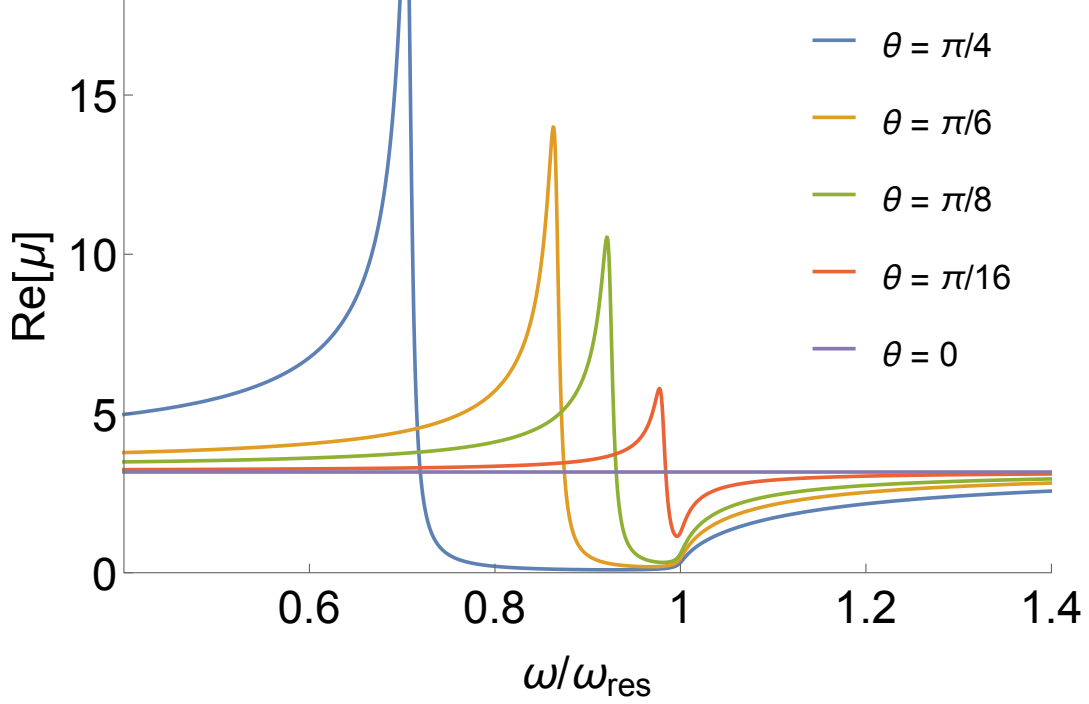


FIG. 5. Dispersion (real part of  $\mu(\omega)$ ) of the extraordinary waves in a magnetic field of 10 T for several different propagation angles  $\theta$ .

Going back to the general case of an arbitrary Fermi level, Eqs. (105), and arbitrary values of  $g$  leads to a biquadratic dispersion equation for  $\mu$ :

$$\frac{\mu^4}{2} \left( (\epsilon_+ + \epsilon_-) \sin^2 \theta + 2\epsilon_{zz} \cos^2 \theta \right) - (\epsilon_+ \epsilon_- \sin^2 \theta + \frac{1}{2} \epsilon_{zz} (\epsilon_+ + \epsilon_-) (1 + \cos^2 \theta)) \mu^2 + \epsilon_+ \epsilon_- \epsilon_{zz} = 0. \quad (109)$$

The polarization coefficients of the normal modes are

$$\frac{E_{\pm}}{E_z} = \frac{-\frac{1}{\sqrt{2}} \mu^2 \sin \theta \cos \theta (\epsilon_{\mp} - \mu^2)}{\epsilon_+ \epsilon_- - \frac{1}{2} \mu^2 (1 + \cos^2 \theta) (\epsilon_+ + \epsilon_-) + \mu^4 \cos^2 \theta}. \quad (110)$$

Equations (109), (110), and (104) provide a complete description of the electromagnetic wave propagation in WSMs. They can be plotted numerically or solved analytically, leading to cumbersome formulas. In the low temperature limit we obtain analytic expressions for all components of the dielectric tensor. Leaving detailed numerical studies to future publications, here we highlight the most interesting cases. Note that a finite separation between Weyl nodes in momentum space by a vector  $2\mathbf{b}$  creates an additional anisotropy vector and gives rise to an additional gyrotropic effect  $g \propto b$  [77]. Therefore, the dispersion shown in Fig. (5) is valid only if this additional  $g$  is small. This will be the case when the Weyl semimetal is created by applying an external magnetic field to a Dirac semimetal, so that the separation of Weyl points is only due to a Zeeman-type interaction which is typically small. We also note that the expressions for the magnetopolariton dispersion for a nonzero  $g$  Eqs. (109) are analytic functions around  $g = 0$ , so the dispersion curves in Fig. (5) will change little when  $g$  is small. The most significant effect of a nonzero  $g$  is the appearance of an elliptical polarization, Eq. (110) instead of the linear one when  $g = 0$ .

### C. Coupling-induced transparency

For quasi-longitudinal propagation,  $\sin^2 \theta \ll 1$  and plasmon-polariton hybridization occurs in the vicinity of the plasmon resonance,  $|\epsilon_{zz}| \ll 1$ . In this case the approximate solution of Eq. (109) is

$$\mu_{1,2}^2 = \frac{1}{(\epsilon_+ + \epsilon_-) \sin^2 \theta + 2\epsilon_{zz}} \left[ \epsilon_+ \epsilon_- \sin^2 \theta + \epsilon_{zz} (\epsilon_+ + \epsilon_-) \pm \sqrt{(\epsilon_+ \epsilon_- \sin^2 \theta)^2 + \epsilon_{zz}^2 (\epsilon_+ - \epsilon_-)^2} \right]. \quad (111)$$

The polarization coefficients become

$$K_{\pm} = \frac{E_{\pm}}{E_z} = \frac{-\frac{1}{\sqrt{2}} \mu^2 \sin \theta}{\epsilon_{\pm} - \mu^2}. \quad (112)$$

In the “non-gyrotropic” limit when  $E_F = 0$  and  $\epsilon_+ = \epsilon_- = \epsilon_{\perp}$ , the extraordinary wave has dispersion

$$\mu_2^2 = \frac{\epsilon_{zz} \epsilon_{\perp}}{\epsilon_{\perp} \sin^2 \theta + \epsilon_{zz}}; \quad K_+ = K_- = -\frac{1}{\sqrt{2}} \frac{\epsilon_{zz}}{\epsilon_{\perp} \sin \theta}. \quad (113)$$

The hybrid resonance corresponds to the vanishing real part of the denominator for  $\mu_2^2$  in Eq. (111) or (113), when  $|\mu_2^2| \gg 1$ .

The effect of *coupling-induced transparency* emerges near the plasmon resonance where  $\mu_2$  can be of the order of 1 or smaller. When the angle  $\theta$  is not too small,  $|\epsilon_{zz}| \ll 1$ ,  $\sin^2 \theta \ll 1$ , but  $|\epsilon_{\pm}| \sin^2 \theta \gg |\epsilon_{zz}|$ , the dispersion and polarization of the “extraordinary” wave (the wave that becomes extraordinary if  $E_F = 0$ ) are simply

$$\mu_2^2 = \frac{\epsilon_{zz}}{\sin^2 \theta}; \quad \frac{E_{x,y}}{E_z} = -\frac{\epsilon_{zz}}{2 \sin \theta} \frac{\epsilon_- \pm \epsilon_+}{\epsilon_+ \epsilon_-}. \quad (114)$$

In this case one can have  $|\mu^2| \ll \epsilon_{\pm}$  whereas the electric field of the wave is directed almost along the magnetic field, i.e. still quasi-longitudinal. Note that  $\mu_2^2$  in Eq. (114) depends only on the  $\epsilon_{zz}$  component, which means that the propagation is not affected at all by the resonant inter-LL absorption losses described by the imaginary parts of  $\epsilon_{\pm}$ . The medium effectively becomes transparent for this wave! More accurately, its losses are determined only by the imaginary part of  $\epsilon_{zz}$ , i.e. disorder-related scattering. Within the transparency band, strong plasmon-photon coupling forces the polarization of the wave to be oriented almost along  $B$ , and therefore it is nearly decoupled from the transitions between LLs. The narrow band of transparency within a broad line of inter-LL absorption is defined by the range of frequencies where  $|\epsilon_{zz}|$  is small enough, namely  $|\epsilon_{zz}| \ll |\epsilon_{\pm}| \sin^2 \theta$ . The situation is similar to the electromagnetically induced transparency (EIT) [80], only in the case of EIT the coupling between two quantum oscillators is provided by a coherent EM drive; see the comparison in [81]. The transparency will be visible if the disorder-related losses determined by  $\text{Im} \epsilon_{zz}$  are lower than the inter-Landau level absorption losses determined by  $\text{Im} \epsilon_{\pm}$ . Introducing the electron scattering rate  $\gamma$  in  $\epsilon_{zz}$ , one can derive the visibility condition as  $\sqrt{\frac{\gamma}{\omega \sin^2 \theta}} < \text{Im} \epsilon_{\pm}$ . The value of  $\gamma$  depends on the particular material, material quality, and temperature. For example, far-infrared spectroscopy of TaAs [68] (without the magnetic field) found the scattering time varying between 3 and 0.4 ps from low to room temperature. A value of  $\text{Im} \epsilon_+ \sim 3 - 5$  around  $\hbar\omega \sim 100$  meV as in Fig. 6 would lead to the visibility condition  $\sin \theta > 0.01 - 0.03$ .

In the same limit the “ordinary” wave has the dispersion  $\mu_1^2 = \frac{2\epsilon_+ \epsilon_-}{\epsilon_+ + \epsilon_-}$  and elliptical polarization in the plane of vectors  $\mathbf{q}$  and  $\mathbf{y}_0$ :  $\frac{E_x}{E_z} = \sin \theta$  and  $\frac{E_y}{E_z} = \frac{i(\epsilon_+ + \epsilon_-)}{\epsilon_+ - \epsilon_-} \sin \theta$ .

### D. Intersubband transitions and optical detection of the chiral anomaly

So far we considered peculiar optical properties of WSMs due to massless 1D chiral fermions at the  $n = 0$  LL. Here we show that resonant inter-LL absorption from  $n = 0$  to  $n \neq 0$  states provides another sensitive method of studying chiral fermions near Weyl nodes and in particular, detecting the chiral anomaly. Consider the propagation of transverse modes in the Faraday geometry when the eigenmodes are left-hand or right-hand circularly polarized (LHC or RHC). Fig. 6 gives an example of the absorption spectrum at low temperatures when the Fermi level  $E_F = 60$  meV is between  $n = 0$  and  $n = 1$  LLs and has the same value for both chiralities. Absorption edges of the lowest-energy

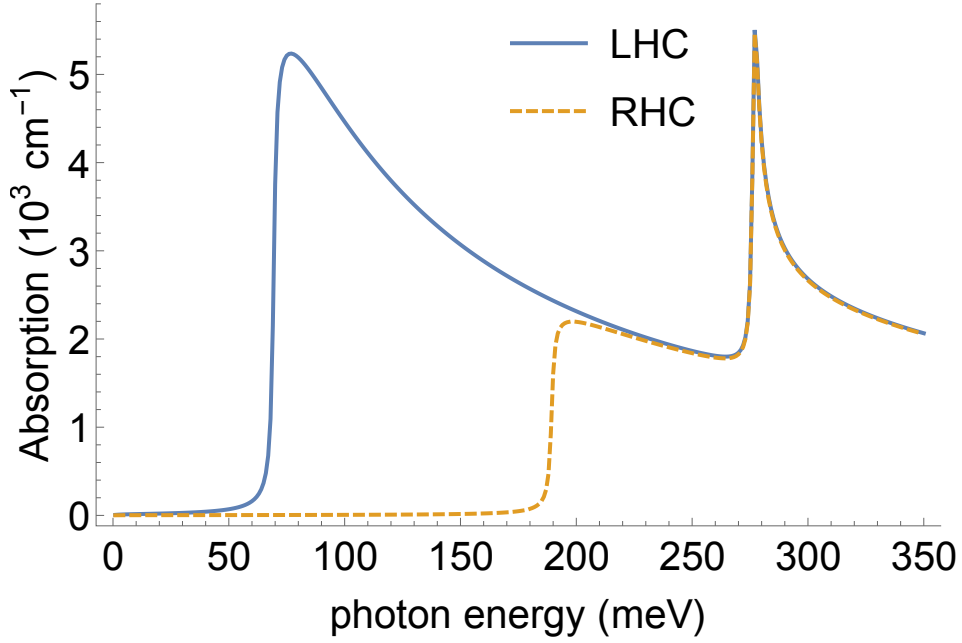


FIG. 6. Absorption spectrum for LHC (solid line) and RHC (dashed line) polarizations in a magnetic field  $B = 10$  T at zero temperature, the Fermi energy of 60 meV, and the relaxation constant  $\gamma = 1$  meV.

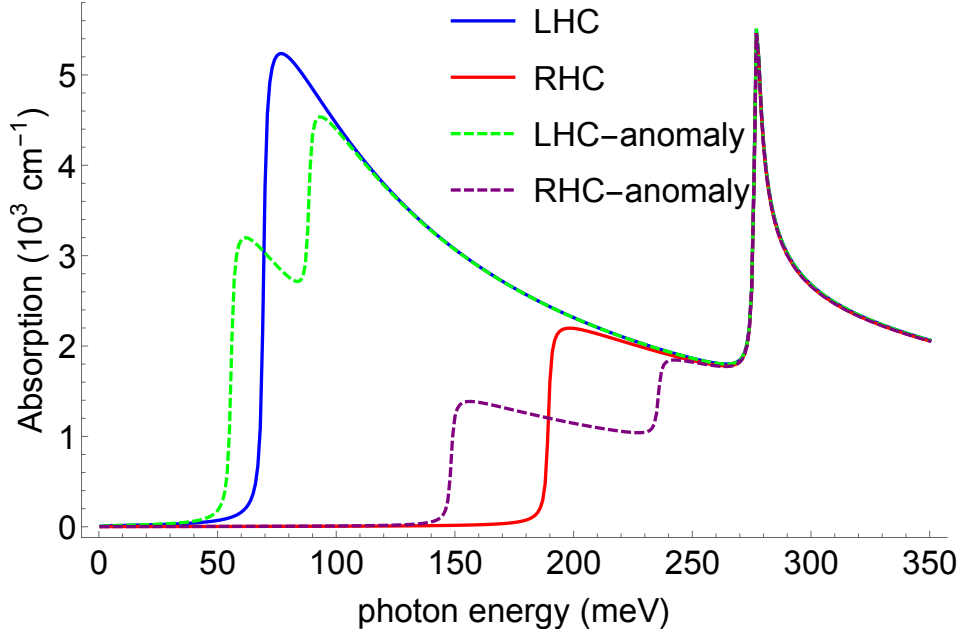


FIG. 7. Absorption spectrum before (solid line) and after (dashed line) a constant electric field  $\mathbf{E} \parallel \mathbf{B}$  is applied which shifts the Fermi levels by  $\pm 30$  meV in the two Weyl nodes. The magnetic field is 10 T and the relaxation constant  $\gamma = 1$  meV.

transitions  $0 \rightarrow 1$ , then  $-1 \rightarrow 0$ ,  $-2 \rightarrow 1$ , and  $-1 \rightarrow 2$  are clearly visible in different polarizations (the last two transitions coincide). In particular, there is a broad range of frequencies between 50 and 200 meV when only the LHC polarization is absorbed. Therefore, a several  $\mu\text{m}$  thin WSM film can serve as a broadband polarizer converting from linear into circular polarization. Note that both the frequency bandwidth and the polarization coefficient are tunable by a magnetic field and/or Fermi level position. Other obvious applications include optical isolators and saturable absorbers.

Fig. 7 shows the evolution of this spectrum when a constant voltage is applied parallel to the magnetic field, which shifts the Fermi levels for the two chiralities by  $\pm 30$  meV. Here we assumed that before applying bias, the Fermi

energy was equal to 60 meV at both Weyl points. As is clear from Fig. 7, when a voltage is applied, an additional absorption edge appears in the spectrum for each polarization, which will be clearly distinguishable as long as the magnitude of the Fermi energy shift is larger than  $k_B T$ . Note that this behavior and the possibility of the optical detection of the chiral anomaly was predicted in Ref. [73].

In conclusion, we showed that unique topological properties of low-energy quasiparticles in WSMS give rise to a plethora of highly unusual magneto-optical effects, which provide an efficient way of studying these fascinating materials and can be utilized in future photonic devices in the terahertz through mid-infrared range. All effects are broadly tunable by varying the magnetic field, electric bias, or the propagation angle. We hope that our study will stimulate further experimental work in this rapidly developing field.

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- [1] Y. Wang, M. Tokman, and A. Belyanin, Second-order nonlinear optical response of graphene, *Phys. Rev. B* 94, 195442 (2016).
  - [2] M. Tokman, Y. Wang, I. Oladyshekin, A. Ryan Kutayiah, and A. Belyanin, Laser-driven parametric instability and generation of entangled photon-plasmon states in graphene, *Phys. Rev. B* 93, 235422 (2016).
  - [3] M. Tokman, Y. Wang, and A. Belyanin, Valley entanglement of excitons in monolayers of transition-metal dichalcogenides, *Phys. Rev. B* 92, 075409 (2015).
  - [4] D. G. Revin, M. Hemingway, Y. Wang, J.W. Cockburn, A. Belyanin, Active mode locking of quantum cascade lasers operating in external ring cavity, *Nature Comm.* 7, 11440 (2016).
  - [5] Q. Zhang, Y. Wang, W. Gao, J. D. Watson, M. J. Manfra, A. Belyanin, and J. Kono, Stability of high-density two-dimensional excitons against a Mott transition in high magnetic fields probed by coherent terahertz spectroscopy, *Phys. Rev. Lett.* 117, 207402 (2016).
  - [6] M. Tokman, Z. Long, S. AlMutairi, Y. Wang, M. Belkin, and A. Belyanin, Enhancement of the spontaneous emission in subwavelength quasi-two-dimensional waveguides and resonators, *Phys. Rev. A* 97, 043801 (2018).
  - [7] M. Tokman, Z. Long, S. AlMutairi, Y. Wang, V. Vdovin, M. Belkin, and A. Belyanin, Purcell enhancement of the photon parametric decay in two-dimensional nonlinear materials, Invited Paper, Special Issue of the APL Photonics, submitted; <https://arxiv.org/abs/1801.07227>.
  - [8] Z. Long, Y. Wang, M. Erukhimova, M. Tokman, A. Belyanin, Magneto-polaritons in Weyl semimetals in a strong magnetic field, *Phys. Rev. Lett.* 120, 037403 (2018).
  - [9] E. M. Purcell, H. C. Torrey, and R. V. Pound, *Phys. Rev.* 69, 37 (1946).
  - [10] Q. Gu, B. Slutsky, F. Vallini, J. S. T. Smalley, M. P. Nezhad, N. C. Frateschi, and Y. Fainman, *Opt. Express* 21,15603 (2013).
  - [11] C.-Y. A. Ni and S. L. Chuang, *Opt. Express* 20, 16450 (2012).
  - [12] K. Ding and C. Z. Ning, *Light: Science & Applications* (2012) 1, e20; doi:10.1038/lssa.2012.20.
  - [13] C. Walther, G. Scalari, M. Amanti, M. Beck, and J. Faist, *Science* 327, 1495 (2010).
  - [14] S. Gwo and C.-K. Shih, *Rep. Prog. Phys.* 79, 086501 (2016).
  - [15] S. V. Gaponenko, *Introduction to Nanophotonics* (Cambridge Univ. Press, 2010).
  - [16] G. M. Akselrod, C. Argyropoulos, T. B. Hoang, C. Ciraci, C. Fang, J. Huang, D. R. Smith, and M. H. Mikkelsen, *Nature Phot.* 8, 835 (2014).
  - [17] J. Lee et al., *Nature* 511, 65 (2014).
  - [18] M. Erukhimova, and M. Tokman. *Optics Lett.* 40, 2739 (2015).
  - [19] M. Erukhimova and M. Tokman. *Phys. Rev. A* 95, 013807 (2017).
  - [20] L.D. Landau and E. M. Lifshitz, *Statistical Physics, Part 1* (Butterworth-Heinemann, Oxford, 1980).
  - [21] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Atom-Photon Interactions: Basic Processes and Applications* (Wiley, New York, 1992).
  - [22] V. M. Fain and Ya. I. Khanin, *Quantum Electronics, Vol. 1* (The MIT Press, Cambridge, MA, 1969).
  - [23] M. O. Scully and M. S. Zubairy, *Quantum Optics* (Cambridge Univ. Press, Cambridge, 1997).
  - [24] V. L. Ginzburg, *Theoretical Physics and Astrophysics* (Pergamon, Oxford, 1979).
  - [25] M. D. Tokman, M. A. Erukhimova, and V. V. Vdovin, *Annals of Physics* 360, 571 (2015).
  - [26] M. Tokman, M. Erukhimova, *Journal of Lumin.* 137, 148 (2013).
  - [27] M. Tokman, X. Yao, and A. Belyanin, *Phys. Rev. Lett.* 110, 077404 (2013).
  - [28] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics* (Pergamon, Oxford, 1977).
  - [29] L. Davidovich, *Rev. Mod. Phys.* 68, 127 (1996).
  - [30] M. Tokman, Y. Wang, and A. Belyanin, *Phys. Rev. B* 92, 075409 (2015).
  - [31] J. Lee et al., *Adv. Optical Mater.* 2016, DOI: 10.1002/adom.201500723.
  - [32] L.D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon Oxford, 1984).
  - [33] M. Tokman, Y. Wang, I. Oladyshekin, A. R. Kutayiah, and A. Belyanin, *Phys. Rev. B* 93, 235422 (2016).
  - [34] G. M. Akselrod, T. Ming, C. Argyropoulos, T. B. Hoang, Y. Lin, X. Ling, D. R. Smith, J. Kong, and M. H. Mikkelsen, *Nano Lett.* 15, 3578 (2015).
  - [35] H.-B. Lin and A. J. Campillo, *Phys. Rev. Lett.* 73, 2440 (1994).
  - [36] P. Bermel, A. Rodriguez, J. D. Joannopoulos, and M. Soljacic, *Phys. Rev. Lett.* 99, 053601 (2007).

- [37] P. Genevet, J.-P. Tetienne, E. Gatzogiannis, R. Blanchard, M. A. Kats, M. O. Scully, and F. Capasso, *Nano Lett.* 10, 4880 (2010).
- [38] M. Kauranen and A. V. Zayats, *Nature Phot.* 6, 737 (2012).
- [39] M. Lapine, I. V. Shadrivov, and Y. S. Kivshar, *Rev. Mod. Phys.* 86, 1093 (2014).
- [40] J. Lee et al., *Nature* 511, 65 (2014).
- [41] D. Smirnova and Y. S. Kivshar, *Optica* 3, 1241 (2016).
- [42] A. R. Davoyan and H. A. Atwater, arXiv:1709.07028 (2017).
- [43] E. Hendry, P.J. Hale, J. Moger, A.K. Savchenko, and S.A. Mikhailov, *PRL* 105, 097401 (2010).
- [44] X. Yao and A. Belyanin, *Phys. Rev. Lett.* 108, 255503 (2012).
- [45] X. Yao, M.D. Tokman, and A. Belyanin, *Phys. Rev. Lett.* 112, 055501 (2014).
- [46] Y. Wang, M. Tokman, and A. Belyanin, *Phys. Rev. B* 94, 195442 (2016).
- [47] L.M. Malard, T.V. Alencar, A.P.M. Barboza, K.F. Mak, and A.M. de Paula, *Phys. Rev. B* 87, 201401 (2013).
- [48] H. Liu, Y. Li, Y. S. You, S. Ghimire, T. F. Heinz, and D. A. Reis, *Nat. Phys.* 13, 262 (2016).
- [49] P. Alonso-Gonzalez, et al., *Nature nanotechnology* 12, 31. 10.1038/nnano.2016.185.
- [50] Yu Ye, Z. Wong, X. Lu, X. Ni, H. Zhu, X.Chen, Y. Wang, and X. Zhang, *Nat. Phot.* 9, 733 (2015).
- [51] C. Gardiner and P. Zoller, *Quantum Noise* (Springer-Verlag, Berlin, Heidelberg, 2004).
- [52] Yu. A. Illinskii and L. V. Keldysh, *Electromagnetic Response of Material Media* (Springer, New York, 1994).
- [53] M. D. Tokman and M. A. Erukhimova, *Phys. Rev. E* 84, 056610 (2011).
- [54] Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, Hoboken, NJ, 2003).
- [55] T. Low, A. Chaves, J. D. Caldwell, A. Kumar<sup>1</sup>, N. X. Fang, P. Avouris, T. F. Heinz, F. Guinea, L. Martin-Moreno, and F. Koppens. *Nat. Materials* 16, 182 (2016).
- [56] J. U. Furst, D.V. Strekalov, D. Elser, A. Aiello, U. L. Andersen, Ch. Marquardt, and G. Leuchs, *Phys. Rev. Lett.* 105, 263904 (2010).
- [57] V. O. Martynov, V A Mironov, and L A Smirnov, *J. Phys. B: At. Mol. Opt. Phys.* 50, 085501 (2017).
- [58] X. Wan, A. M. Turner, A. Vishwanath, and S. Y. Savrasov, *Phys. Rev. B* 83, 205101 (2011).
- [59] P. Hosur and X. Qi, *Compt. Rend. Phys.* 14, 857 (2013).
- [60] O. Vafek and A. Vishwanath, *Ann. Rev. Condens. Matter Phys.* 5, 83 (2014).
- [61] S.-Y. Xu, I. Belopolski, N. Alidoust, M. Neupane, G. Bian, et al., *Science* 349, 613 (2015).
- [62] B. Q. Lv, H. M. Weng, B. B. Fu, X. P. Wang, H. Miao, et al., *Phys. Rev. X* 5, 031013 (2015).
- [63] S.-Y. Xu, N. Alidoust, I. Belopolski, Z. Yuan, G. Bian, et al., *Nat. Phys.* 11, 748 (2015).
- [64] J. Xiong, S. K. Kushwaha, T. Liang, J. W. Krizan, M. Hirschberger, W. Wang, R. J. Cava, and N. P. Ong, *Science* 350, 413 (2015).
- [65] X. Huang, L. Zhao, Y. Long, P. Wang, D. Chen, et al., *Phys. Rev. X* 5, 031023 (2015).
- [66] D. T. Son and B. Z. Spivak, *Phys. Rev. B* 88, 104412 (2013).
- [67] Q. Li, D.E. Kharzeev, C. Zhang, Y. Huang, I. Pletikosic, et al., *Nature Phys.* 12, 550 (2016).
- [68] B. Xu, Y. M. Dai, L. X. Zhao, K. Wang, R. Yang, et al., *Phys. Rev. B* 93, 121110(R) (2016).
- [69] B. Z. Spivak and A. V. Andreev, *Phys. Rev. B* 93, 085107 (2016).
- [70] J. Zhou, H. Chang, and D. Xiao, *Phys. Rev. B* 91, 035114 (2015).
- [71] F.M.D. Pellegrino, M. I. Katsnelson, and M. Polini, *Phys. Rev. B* 92, 201407(R) (2015).
- [72] C. J. Tabert, J. P. Carbotte, and E. J. Nicol, *Phys. Rev. B* 93, 085426 (2016).
- [73] P.E.C. Ashby and J. P. Carbotte, *Phys. Rev. B* 89, 245121 (2014).
- [74] J. Ma and D. A. Pesin, *Phys. Rev. B* 92, 235205 (2015).
- [75] E.V. Gorbar, V.A. Miransky, I.A. Shovkovy, and P.O. Sukachov, *Phys. Rev. B* 95, 115202 (2017).
- [76] P.E.C. Ashby and J. P. Carbotte, *Phys. Rev. B* 87, 245131 (2013).
- [77] A.A. Zyuzin and A.A. Burkov, *Phys. Rev. B* 86, 115133 (2012).
- [78] S. Dai, Q. Ma, M. K. Liu, T. Andersen, Z. Fei, et al., *Nat. Nanotech.* 10, 682 (2015).
- [79] A. Poddubny, I. Iorsh, P. Belov, and Y. Kivshar, *Nat. Phot.* 7, 958 (2013).
- [80] M. Fleischhauer, A. Imamoglu, and J. P. Marangos, *Rev. Mod. Phys.* 77, 633 (2005).
- [81] M. D. Tokman, M. A. Erukhimova, *Radiophysics and Quantum Electronics*, vol. 57, 821-836 (2015).

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**Abstract**

Six projects supported by the award were completed. They resulted in seven published papers and one more paper is submitted as an invited paper to APL-Photonics. (1) A rigorous quantum-mechanical theory of the second-order nonlinear response of two-dimensional (2D) massless Dirac fermions in graphene and topological insulators has been developed. The resulting nonlinear susceptibility tensor satisfies all symmetry and permutation properties with respect to its indices and arguments, is gauge-invariant, and has a correct quasiclassical limit for low frequencies. The theory has been applied to a number of nonlinear optical processes which form the basis of ultracompact, high-performance optoelectronic devices for ultrafast secure communications, remote sensing, and imaging. (2) A new optical mechanism of spin-valley entanglement of excitons in transition metal dichalcogenide monolayers has been proposed which can be utilized in future systems for quantum communications and information processing. (3) A theory of the third-order nonlinearity of 2D electron gas in coupled quantum well systems has been developed and applied to the four-wave mixing and phase locking of laser modes in quantum cascade lasers (QCLs). The collaborators at Sheffield experimentally demonstrated active mode locking of mid-infrared QCLs, which is a major milestone in laser science. (4) In the presence of a magnetic field, a nonperturbative theory for 2D

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magnetoexcitons interacting with terahertz radiation has been developed and applied to explain the striking stability of 2D magnetoexcitons against dissociation (the absence of the Mott transition) at high magnetic fields, observed by experimental collaborators at Rice University. (5) A strong enhancement of both spontaneous emission and nonlinear optical response of graphene and other 2D materials in subwavelength 2D cavities has been predicted and applied to the design of nonlinear photonic circuits. (6) The optical response and electrodynamic properties of massless Dirac fermions in Dirac and Weyl semimetals in a magnetic field have been investigated. The results indicate that Weyl semimetals can serve as efficient polarizers, light modulators, and natural hyperbolic materials in the terahertz and infrared spectral ranges, with applications in ultracompact photonic chips.

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[3] M. Tokman, Y. Wang, and A. Belyanin, Valley entanglement of excitons in monolayers of transition-metal dichalcogenides, Phys. Rev. B 92, 075409 (2015).

[4] D. G. Revin, M. Hemingway, Y. Wang, J.W. Cockburn, A. Belyanin, Active mode locking of quantum cascade lasers operating in external ring cavity, Nature Comm. 7, 11440 (2016).

[5] Q. Zhang, Y. Wang, W. Gao, J. D. Watson, M. J. Manfra, A. Belyanin, and J. Kono, Stability of high-density two-dimensional excitons against a Mott transition in high magnetic fields probed by coherent terahertz spectroscopy, Phys. Rev. Lett. 117, 207402 (2016).

[6] M. Tokman, Z. Long, S. AlMutairi, Y. Wang, M. Belkin, and A. Belyanin, Enhancement of the spontaneous emission in subwavelength quasi-two-dimensional waveguides and resonators, Phys. Rev. A 97, 043801 (2018).

[7] M. Tokman, Z. Long, S. AlMutairi, Y. Wang, V. Vdovin, M. Belkin, and A. Belyanin, Purcell enhancement of the photon parametric decay in two-dimensional nonlinear materials, Invited Paper, Special Issue of the APL Photonics, submitted; <https://arxiv.org/abs/1801.07227>.

[8] Z. Long, Y. Wang, M. Erukhimova, M. Tokman, A. Belyanin, Magneto-polaritons in Weyl semimetals in a strong magnetic field, Phys. Rev. Lett. 120, 037403 (2018).

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