

**Localization of phonon polaritons in disordered polar media**

Arkady M. Satanin\* and Yong S. Joe†

*Center for Computational Nanoscience, Department of Physics and Astronomy, Ball State University, Muncie, Indiana 47306, USA*

Chang Sub Kim‡

*Department of Physics and Institute for Condensed Matter Theory, Chonnam National University, Gwangju 500-757, Korea*

Mikhail I. Vasilevskiy

*Centro de Fisica, Universidade do Minho, Campus de Gualtar, 4710-057 Braga, Portugal*

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The localization of the hybrid modes of phonons and photons in polar matter is investigated in the presence of random scatterers theoretically. We employ the self-consistent generalized Born-Huang approach to derive effective equations describing the phonon-polariton fields. Based on these equations, the density of states and various localization properties are exploited in two-dimensional systems both analytically and numerically within the framework of the Anderson model with a non-Hermitian effective Hamiltonian. Consequently, it is shown that the disorder effect brings some intriguing features which include the appearance of the localized states in the polariton bottleneck in the energy spectrum and the collapse of the energy gap. In addition, an analysis is given of the polariton level-spacing distribution.

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**I. INTRODUCTION**

The optical properties of a solid material are mainly determined by the interaction of photons with certain elementary excitations. The optical response owing to electron and phonon excitations in media with intrinsic structure (such as low-dimensional systems and photonic crystals) possesses features that are attractive for optoelectronic applications because the control over the optical properties can be achieved by adjusting the geometrical parameters of the system [1]. The recently realized idea of random lasers demonstrates that the intrinsic structure can be just disorder [2]. Since the wavelengths of an incident photon and of an optical phonon eventually coupling to it are large compared to the lattice constant, it is supposed that the light and optical phonon waves can be described in a continuous approximation. It is well-known that the interaction between an electromagnetic radiation and optical phonon modes in ionic crystals (like alkali halides) and polar semiconductors (we shall use the term *polar media* as a more general one for both classes of materials) may be considered by using a phenomenological theory, for instance, the Born-Huang approach [3]. In this approach, an ion in a certain unit cell moves in a self-consistent field of all other ions. The coupling between a photon and a phonon is strong near the optical phonon-photon resonance and gives rise to a new quasiparticle, a polariton [4–6]. The polaritons are always formed in a dielectric medium when the light dispersion curve crosses that

of a transverse excitation to which it is coupled.

The linear version of the Born-Huang theory and its modifications have been extensively developed and recently applied to study the properties of optical phonons and their coupling to the electromagnetic radiation in bulk and low-dimensional structures [7–12]. The energy transport by polaritons in realistic (disordered) dielectric media can be limited by scattering processes. The scattering of polaritons on local defects has been investigated by Hopfield [13] and Maddox and Mills [7]. The concept of local polaritons (with the phonon part coupled to an impurity), introduced in Ref. [14], allowed for the explanation of weak features in the far-infrared spectra of so-called one-mode mixed crystals. However, the problem of polariton scattering on ensembles of scatterers and their localization has not been addressed yet. The Anderson localization, which stands for the phenomenon consisting in the change of the electron wave function from extended to a localized one as the magnitude of disorder is increased, has been one of the main subjects in the mesoscopic physics, intensively studied both experimentally and theoretically. Now it is well-understood that the Anderson localization is a fundamental property of waves in a disordered medium. The recent development of photonic crystals provoked increasing research activity exploring the concepts of the physics of electronic phenomena with application to classical waves [15,16]. Accordingly, there has been a growing interest in studies of localization of bosonic fields, such as electromagnetic waves [17], sound (acoustical phonons), system of coupled scalar fields [18], plasmons [19], and optical phonons [20] in strongly scattering nonelectronic materials [21]. In particular, the Anderson localization of phonons and photons in disordered matters has been considered in Refs. [17,22,23,25,26]. As far as noninteracting phonon-photon systems are concerned, the physics of localization is well-understood. The common belief based on the scaling theory arguments [16] is that, in one and two dimen-

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\*Electronic address: arkady@ipm.sci-nnov.ru; Permanent address: Institute for Physics of Microstructures, RAS, GSP-105, Nizhny Novgorod, 603950, Russia.

†Electronic address: ysjoe@bsu.edu

‡Electronic address: cskim@boltzmann.chonnam.ac.kr

sions all states are localized for any magnitude of disorder, while for three-dimensional systems there exists a nonzero strength of disorder, leading to the Anderson transition. In practice, there is a major task to investigate the localization of light because the energy of photons is always higher than the effective scattering potential in disordered media. The interaction of light with phonons gives a new tool for the realization in the regime of strong localization of light.

In this work we investigate the localization of the phonon-polaritons in disordered polar matter. To this end we first use the generalized Born-Huang approach to derive the effective field equations for polariton propagation. In this scheme the transverse optical excitation, i.e., the hybridization of photons and transverse optical phonons, is described by introducing the coupled vector-potential and mechanical displacement fields. An analytical dispersion relation for the phonon-polaritons in a homogeneous medium, following from these equations, manifests an energy gap that appears as a consequence of the fields' coupling. For an inhomogeneous system, the dielectric function and phonon-photon coupling parameter are considered as local characteristics of the medium (i.e., as random functions). It is shown that the problem of polariton localization can be formulated in terms of the Anderson model with a nonsymmetric matrix in the space of doubled dimension, which describes the coupled phonon-photon modes. We have performed an exact diagonalization of the nonsymmetric matrix for a model two-dimensional system in order to find the eigenfunctions and eigenvalues near the polariton energy gap. The density of states (DS) and some characteristics of localization have been calculated. The investigation of DS near the polariton "bottleneck" has shown that disorder results in localized modes appearing at both sides inside the polaritonic gap, which form DS tails extending far into the gap. As the magnitude of disorder increases, the tails overlap and the gap collapses. To examine the localization of the coupled phonon-photon fields, we investigated the behavior of the so-called generalized participation ratio, calculated from the eigenstates obtained numerically. In addition, the nearest-neighbor level statistics have been studied. Although generally the polariton localization is enhanced over broad energy ranges as the magnitude of disorder increases, we found a different behavior for the level statistics of the upper and lower polariton branches. While the level distribution for the upper branch is a typical Wigner function, the level distribution for the lower one has a long tail for large energy intervals between subsequent polariton levels. A correlation between this behavior and the DS peak near the edge of the lower polariton band has been found.

The paper is organized as follows. The coupled field equations are derived and solved for the case of homogeneous medium in Sec. II. In Sec. III, the model of disorder is explained, the matrix form of the equations is introduced, and some analytical results are presented. In Sec. IV, we perform the exact diagonalization for two-dimensional systems, find the eigenfunctions and eigenvalues near the polaritonic gap, and calculate and discuss DS and some characteristics of localization. Section V is devoted to conclusions.

## II. GENERAL THEORY OF PHONON POLARITONS

### A. Model and basic equations

Let us consider a polar medium, i.e., an ionic crystal or a polar semiconductor with two ions per unit cell. These ions possess a certain effective charge (known as transverse charge). The medium has a background dielectric constant  $\epsilon_\infty$ . We suppose for simplicity that the medium is isotropic. In the spirit of Born and Huang's ideas [3], we shall assume that (i) the wavelength of light ( $\lambda$ ) is much larger than the lattice constant ( $a$ ), (ii) each ion moves in a self-consistent electromagnetic field created by all other ions, and (iii) the coupling of the electromagnetic field to the mechanical displacement can be considered in the dipole approximation, so that it is sufficient to consider only the relative displacements of two ions in the same unit cell. The coupled mechanical and electromagnetic equations of motion can be derived using a Lagrangian of the system, which can be constructed from independent quadratic invariants of two vector fields, the relative displacement between two ions  $\mathbf{u}(\mathbf{r}, t)$  and the electromagnetic field  $\mathbf{E}(\mathbf{r}, t)$  (and their time and spaces derivatives) [13,27]. The generalized Born-Huang field equations are obtained in a straightforward way using the variational principle,

$$\delta \int dt dV \mathcal{L} = 0, \quad (1)$$

where  $\mathcal{L}$  is the Lagrangian density. One of the advantages of this method is that any loss mechanisms may be included into consideration by using the Rayleigh dissipative function [28,30].

It is convenient to decompose the displacement and electric field into transverse and longitudinal components. We choose the Coulomb gauge ( $\nabla \cdot \mathbf{A} = 0$ ), so that

$$\mathbf{E} = -\frac{1}{c} \frac{\partial \mathbf{A}}{\partial t}, \quad \mathbf{H} = \nabla \times \mathbf{A}, \quad (2)$$

where  $\mathbf{H}$  is the magnetic field. Note that in the Coulomb gauge the vector potential is a purely transverse field. In this work we shall neglect the mixing of transverse (TO) and longitudinal (LO) optical phonon modes.

The Lagrangian density for transverse fields can be written as [29]

$$\mathcal{L} = \frac{1}{2} \left[ \rho \left( \frac{\partial \mathbf{u}}{\partial t} \right)^2 + \Lambda (\nabla \mathbf{u})^2 - \gamma \mathbf{u}^2 \right] + \frac{1}{8\pi} \left[ \frac{\beta}{c^2} \left( \frac{\partial \mathbf{A}}{\partial t} \right)^2 - (\nabla \times \mathbf{A})^2 \right] - \frac{\alpha}{c} \mathbf{u} \cdot \frac{\partial \mathbf{A}}{\partial t}. \quad (3)$$

The three terms in the Lagrangian (3) represent the mechanical part, the electromagnetic field, and the interaction between fields (in the dipole approximation), respectively. The mechanical properties of the medium are characterized essentially by two parameters, the reduced mass density  $\rho$ , and an effective elastic constant  $\gamma = \omega_{TO}^2 \rho$ . As it is usual for macroscopic models [8,11,12], we take into account the spatial dispersion of phonons by using a phenomenological parameter  $\Lambda$ , which characterizes the curvature of the TO phonon

branch in the long wavelength limit. Since we are going to consider only nonmagnetic systems, the electromagnetic properties of the medium are characterized only by a local high-frequency dielectric constant  $\beta$  (for a homogeneous medium  $\beta = \varepsilon_\infty$ ). The coupling between the mechanical and electromagnetic field is characterized by a parameter  $\alpha$  (in the absence of disorder) given by

$$\alpha^2 = \frac{(\varepsilon_0 - \varepsilon_\infty)\gamma}{4\pi},$$

where  $\varepsilon_0$  is the low-frequency dielectric constant. We would like to emphasize that this restricted model is reasonable for light frequencies close to  $\omega_{TO}$ , which is the region of primary concern in many experimental situations. In general, the optical excitations can interact with acoustical phonons giving rise to decay of the excitations and, consequently, to dissipation of the electromagnetic field energy. If this interaction is important, the resonant optical system is coupled to a non-resonant dissipative system. As it has been mentioned above, this effect can be incorporated by introducing the Rayleigh dissipative function [30]:

$$R = \frac{\rho\nu_u}{2}\dot{\mathbf{u}}^2 + \frac{\rho\nu_A}{2c}\dot{\mathbf{A}}^2, \quad (4)$$

where  $\nu_u$  and  $\nu_A$  are decay parameters for optical phonons and electromagnetic field, respectively.

Using Eqs. (1), (3), and (4), we obtain the coupled equations of motion for two harmonic fields with damping:

$$\rho \frac{\partial^2 \mathbf{u}}{\partial t^2} + \rho\nu_u \frac{\partial \mathbf{u}}{\partial t} + \Lambda \nabla^2 \mathbf{u} + \gamma \mathbf{u} = -\frac{\alpha}{c} \frac{\partial \mathbf{A}}{\partial t}, \quad (5)$$

$$\frac{\beta}{c^2} \frac{\partial^2 \mathbf{A}}{\partial t^2} + \frac{\rho\nu_A}{c} \frac{\partial \mathbf{A}}{\partial t} - \nabla^2 \mathbf{A} = \frac{4\pi\alpha}{c} \frac{\partial \mathbf{u}}{\partial t}. \quad (6)$$

### B. Polariton excitations in homogeneous media

In an isotropic medium without losses ( $\nu_u = \nu_A = 0$ ) and disorder, Eqs. (5) and (6) yield usual harmonic waves,

$$\mathbf{u}(\mathbf{r}, t) = \mathbf{u}_0 e^{i(\mathbf{k}\cdot\mathbf{r} - \omega t)},$$

$$\mathbf{A}(\mathbf{r}, t) = \mathbf{A}_0 e^{i(\mathbf{k}\cdot\mathbf{r} - \omega t)},$$

where  $\mathbf{u}_0$  and  $\mathbf{A}_0$  are the amplitudes. Substituting these expressions into Eqs. (5) and (6) and neglecting for simplicity the phonon dispersion we can easily find the dispersion relation for polaritons in the form

$$\omega_{\pm}^2(k) = \frac{1}{2}(\omega_{LO}^2 + \varepsilon_\infty^{-1}c^2k^2 \pm \sqrt{(\omega_{LO}^2 + \varepsilon_\infty^{-1}c^2k^2)^2 - 4\varepsilon_\infty^{-1}\omega_{TO}^2c^2k^2}), \quad (7)$$

where the Lyddane-Sachs-Teller relation [3]  $\omega_{LO} = \sqrt{\varepsilon_0/\varepsilon_\infty}\omega_{TO}$  has been used,  $\omega_+(k)$  and  $\omega_-(k)$  are the upper and lower branches of polariton excitations.

We would like to recall that phonon polaritons are admixture of electromagnetic and mechanical fields. A simple

physical picture for these excitations may be obtained from the analogy with the classical coupling of two harmonic oscillators of frequencies  $\omega_1$  and  $\omega_2$ . Without coupling, the oscillators vibrate at these independent frequencies. However, if the oscillators are connected by a spring, they no longer vibrate with the frequencies  $\omega_1$  and  $\omega_2$ . Instead, two different coupled vibrational modes arise, corresponding to two polariton modes in the present case.

The linear interaction of two harmonic fields in Eqs. (5) and (6) can be described by a dimensionless parameter

$$\bar{\alpha} = \alpha \sqrt{\frac{4\pi}{\gamma\varepsilon_\infty}} = \sqrt{\frac{\varepsilon_0 - \varepsilon_\infty}{\varepsilon_\infty}}.$$

Two regimes can be pointed out, according to the value of this parameter with respect to unity, which can be designated as strong and weak coupling. Anticipating the consideration of disorder, we can expect that for  $\alpha \ll \sqrt{\gamma\beta/4\pi}$ , the system of photons and phonons is essentially noninteracting and all the well-known results [21] can be applied to the localization of two independent boson fields. In the opposite case of  $\alpha \gg \sqrt{\gamma\beta/4\pi}$ , one can expect that the formation of a strongly coupled polariton can result in significantly new physics.

## III. DISORDERED MEDIA

### A. Model of disorder

When a polariton wave propagates through a real crystal, it is deflected from its initial direction by any static defect that may be present. So, the motion of a polariton in a disordered region of the crystal is such that the polariton is strongly scattered. In fact, the variation of any of the parameters that enter Eqs. (5) and (6) can affect the polariton propagation. Scattering of the polariton on fluctuations of these parameters may give rise to localized modes. One can expect that disorder can reduce the width of the polariton gap and even lead to its collapse when the amplitude of the fluctuations is increased. At this point, we shall specify the model of the scatterers. Following the work of Maddox and Mills [7], we assume that defects in the polar medium can be modeled by a local deviation of the mechanical and dielectric constants and the coupling parameter  $\alpha$ . We shall consider three fluctuating parameters, (i)  $\omega_{TO}$  (or, equivalently,  $\gamma$  since  $\rho$  will be considered constant), which should lead to the localization of phonons, (ii)  $\beta$  varying around  $\varepsilon_\infty$ , which should affect the photons, and (iii)  $\alpha$ , which defines the phonon-photon interaction and therefore influences both of them. In a disordered medium, these parameters will be taken as random functions of the spatial coordinates. So, we shall consider

$$\gamma(\mathbf{r}) = \langle \gamma \rangle + \delta\gamma(\mathbf{r}),$$

$$\beta(\mathbf{r}) = \langle \beta \rangle + \delta\beta(\mathbf{r}),$$

$$\alpha(\mathbf{r}) = \langle \bar{\alpha} \rangle + \delta\alpha(\mathbf{r}),$$

where  $\langle (\dots) \rangle$  denotes the corresponding mean values ( $\langle \beta \rangle = \varepsilon_\infty$  and  $\langle \gamma \rangle = \rho\omega_{TO}^2$ ). Generally speaking, these param-

eters are not totally independent. There are rather complex relations between them, which cannot be adequately established at the phenomenological level. At this stage, we shall neglect any correlation between the fluctuating parameters. The role of such correlations will be considered in a future work.

We have extended the Lagrange approach to inhomogeneous media where the random functions introduced above vary at a scale much smaller than the photon and phonon wavelengths. Consequently, we have obtained equations coinciding in form with Eqs. (5) and (6). Neglecting the dissipation, we seek their solutions in the form:

$$\mathbf{u}(\mathbf{r}, t) = \mathbf{u}(\mathbf{r})e^{-i\omega t}, \quad \mathbf{A}(\mathbf{r}, t) = \mathbf{A}(\mathbf{r})e^{-i\omega t},$$

where  $\mathbf{u}(\mathbf{r})$  and  $\mathbf{A}(\mathbf{r})$  are the spatially dependent amplitudes of the fields, which can be found from the following equations:

$$-\Lambda \nabla^2 \mathbf{u} + (\rho\omega^2 - \gamma)\mathbf{u} = -i\frac{\alpha}{c}\omega\mathbf{A}, \quad (8)$$

$$\nabla^2 \mathbf{A} + \frac{\beta\omega^2}{c^2}\mathbf{A} = \frac{4\pi\alpha i\omega}{c}\mathbf{u}. \quad (9)$$

### B. Matrix form of the equations of motion

In order to symmetrize Eqs. (8) and (9), we make a transformation,

$$\mathbf{u} \rightarrow \frac{1}{\sqrt{\rho}}\mathbf{u}, \quad \mathbf{A} \rightarrow c\sqrt{\frac{4\pi}{\rho\beta}}\mathbf{A}.$$

Then, the resulting equations for the coupled fields can be written as follows:

$$\frac{\Lambda}{\sqrt{\rho}}\nabla^2 \frac{1}{\sqrt{\rho}}\mathbf{u}(\mathbf{r}) + \frac{\gamma}{\rho}\mathbf{u}(\mathbf{r}) = \omega^2\mathbf{u} + i\omega\alpha\sqrt{\frac{4\pi}{\rho\beta}}\mathbf{A}(\mathbf{r}), \quad (10)$$

$$-\frac{c^2}{\sqrt{\beta}}\nabla^2 \frac{1}{\sqrt{\beta}}\mathbf{A}(\mathbf{r}) = \omega^2\mathbf{A}(\mathbf{r}) - i\omega\alpha\sqrt{\frac{4\pi}{\rho\beta}}\mathbf{u}(\mathbf{r}). \quad (11)$$

Next, we find it more convenient to rewrite the above equations in the form

$$\mathcal{H}_u \mathbf{u}(\mathbf{r}) = \omega^2 \mathbf{u}(\mathbf{r}) + i\omega \mathcal{V}_{uA} \mathbf{A}(\mathbf{r}), \quad (12)$$

$$\mathcal{H}_A \mathbf{A}(\mathbf{r}) = \omega^2 \mathbf{A}(\mathbf{r}) - i\omega \mathcal{V}_{Au} \mathbf{u}(\mathbf{r}), \quad (13)$$

where definitions have been made for the operators,

$$\mathcal{H}_u \equiv \frac{\Lambda}{\sqrt{\rho}}\nabla^2 \frac{1}{\sqrt{\rho}} + \frac{\gamma}{\rho}, \quad \mathcal{V}_{uA} \equiv \alpha\sqrt{\frac{4\pi}{\rho\beta}}, \quad (14)$$

$$\mathcal{H}_A \equiv -\frac{c^2}{\sqrt{\beta}}\nabla^2 \frac{1}{\sqrt{\beta}}, \quad \mathcal{V}_{Au} = \mathcal{V}_{uA}. \quad (15)$$

We can eliminate one of the variables from Eqs. (12) and (13), for instance,  $\mathbf{A}$ . Then we arrive at a quadratic algebraic eigenvalue problem,

$$(\mathcal{P}_u - \epsilon \mathcal{Q}_u + \epsilon^2 I)\mathbf{u} = \mathbf{0}, \quad (16)$$

where  $\mathcal{P}_u$  and  $\mathcal{Q}_u$  are operators which are defined by

$$\mathcal{P}_u = \mathcal{V}_{uA} \mathcal{H}_A \mathcal{V}_{uA}^{-1} \mathcal{H}_u, \quad (17)$$

$$\mathcal{Q}_u = \mathcal{H}_u + \mathcal{V}_{uA} \mathcal{H}_A \mathcal{V}_{uA}^{-1} + \mathcal{V}_{uA} \mathcal{V}_{Au}, \quad (18)$$

where  $I$  is the identity operator and a new spectral variable  $\epsilon = \omega^2$  has been introduced. If, instead, we eliminate  $\mathbf{u}$  from Eqs. (12) and (13), we get

$$(\mathcal{P}_A - \epsilon \mathcal{Q}_A + \epsilon^2 I)\mathbf{A} = \mathbf{0}, \quad (19)$$

where  $\mathcal{P}_A$  and  $\mathcal{Q}_A$  are defined as

$$\mathcal{P}_A = \mathcal{V}_{Au} \mathcal{H}_u \mathcal{V}_{Au}^{-1} \mathcal{H}_A, \quad (20)$$

$$\mathcal{Q}_A = \mathcal{H}_A + \mathcal{V}_{Au} \mathcal{H}_u \mathcal{V}_{Au}^{-1} + \mathcal{V}_{Au} \mathcal{V}_{Au}. \quad (21)$$

Thus we have constructed a nonlinear eigenvalue problem [31] which occurs for systems of coupled differential equations when the coupling between them depends on the derivatives. For example, the nonlinear eigenvalue problem is encountered for coupled circuits of capacitors, coils, and resistors. It is worth mentioning that a similar problem arises also in the scattering theory [32,33]. From the theory of matrices it is known that, when  $\mathcal{P}$  and  $\mathcal{Q}$  are symmetric, Eqs. (16) and (19) have positive eigenvalues [31], corresponding to nondecaying harmonic modes.

Further, we find that it is possible to linearize the eigenvalue problem by doubling the dimension of the system. For instance, we can rewrite Eq. (16) in the form

$$I\mathbf{v} = \epsilon\mathbf{u}, \quad (22)$$

$$-\mathcal{P}_u \mathbf{u} + \mathcal{Q}_u \mathbf{v} = \epsilon \mathbf{v}, \quad (23)$$

where  $\mathbf{v} = \epsilon\mathbf{u}$ . Now we can rewrite Eq. (22) in the matrix form,

$$\mathcal{M}_u \begin{pmatrix} \mathbf{u} \\ \mathbf{v} \end{pmatrix} = \epsilon \begin{pmatrix} \mathbf{u} \\ \mathbf{v} \end{pmatrix}, \quad (24)$$

where

$$\mathcal{M}_u = \begin{pmatrix} 0 & I \\ -\mathcal{P}_u & \mathcal{Q}_u \end{pmatrix}. \quad (25)$$

After we find the eigenvectors  $\mathcal{M}_u$ , we obtain  $\mathbf{u}$  as an upper part of the eigenvector column. The eigenvectors  $\mathbf{A}$  can be extracted from the relation

$$\mathbf{A} = \frac{1}{i\sqrt{\epsilon}} \mathcal{V}_{Au}^{-1} (\mathcal{H}_A - \epsilon)\mathbf{u}. \quad (26)$$

Thus we solve the eigenvalue problem for a nonsymmetric matrix Eq. (25), as an alternative way of solving Eq. (16).

### C. Perturbation theory analysis

Let us recall first that, if  $\alpha=0$ , i.e., when there are two independent (bosonic) fields, the localization of both

phonons and photons is similar to that of electrons in a random potential. It is generally accepted that all the photon and phonon states are localized in one- and two-dimensional systems. Hence it is possible to check whether there is Anderson localization of photons or phonons in the systems by considering random and real symmetric matrices  $\mathcal{H}_u$  and  $\mathcal{H}_A$  (with  $\alpha=0$ ).

There are two types of localized states, commonly referred to as strongly and weakly localized ones [16]. The localization of light and phonons may be characterized by the mean free path  $l_{light}$  and  $l_{ph}$ , respectively. For instance, the mean free path of light in a three-dimensional dielectric medium with an array of embedded short range scatterers (whose radii  $r$  is  $r \ll \lambda$ ) can be estimated in terms of the difference of dielectric function  $\delta\epsilon$  and the density  $n_s$ :  $l_{light} \sim (c/\omega)^4/n_s(\delta\epsilon)^2 r^6$ . The localization occurs when the characteristic spatial scale of coherent multiple scattering (mean free path) becomes of the order of the wavelength,  $l_{light} \sim \lambda$ . For almost dispersionless optical phonons the localization length may be of the order of lattice constant,  $l_{ph} \sim a$ . More details concerning the localization of optical phonons and photons in disordered media can be found in Refs. [23,24] and Refs. [17,34], respectively.

Here, we consider the situation that the photon-phonon coupling is weak ( $\alpha < \sqrt{\gamma\beta/4\pi}$ ) for a perturbative analysis. In order to understand qualitatively what happens to the polaritons of the lower ("phonon") branch, we rewrite Eqs. (12) and (13) in the following form:

$$\mathcal{H}_u \mathbf{u}(\mathbf{r}) - \epsilon \mathbf{u}(\mathbf{r}) = \epsilon \mathcal{V}_{uA} \frac{1}{\mathcal{H}_A - \epsilon} \mathcal{V}_{Au} \mathbf{u}(\mathbf{r}). \quad (27)$$

Suppose that we know (localized) eigenfunctions of the operator  $\mathcal{H}_u$ ,

$$\mathcal{H}_u \mathbf{u}_j(\mathbf{r}) = \epsilon_j \mathbf{u}_j(\mathbf{r}). \quad (28)$$

Then, the matrix elements of the perturbation operator [right-hand side of Eq. (27)] between these eigenstates can be easily calculated (although it is necessary to take into account the singular character of the perturbation operator). Extracting the imaginary part, we find the decay rate  $\Gamma_j$  of a state  $\mathbf{u}_j(\mathbf{r})$  as

$$\begin{aligned} \Gamma_j &= \pi \epsilon_j \langle j | \mathcal{V}_{uA} \delta(\mathcal{H}_A - \epsilon_j) \mathcal{V}_{Au} | j \rangle \\ &= \pi \epsilon_j \sum_a |\langle j | \mathcal{V}_{uA} | a \rangle|^2 \delta(E_A^a - \epsilon_j), \end{aligned} \quad (29)$$

where  $E_A^a$  and  $|a\rangle$  are the eigenvalues and eigenvectors of  $\mathcal{H}_A$ . Thus the phonon eigenmodes decay as a consequence of the interaction with the photon bath. In turn, photons also decay via interaction with phonons. If, for instance, photons are not localized in the disordered medium under consideration, they can transport energy from one localized phonon mode to another facilitating their delocalization.

Thus in disordered polar media there are weakly localized photons and strongly localized phonons for the frequencies far from gap. Near the polaritonic gap where the coupling is strong the perturbative approach cannot be used to describe the localization of polariton.

#### IV. SPECTRAL CHARACTERISTICS OF THE DISORDERED SYSTEM

We investigate here some characteristics of the phonon-polaritons in a two-dimensional system numerically, which are the density of states, the participation ratio, and the distribution function of the levels. The continuous, partial differential equations are discretized into algebraic ones and then are solved by the standard techniques. In a disordered system, the phonon and photon momenta are not conserved and Bloch theorem cannot be used. Accordingly, one has to invoke a sufficiently large supercell with appropriate boundary conditions. One frequently used scheme is to impose the quasiperiodical boundary condition, i.e., to study a superlattice of identical disordered supercells, which has been used in our calculations.

##### A. Numerical method for discrete model of inhomogeneous medium

We consider a two-dimensional system where the parameters depend on the coordinates  $(x, y)$ . The continuum space is divided into a mesh. We choose a mesh with sites  $(x_n, y_m)$ ,  $x_n = n\Delta$  and  $y_m = m\Delta$  ( $n, m = 1, 2, \dots, N$ ), where  $\Delta$  is the minimal distance. The relative displacement and the vector potential at various mesh points are denoted by  $\mathbf{u}_{n,m} = \mathbf{u}(x_n, y_m)$  and  $\mathbf{A}_{n,m} = \mathbf{A}(x_n, y_m)$ . We may interpret them as generalized coordinates of the system in a phase space of dimension  $2N \times 2N$ . The Hamiltonian operators  $\mathcal{H}_u$  and  $\mathcal{H}_A$  should be considered now in the discrete space as

$$(\mathcal{H}_u \mathbf{u})_{n,m} = \frac{\Lambda}{\sqrt{\rho_{n,m} \rho_{n',m'}}} \sum (\nabla^2)_{n,m;n',m'} \frac{1}{\sqrt{\rho_{n',m'}}} \mathbf{u}_{n',m'}, \quad (30)$$

$$(\mathcal{H}_A \mathbf{A})_{n,m} = - \frac{c^2}{\sqrt{\beta_{n,m} \beta_{n',m'}}} \sum (\nabla^2)_{n,m;n',m'} \frac{1}{\sqrt{\beta_{n',m'}}} \mathbf{A}_{n',m'}, \quad (31)$$

where

$$\begin{aligned} (\nabla^2)_{n,m;n',m'} &= \frac{1}{\Delta^2} [(\delta_{n,n'+1} + \delta_{n,n'-1} - 2\delta_{n,n'}) \delta_{m,m'} + (\delta_{m,m'+1} \\ &\quad + \delta_{m,m'-1} - 2\delta_{m,m'}) \delta_{n,n'}]. \end{aligned} \quad (32)$$

Note that the symmetric interaction operator has only diagonal elements

$$(\mathcal{V}_{uA})_{n,m;n',m'} = \alpha_{n,m} \sqrt{\frac{4\pi}{\rho_{n,m} \beta_{n,m}}} \delta_{n,m;n',m'}. \quad (33)$$

The operators  $\mathcal{P}_u$  and  $\mathcal{Q}_u$  may be found from Eqs. (17) and (18) using a simple matrix algebra. After that, we obtain the matrix  $\mathcal{M}_u$ . Now we can solve numerically the eigenvalue problem which is defined by Eq. (24) using a standard routine.

First, we consider the case when there is no disorder in the system. In this case, we can seek for the solutions of discretized Eq. (24) in the form

$$\mathbf{u}_{n,m} = \mathbf{u}_0 e^{i(k_x n + k_y m)\Delta}, \quad \mathbf{v}_{n,m} = \mathbf{v}_0 e^{i(k_x n + k_y m)\Delta}.$$

After simple manipulations, we obtain the dispersion relation,

$$[\omega_{ph}^2(k) - \epsilon][\omega_l^2(k) - \epsilon] - \epsilon\omega_{TO}^2\bar{\alpha}^2 = 0. \quad (34)$$

Here, the dispersions for phonons

$$\omega_{ph}^2(k) = \omega_{TO}^2 + 2\frac{\Lambda}{\rho\Delta^2}(2 - \cos k_x\Delta - \cos k_y\Delta), \quad (35)$$

and for photons

$$\omega_l^2(k) = 2\frac{c^2}{\beta\Delta^2}(2 - \cos k_x\Delta - \cos k_y\Delta) \quad (36)$$

have been introduced, respectively. If  $\bar{\alpha}=0$  and  $\Delta$  tends to zero, we recover the standard dispersion relations for both waves. It is clear that when  $\Delta$  is small compared to the wavelength ( $\Delta \ll \lambda$ ), the field modes of the discrete model can be exactly described by the excitations of the continuous system. The generalization of the above equations for a three-dimensional system is straightforward.

When the disorder is taken into account, one has to seek for the full numerical treatment. For the numerical simulation we will further simplify our model by assuming that only waves with one particular polarization are excited in the system, i.e., we will consider just scalar waves. We then set up the non-Hermitian matrix Eq. (25) and solve the eigenvalue problem. The eigenvectors  $U_j(i)$  and eigenvalues  $\epsilon_j$  for such a generalized Anderson model can be obtained by a numerical diagonalization using one of the standard algorithms.

### B. Density of states

Once the eigenvalue problem of Eq. (24) is solved, the polariton DS can be calculated as

$$g(\epsilon) = \sum_j \delta(\epsilon - \epsilon_j). \quad (37)$$

The density of states is a self-averaging quantity, so, taking the average of Eq. (37) over different disorder realizations is meaningful and represents the properties of the system in the thermodynamical limit.

Before presenting the numerical results, we shall obtain an approximate expression for the density of states, valid for the energy region near the gap, for a nearly homogeneous system. It will help us to identify the effects introduced by disorder. For the sake of simplicity, we will neglect the optical phonon dispersion and consider the limit of  $\Delta \rightarrow 0$ . In this case, instead of Eq. (34), we obtain:

$$(v^2k^2 - \epsilon)(\epsilon_{TO} - \epsilon) - \epsilon\omega_{TO}^2\bar{\alpha}^2 = 0, \quad (38)$$

where  $\epsilon_{TO} = \omega_{TO}^2$  and  $v = c/\sqrt{\beta}$ . This expression can be used near the bottleneck, for a narrow spectral interval near the edge,  $\delta\epsilon = \alpha^2\Delta^2\epsilon_{TO}/v^2\pi^2 \ll \epsilon_{TO}$ , corresponding to the photon wave vectors  $k \leq \pi/\Delta$ . Using Eq. (38), we find the number of states for the two-dimensional system as

$$N(\epsilon) = \frac{S}{4\pi v^2} k^2(\epsilon), \quad (39)$$

where  $S$  is the area. A simple calculation gives

$$N(\epsilon) = \frac{S\epsilon}{4\pi v^2} \begin{cases} \frac{\epsilon_{LO} - \epsilon}{\epsilon_{TO} - \epsilon}, & \epsilon < \epsilon_{TO} - \delta\epsilon \\ 0, & \epsilon_{TO} < \epsilon < \epsilon_{LO} \\ \frac{\epsilon - \epsilon_{LO}}{\epsilon - \epsilon_{TO}}, & \epsilon > \epsilon_{LO}. \end{cases} \quad (40)$$

Then, the density of states is given as

$$g(\epsilon) = \frac{\partial N(\epsilon)}{\partial \epsilon} = \frac{S}{4\pi v^2} \begin{cases} \frac{\epsilon^2 - 2\epsilon\epsilon_{TO} + \epsilon_{TO}\epsilon_{LO}}{(\epsilon_{LO} - \epsilon)^2}, & \epsilon < \epsilon_{TO}; \quad \epsilon > \epsilon_{LO} \\ 0, & \epsilon_{TO} < \epsilon < \epsilon_{LO}, \end{cases} \quad (41)$$

where  $\epsilon_{LO} = \epsilon_{TO} + \bar{\alpha}^2$ .

We adopted the material parameters of the polar semiconductor GaP in our calculations, taking  $\epsilon_0 = 10.20$ ,  $\epsilon_\infty = 8.50$ ,  $\omega_{TO} = 11.1$  THz, and  $\omega_{LO} = 12.1$  THz. We used 1 (THz)<sup>2</sup> as a unit of the spectral parameter. The dimensionless interaction parameter is  $\bar{\alpha} = 4.96$ . A square net with the dimensions  $N \times N = 40 \times 40$  was used, so, the size of the matrix  $\mathcal{M}$  was  $\mathcal{N} \times \mathcal{N} = 3200 \times 3200$ , where  $\mathcal{N} = 2N^2$ . In order to test our program, we compared the numerical and analytical results for the density states for the homogeneous system. The result is shown in Fig. 1. As it can be seen from Fig. 1(b), Eq. (41) fits quite well the numerical data near the gap (the interval  $121.0 < \epsilon < 145.2$ ). It is worth noting that DS shows a peak near the center of the upper band (close to  $\epsilon = 600$ ). This reflects the well-known van-Hove DS singularity in 2D systems. The peak of the density of states at the lower edge of the gap is another example of such singularity. Contrary to the former, which appears only in the discrete model, the latter is characteristic of the 2D polariton system.

Now we shall focus on the spectral region near the gap and consider the effect of disorder. The uncorrelated (dimensionless) random parameters were distributed with equal probabilities within the intervals,

$$0 < \delta\alpha(n,m) < \delta\alpha,$$

$$0 < \delta\beta(n,m) < \delta\beta,$$

where  $\delta\alpha$  and  $\delta\beta$  are parameters that specify the magnitude of disorder. The intervals of variation of the fluctuating parameters are restricted by their values for the constituents of the disordered medium. For instance, when a two-component composite medium is considered, the magnitude of fluctuations can be estimated as the difference between the values of these parameters between the host material and the inclusions. The phonon frequency  $\omega_{TO}$  is kept constant for a while. Two random parameters play different roles in the localization. It is clear that the fluctuations of  $\alpha$  should

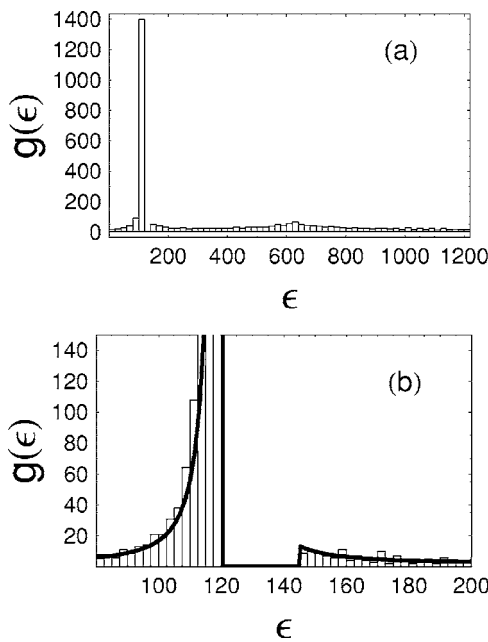


FIG. 1. (a) Polariton DS calculated for a perfect 2D system showing a gap between  $\epsilon=121.0$  and  $145.2$ . (b) A fragment of the histogram of (a) and approximate analytical function (solid line) for the density of states near the gap.

modulate the gap, while  $\beta$  determines mostly DS in the upper band. Let us investigate their effect on the density of states near the polariton “bottleneck.”

Numerical results for DS, averaged over 16 realizations, are presented in Fig. 2 for different magnitudes of the dimensionless coupling constant fluctuations,  $\delta\alpha=0.25, 0.5, 0.75,$  and  $1.0$ , while fixing the dielectric constant. One can see that, when the magnitude of disorder increases, the polariton modes appear inside the gap forming tails of the density of states at both sides of the gap. If the fluctuations of the dielectric constant are also taken into account (Fig. 3), the tails overlap and the gap collapses, meaning that the states become available in the gap. The collapse of the gap can be seen better in Fig. 4, where the eigenvalues are depicted in ascending order for a typical realization of the random parameters. One can observe clearly that the energy levels ap-

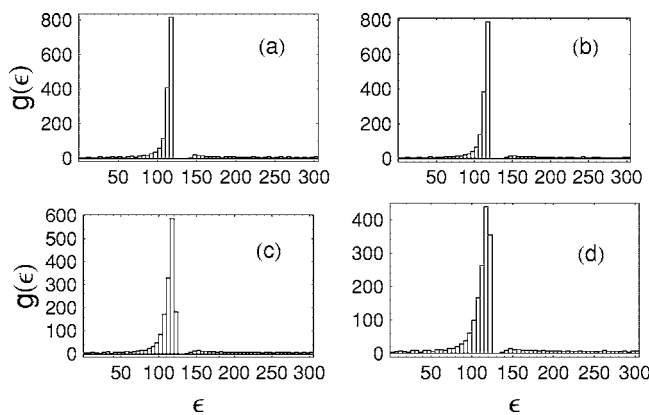


FIG. 2. DS calculated for different values of the dimensionless magnitude of disorder,  $\delta\alpha=(a) 0.25,$  (b)  $0.5,$  (c)  $0.75,$  and (d)  $1.0$ .

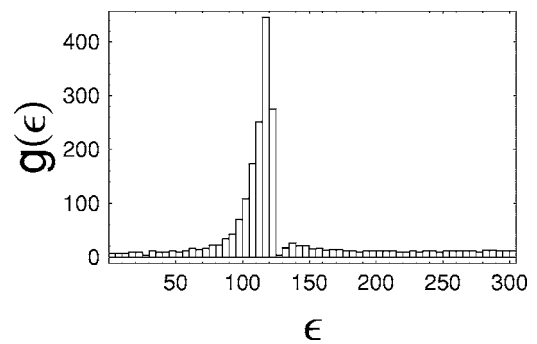


FIG. 3. DS for a strongly disordered system with  $\delta\alpha=1.0$  and  $\delta\beta=1.0$ .

pear in the interval  $121.0 < \epsilon < 145.2$ . It should be pointed out that the DS peak at the edge of the lower polariton band is decreased with disorder but remains large. It will be shown in the next section that this is important for the level distributions.

### C. Participation ratio

Let us now examine the localization of the polariton modes near the gap. It can be tested by calculating the participation ratio (PR) of each mode  $p(\epsilon_j)$ , defined as

$$p(\epsilon_j) = \left( \sum_i U_j^A(i) \right)^{-1}, \quad (42)$$

where the eigenvector  $U_j(i)$  corresponds to an eigenvalue  $\epsilon_j$  of the matrix  $\mathcal{M}_u$ . Alternatively, we could introduce PR for A-type eigenmodes in the same manner. The sum in Eq. (42) runs over all sites of the generalized (doubled) lattice. The participation ratio is usually considered as a measure of localization in numerical studies of disordered systems. In our case,  $p \sim \mathcal{N}$  for a completely extended state,  $p \sim 1$  for a state confined to a single site, and, approximately,  $p \sim M$  for a state which is extended over  $\sim M$  sites in the supercell. Since we are considering a nonlinear eigenvalue problem (equivalent to the original problem of two coupled fields), each eigenvector has the structure,

$$U_j(i) = \begin{pmatrix} u_j(i) \\ \epsilon_j \mu_j(i) \end{pmatrix}. \quad (43)$$

Using Eq. (43), we can recast Eq. (42) into a simpler form,

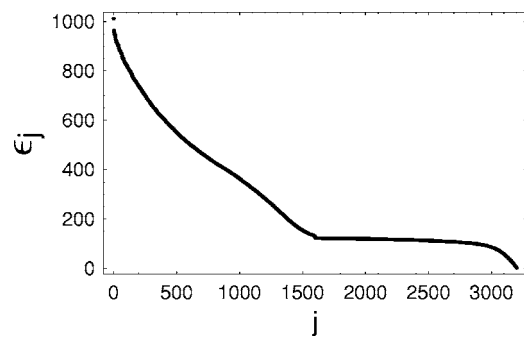


FIG. 4. Eigenvalues in ascending order for one typical disorder realization with  $\delta\alpha=1.0$  and  $\delta\beta=1.0$ .

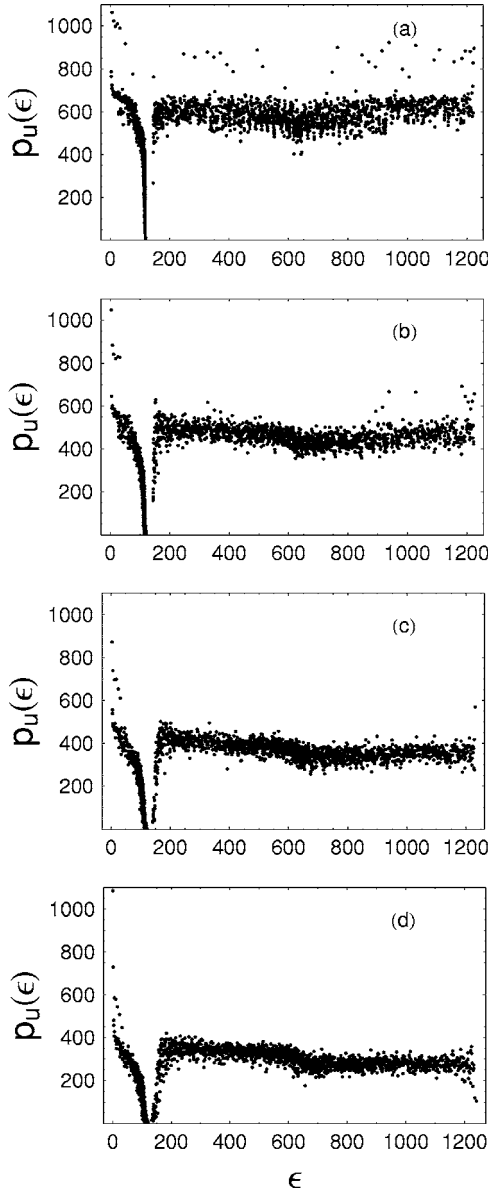


FIG. 5. Participation ratio of polariton modes for different magnitudes of coupling constant disorder,  $\delta\alpha =$  (a) 0.25, (b) 0.5, (c) 0.75, and (d) 1.0 with  $\delta\beta = 0$ .

$$p(\epsilon_j) = \frac{1}{1 + \epsilon_j^4 \left( \sum_i u_j^4(i) \right)^{-1}}, \quad (44)$$

where  $u_j(i)$  is a  $u$ -type eigenvector of Eq. (24).

Figure 5 demonstrates the behavior of PR with the increase of disorder. The fluctuations of  $\alpha$  give rise to the localized states in the gap. Moreover, they also result in the localization of practically all the polariton modes distributed over a very broad energy range. Adding disorder of the dielectric constant strongly enhances the localization of the short-wavelength excitations in the upper polariton band, as it can be seen in Fig. 6.

#### D. Statistics of spectral intervals

An alternative way to examine the localization, based on the random matrix theory [35], is to study the statistics of the

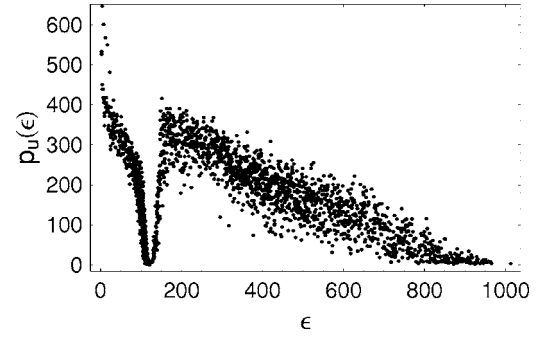


FIG. 6. Participation ratio for a strongly disordered system with  $\delta\alpha = 1.0$  and  $\delta\beta = 1.0$ .

intervals between neighboring modes in the spectrum, which are commonly referred to as level-spacing distributions (LSDs). The statistical properties of spectra of complex systems can be modeled by universal LSDs corresponding to certain ensembles of random matrices [35]. For example, a Poissonian spectrum of normalized level spacings,

$$s_j = (\epsilon_{j+1} - \epsilon_j) / \langle \epsilon_{j+1} - \epsilon_j \rangle,$$

is characteristic of uncorrelated ensembles of random matrices. If there is a correlation as for the Gaussian orthogonal ensemble (GOE), the corresponding LSD is of the form closely approximated by the Wigner expression [35],

$$P_1(s) = \frac{\pi}{2} s e^{-(\pi/4)s^2}. \quad (45)$$

As far as disordered systems are concerned, it has been shown that the localization transition in the three-dimensional Anderson model is accompanied by a change of the LSD [36,37]. On the metallic side, it follows Eq. (45), while on the insulating side it is described by the Poisson law  $P_0(s) = e^{-s}$ . Multifractal eigenstates—neither extended nor exponentially localized—have been found in 2D and 1D disordered electronic systems [38], with the LSD different from both  $P_0(s)$  and  $P_1(s)$ .

Our effective Hamiltonian matrix  $\mathcal{M}$  in Eq. (25) with random elements belongs to a general nonorthogonal ensemble of random matrices. The mathematical treatment of random matrices with no symmetry conditions goes back to the pioneering work by Ginibre [39]. For instance, the non-Hermitian random matrices have been used to describe the generic statistical properties of resonances in open quantum chaotic systems and to characterize the typical features of dissipative chaotic quantum maps [35,40]. The corresponding LSD is given by [39]

$$P_2(s) = \frac{3^4 \pi^2}{2^7} s^3 \exp\left[-\frac{3^2 \pi}{2^4} s^2\right]. \quad (46)$$

Like Eq. (45), it is also of the form suggested by Wigner [41],

$$P_W(s) \sim s^\sigma e^{-c_\sigma s^2}, \quad (47)$$

where the parameter  $\sigma$  and the constant  $c_\sigma$  depend on the restrictions imposed on the matrices. An important difference



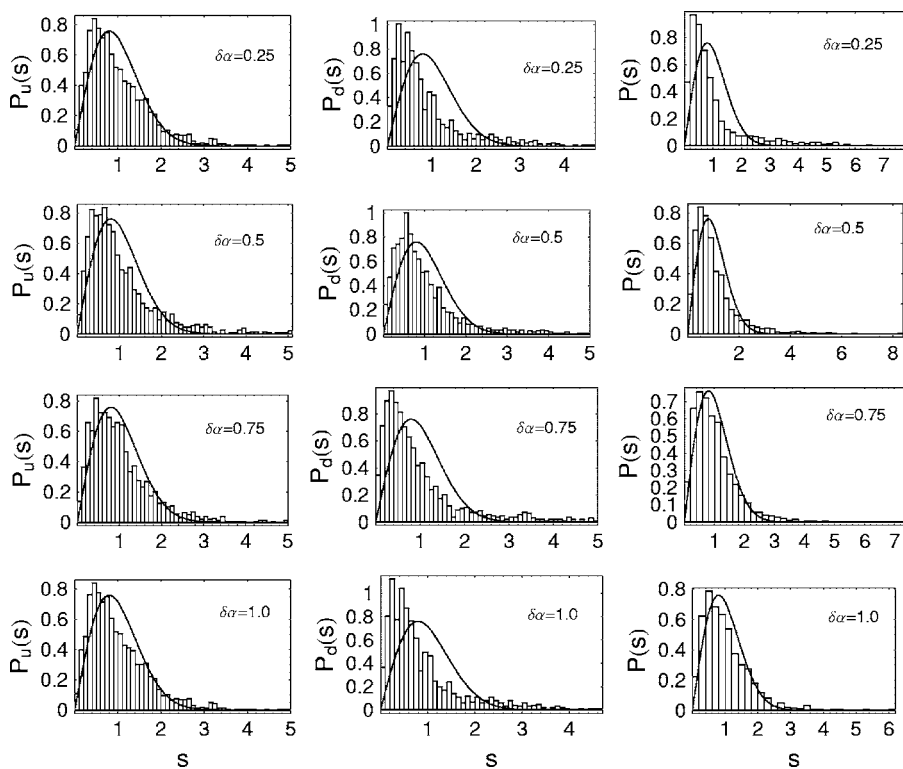


FIG. 7. Distributions of inter-level spacings (histograms) for three different spectral intervals explained in the text, for different magnitude of disorder of the coupling constant,  $\delta\alpha=0.25, 0.5, 0.75,$  and  $1.0$ , while keeping  $\delta\beta=0$ . The curve is the Wigner function  $P_1$ .

between the Wigner-type distributions and the Poisson distribution is their behavior for  $s \rightarrow 0$ . For the former, the probability of very small level spacings tends to zero indicating the repulsion of the levels. Let us also note that purely photonic or phononic excitations belong to the GOE universality class [34,35]. We have investigated LSD for the polariton system with disorder numerically. We considered three spectral regions of the eigenvalues including (i) those above the upper polariton band edge of the perfect system [ $\epsilon_j > \epsilon_{LO}$ , their distribution function is called  $P_u(s)$ ], (ii) eigenvalues lying below the gap in the lower branch edge [ $0 < \epsilon_j < \epsilon_{TO}$ , with their LSD denoted by  $P_d(s)$ ], and (iii) the whole polariton spectrum, with the LSD  $P(s)$ . The calculated distribution functions are depicted in Fig. 7 for different magnitudes of disorder,  $\delta\alpha=0.25, 0.5, 0.75,$  and  $1.0$  with  $\delta\beta=0$ . As can be seen from Fig. 7, the distribution of the nearest-neighbor spacings for the spectral regions (i) and (ii) are reasonably well-described by a Wigner function Eq. (47) with  $\beta$  slightly smaller than unity. As  $\delta\alpha$  increases, the distribution for the lower band [ $P_d(s)$ ] noticeably shifts to the region of the smaller  $s$ . The results calculated for the strong disorder case with dielectric constant fluctuations included ( $\delta\beta \neq 0$ ), presented in Fig. 8. One can see the quite different level statistics between the upper and lower polariton branches.  $P_u(s)$  is still reasonably described by the Wigner function  $P_W(s)$ , while the distribution for the lower branch implies a significantly reduced level repulsion [although it does not seem to tend to  $P_0(s)$ ]. We think that the explanation is related to the singular behavior of the density of states near the edge of the lower polariton band. The phonon-like excitations have a very heavy "effective mass" (and, consequently, large DS) near the band edge. This results in a smaller overlap of their localized eigenfunctions and, consequently, a smaller level

repulsion. Another feature of LSDs of Figs. 7 and 8 is the long tail observed for the large nearest-neighbor energy intervals. This behavior may be understood as a consequence of the interaction between the upper and lower bands, which results in an additional repulsion of the levels. As for the localized/delocalized nature of these states, the different LSD may be the manifestation of their multifractal nature, which is characteristic of various 2D systems and also of the 3D Anderson model exactly at the metal-insulator transition [37,38]. The distribution for the whole polariton spectrum retains its Wigner's shape even for strong disorder because the portion of levels near the gap edges is not very large. Finally, let us point out that the Ginibre's distribution Eq. (46) occurs when the fluctuations of the different parameters in the effective Hamiltonian have the same magnitude, so it can hardly be reached for the system under consideration.

## V. CONCLUDING REMARKS

In summary, we have studied the density of states and localization of phonon-polaritons in disordered polar media with fluctuating phonon-photon coupling constant and light velocity. The generalized Born-Huang equations of motion for the coupled fields were derived, which have been used to show that the problem of polariton localization can be formulated in terms of the generalized Anderson model with a non-Hermitian effective Hamiltonian.

We have performed an exact numerical diagonalization of such a Hamiltonian for a two-dimensional system and studied the disorder effects on the localization of polaritons in the regime where the energy scale of disorder and phonon-photon interaction strength are of the same order of magnitude. The investigation of the behavior of the density of

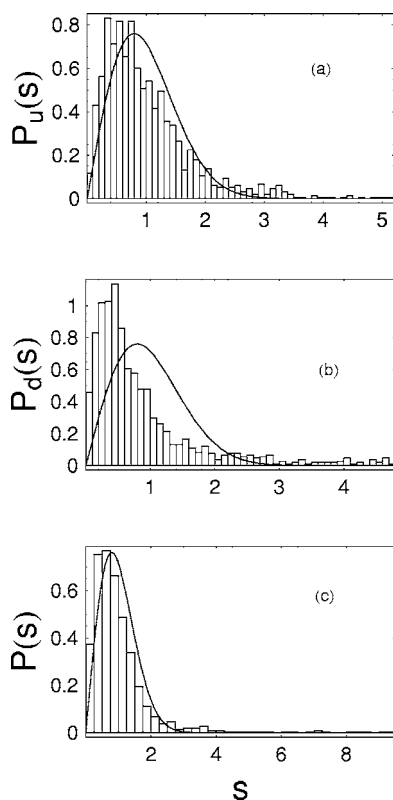


FIG. 8. Distribution of the interlevel spacings in the strong disorder regime,  $\delta\alpha=1.0$  and  $\delta\beta=1.0$  for the upper (a) and lower (b) polariton bands and (c) for the whole polariton spectrum. The curve is the Wigner function  $P_1$ .

states and the participation ratio near the polariton gap have shown that, when the magnitude of disorder is increased, localized modes appear forming tails inside the gap. At a certain magnitude of fluctuations of the coupling parameter, the tails in the gap overlap and the collapse of the gap is observed. This is facilitated by the fluctuations of the dielectric constant, although this type of disorder alone does not eliminate the gap. It has also been observed that the frequency range where the localization is enhanced by the fluc-

tuations in the phonon-photon interaction includes practically the whole polariton spectrum. In addition, the fluctuating dielectric constant enhances the localization of the higher-energy polaritons because of their stronger Rayleigh scattering (compared to the lower-energy excitations).

We have also investigated the distribution of the spectral intervals between the nearest-neighbor polariton energy levels. The statistics of the whole spectrum and of the levels belonging to the upper polariton branch do not show qualitative changes in a broad range of the magnitude of disorder and are reasonably described by the Wigner distribution with  $\sigma \sim 1$ . At the same time, we found substantial changes in the distribution for the lower (phonon) polariton branch, which correlates with the higher DS in this spectral region. The interpretation of this observation in terms of the localization of these states is not, however, completely clear and needs further investigation. Other targets of the future work should include the consideration of the finite lifetime of optical phonons (leading to a complex matrix  $\mathcal{M}=\mathcal{M}'+i\mathcal{M}''$ ) and of the disorder-induced coupling between LO and TO phonons (which is known to be important in nanostructures [8,12]). The developed approach, after an extension to anisotropic media, can be used for studying polariton excitations in polar crystals. In this case an interesting effect of polariton mode's interaction may arise as a consequence of mixing of transverse and longitudinal components of the electromagnetic field.

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