Polaritons in 2D-crystals and localized modes in narrow waveguides

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Abstract

We study 2D-polaritons in an atomically thin dipole-active layer (2D-crystal) placed inside a parallel-plate waveguide, and investigate the possibility to obtain the localized waveguide modes associated with atomic defects. Considering the waveguide width, $l$, as an adjustable parameter, we show that in the waveguide with $l \sim 10^4 a$, where $a$ is the lattice parameter, the localized mode can be provided by a single impurity or a local structural defect.
I. INTRODUCTION

The mechanism of the photon localization via the resonant coupling between photons and the local excitations inside the polariton gap was for the first time proposed in Refs. [1]. Considering a dipole-active impurity atom in an isotropic frequency-dispersive medium, the authors discovered the photon-atom bound states. They showed that when the atom transition frequencies fall inside the polariton gap, the radiative relaxation of the bound states is suppressed, and the field is localized around the impurity.

In the recent papers [3,4], we considered another type of the impurity-induced polariton states associated with the phonon local states in polar crystals. It was shown that near the bottom of the polariton gap, the local states are predominated by the long-wavelength modes and have the macroscopic localization and coherence radiuses. We attributed these features to the singularity of the density of states at the gap bottom, which also causes the absence of the lower threshold for the impurity strength. The singularity is provided by the long-wavelength polaritons and is generic for any isotropic dipole-active phonon mode with the negative dispersion.

The long-wavelength nature of the polariton states allowed us to analyze the crossover between the polariton and phonon local states within the continuum approximation. Our results show that the crossover takes place in a relativistically narrow interval near the bottom of the polariton gap [5]. A small width of the crossover region and a strong suppression of the photon content of the polariton local states are caused by the fact that the typical momentum of the modes dominating in these states, \( k_{\text{max}} \sim \beta^{1/2} a^{-1} \), is much greater than the cross-resonance momentum, \( k_0 \sim \beta a^{-1} \), where \( a \) is the lattice parameter, and \( \beta = v/c \) is the ratio between the phonon velocity and the speed of light. To eliminate this disproportion, one needs to lower the group velocity of electromagnetic waves in the active medium. It can be achieved if the medium is placed inside a waveguide. For instance, in the parallel-plate waveguide, the dispersion law of the propagating modes, \( \omega_n(k) = \sqrt{\left( \frac{\pi n}{2L} \right)^2 + k^2} \), provides the reduction of their group velocity in the long-wavelength region. The phonon and photon velocities become comparable in the cross-resonance region if the spacing between plates, \( l \), is of the order of \( 10^6 a \). However, as long as the waveguide spectrum contains the activationless mode \( (n=0) \), there is a guarantee that \( k_{\text{max}} \) is far away from the cross-resonance point.

In the present paper, we investigate the polarization waves in an atomically thin dielectric layer (ionic 2D-crystal) placed in a narrow parallel-plate waveguide. The structural stability of 2D-crystal is provided by a strong confining potential, which also eliminates the activationless mode from the waveguide spectrum. A sub-micron crystal film grown on a substrate can be a physical realization of this model. The coherent interaction between the waveguide modes and the polarization waves gives rise to new excitations - 2D-polaritons. They form two polariton bands, and the position of the maximum in the lower polariton branch depends on the waveguide width. We show that \( k_{\text{max}} \sim k_0 \) for \( l \sim \beta^{-2/3} a \), and \( k_{\text{max}} \) tends to \( \beta^{2/3} a^{-1} \) when \( l \gg \beta^{-2/3} a \). In the latter case, the frequency region of the polariton local states is enlarged by the factor of \( \beta^{-1/3} \sim 10^2 \), comparing to the 3D-case. The electric field in these states is always delocalized across the 2D-crystal within the distance of the order of \( l \) from the defect due to the contribution from the upper polariton band. Upon increase of the impurity strength, the localization length decreases and contributions to the
field from the lower and upper branches begin to compete everywhere in the waveguide. When the localization length becomes comparable with the waveguide width, the field is no longer confined near the dielectric layer and the polariton local state transforms into the localized waveguide mode. We estimate the corresponding value of the impurity strength, and show that, in the waveguide with \( l \sim \beta^{-2/3} a \), the localized mode can be provided by a single impurity or a structural defect.

II. POLARITONS IN THE IONIC 2D-CRYSTAL

Let us consider an atomically thin dielectric layer between two perfectly conducting sheets [Fig.1]. The layer presents a regular ionic 2D-crystal, which is stabilized in \( z = 0 \) plane by a strong restraining potential. This increases the activation energy of the off-layer lattice vibrations and shifts the corresponding phonon modes much higher than the modes with the in-plane polarization. Assuming an infinitely strong confining potential we eliminate the off-plane phonons from our model. Among the remaining in-plane phonons, for the sake of simplicity, we consider only a single transverse dipole-active mode with the isotropic spectrum and negative dispersion. In the long-wavelength region, we can use the standard approximation, \( \Omega^2(k) \approx \Omega_0^2 - v^2 k^2 \), where \( v \) sets the range of the typical phonon velocities. The dipole-active excitations in a thin layer have a high decay rate, unless induced electromagnetic radiation from the layer is compensated. In our model it is provided by the coherent coupling between the lattice excitations and the eigen modes of the waveguide. In the parallel plate waveguide, there are two types of propagating modes [Fig.1]. TM-modes (transverse-magnetic) involve electric field directed across the dielectric layer and, therefore, they cannot be activated in our model. Electric field in TE-modes (transverse-electric) is directed along the layer and, therefore, these modes can be excited along with the the transverse-optical phonons. Introducing the surface polarization \( Q(x,y) \) associated with these 2D-phonons and considering its dynamics together with the corresponding TE-modes of the waveguide, we obtain the following system of equations:

\[
\frac{c^2 \partial^2 E_k}{\partial z^2} + \left( \omega^2 - c^2 k^2 \right) E_k = -4\pi \omega^2 Q_k \delta(z),
\]

\[
\left[ \omega^2 - \Omega^2(k) \right] Q_k = -\frac{ad^2}{4\pi} E_k(0),
\]

where \( Q_k \) and \( E_k(z) \) are the 2D-Fourier amplitudes of the polarization and electric fields, \( k \) is a 2D-wave vector, \( d \) is the phonon-photon amplitudes of the localization and electric fields, \( k \) is a 2D-wave vector, \( d \) is the phonon-photon coupling parameter (ion plasma frequency). For the “order of magnitude” estimates we assume in this paper that \( d \sim \Omega_0 \sim v/a \). For later convenience we introduce the dimensionless variables \( l k \rightarrow k \), \( l \omega / c \rightarrow \omega \), and \( l \Omega / c \rightarrow \Omega \), so that \( \Omega^2(k) \approx \Omega_0^2 - \beta^2 k^2 \), and \( \Omega_0 \sim k_0 \sim \eta \beta \), where \( \eta = l/a \) is a large parameter.

The electric field in Eq.(1) is confined between the conducting plates and its normal derivative at the dielectric sheet has a discontinuity caused by the surface polarization. Solving Eqs.(1, 2) under these conditions, we obtain the following dispersion equation:

\[
\frac{\Omega^2 - \omega^2}{\omega^2} = \delta \frac{\tan \sqrt{\omega^2 - k^2}}{\sqrt{\omega^2 - k^2}}.
\]
where \( \delta = \alpha l d^2 / 2 c^2 \sim \eta \beta^2 \) is a small parameter.

This equation defines a series of polariton branches, \( \omega = \omega_n(k) \), with their activation frequencies given by the equation [Fig. 2,3]:

\[
\omega_n^2 - \Omega_0^2 = -\delta \omega_n \tan \omega_n .
\]  

(4)

Analysis of Eq. (3) shows that the upper \((n \geq 1)\) branches have the quadratic “large momenta” asymptotes:

\[
\omega_n^2(k) \approx \omega_n^2 + k^2 ,
\]

(5)

where \( \omega_n \approx \pi (n - 1 / 2) \gg \Omega_0 \). Therefore, their spectral bands all overlap and form a common upper polariton band.

The lower \((n = 0)\) polariton branch is separated from the others by the polariton gap, the bottom of which coincides with the maximum in this branch. Evaluation of the group velocity at the center of the Brillouin zone,

\[
\left[ \frac{d \omega_0^2(k)}{dk^2} \right]_{k=0} \propto -\beta^2 + \frac{\Omega_0^2 - \omega_0^2}{2 \omega_0^2} \left( \frac{2 \omega_0}{\sin 2 \omega_0} - 1 \right) \sim -\beta^2 + \frac{\delta \Omega_0^2}{3} ,
\]

(6)

shows that the lower branch has a negative dispersion if \( l \lesssim \beta^{-2/3} a \sim 10^4 a \). In such narrow waveguides the photon-phonon coupling is negligible and the phonon branch, with its maximum located at \( k = 0 \), remains practically unaffected by the field.

As \( l \) increases, the maximum moves away from \( k = 0 \), reaching the cross-resonance region, \( k \sim k_0 \), at \( l \sim \beta^{-2/3} a \). We restrict our further consideration to the case, \( l \gg \beta^{-2/3} a \), only. It guarantees that the maximum of the lower branch is located far away from \( k_0 \), in the region where \( k \gg \Omega, \omega \), and Eq.(3) can be approximated as follows:

\[
\Omega^2 - \omega^2 \approx \frac{\delta}{2k^3} \omega^2 (\omega^2 + 2k^2) .
\]

(7)

A positively defined solution of Eq.(7) gives the “large momenta” asymptote of the lower polariton branch,

\[
\omega_0^2(k) \approx \Omega^2 \left( 1 - \frac{\delta}{k} \right) .
\]

(8)

The corresponding dispersion curve reaches its maximum at the point

\[
k_{\text{max}} \approx l \left( \frac{\alpha d^2 \Omega_0^2}{2 v^2 c^2} \right)^{1/3} \sim \eta \beta^{2/3} \gg 1,
\]

(9)

where it sets the bottom of the polariton gap:

\[
\omega_{\text{max}}^2 \approx \Omega_0^2 - 3 \frac{l^2}{c^2} \left( \frac{\alpha v d^2 \Omega_0^2}{2 c^2} \right)^{2/3} \sim \Omega_0^2 - 3 \beta^{4/3} \left( \frac{d^2 \Omega_0}{c^2} \right)^{2/3} \frac{l^2}{c^2} .
\]

(10)

In the immediate vicinity of \( k_{\text{max}} \) the polariton dispersion law can be presented as follows:
\[ \omega_0^2(k) \approx \omega_{\text{max}}^2 - 3\beta^2(k - k_{\text{max}})^2, \quad (11) \]

and the asymptote of the density of states near the gap bottom has the form:

\[ \rho(\omega^2) \propto \frac{k_{\text{max}}}{2\pi\beta \sqrt{3(\omega_{\text{max}}^2 - \omega^2)}} \sim \frac{c}{a} \frac{\eta^{2/3}}{\Omega_0 \sqrt{\omega_{\text{max}}^2 - \omega^2}}, \quad (12) \]

Restoring the true dimensionality of the variables and comparing our results with those obtained for 3D-polaritons:

\[ k_{\text{max}} \sim \beta^{1/2} a^{-1}, \omega_{\text{max}}^2 \sim \Omega_0^2 - \beta d \Omega_0, \]
\[ \rho(\omega^2) \propto \frac{a(ak_{\text{max}})^2}{v \sqrt{\omega_{\text{max}}^2 - \omega^2}} \sim \frac{\beta}{\Omega_0 \sqrt{\omega_{\text{max}}^2 - \omega^2}}, \]

one can see that the maximum of the polariton curve is now located closer to the cross-resonance point, \( k_0 \sim \beta a^{-1} \), and the bottom of the gap is shifted toward the phonon activation frequency. Also, the singularity of the density of states is strengthened by the factor of \( \beta^{-1/3} \sim 10^2 \). Setting \( \rho(\omega^2) \sim 1/\Delta \), where \( \Delta \) is the width of the polariton band, one can estimate the frequency range where the singularity prevails:

\[ \sqrt{\omega_{\text{max}}^2 - \omega^2} \sim \beta^{2/3} \frac{\Delta}{\Omega_0} \sim \beta^{2/3} \Omega_0. \quad (13) \]

Comparing to the 3D-case, it is enlarged by the factor of \( \beta^{-1/3} \sim 10^2 \), what broadens the region of the local states dominated by long-wavelength modes.

**III. DEFECT-INDUCED LOCAL STATES**

If a point-like defect is embedded in the dielectric layer, it modifies Eq.(2) as follows:

\[ [\omega^2 - \Omega^2(k)] \mathcal{Q}_k = -\frac{ad^2}{4\pi} E_k(0) + \frac{\alpha a^2 Q(0)}{4\pi S}, \quad (14) \]

where \( Q(0) \) is the polarization of a defect and \( S \) is the total area of the layer. The strength of the defect, \( \alpha \), depends on its mobility and binding energy in a crystal. In the case of an isotope impurity \( \alpha = -\omega^2 \delta m/m \); for a non-isotope impurity or a structural defect we assume that \( \alpha \sim \pi \Omega_0^3 \), where \( \pi \) is a numerical parameter.

Solving Eqs.(1,14) we obtain ( in the dimensionless variables \( \omega, k, \text{and} \Omega \)):

\[ E_k(z) = \frac{\alpha a^2 l Q(0)}{2 S \gamma} \times \frac{\omega \sin \zeta Z}{\zeta \cos \zeta} \times \left( \omega^2 - \Omega^2 + \delta \frac{\omega^2 \tan \zeta}{\zeta} \right)^{-1}, \quad (15) \]

where we denote \( Z = l^{-1}(l \pm z) \) and \( \zeta = \sqrt{\omega^2 - k^2} \).

Using this equation one can express \( Q_k \) via the polarization of the defect, \( Q(0) \), and obtain then the spectral equation for the local state:

\[ 1 = \frac{\alpha}{4\pi c^2} \left( \frac{a}{2\pi} \right)^2 \times \int dk \left( \omega^2 - \Omega^2 + \delta \frac{\omega^2 \tan \zeta}{\zeta} \right)^{-1}. \quad (16) \]
When the frequency approaches $\omega_{\text{max}}$, the integral diverges at the “surface” $k^2 = k_{\text{max}}^2$. Near $\omega_{\text{max}}$ we can approximate Eq.(16) as follows:

$$1 \approx \frac{\alpha^2 (a^2 \pi^2)}{2c^2} \times \int_0^\infty \frac{kdk}{\omega^2 - \omega_{\text{max}}^2 + 3\beta^2 (k - k_{\text{max}})^2},$$

(17)

Retaining here only the singular part of the integral, we finally obtain:

$$\sqrt{\omega^2 - \omega_{\text{max}}^2} \approx \frac{\alpha a (\kappa_{\text{max}})}{8\pi c^2 \beta^3} \sim \frac{\alpha \eta \beta^{5/3}}{3}.$$

(18)

Comparing to the 3D-case $\left(\sqrt{\omega^2 - \omega_{\text{max}}^2} \sim \alpha \eta \beta^2\right)$, the separation of the local state from the bottom of the gap is enlarged by the factor of $\beta^{-1/3}$.

Equation (18) defines the eigen frequency of the local state near the bottom of the gap. To evaluate the localization radius of this state one needs to consider the spatial distribution of the electric and the polarization fields. The inverse Fourier transformation of Eq. (15) gives us:

$$E(z, r) = \frac{\alpha \omega^2 a^2 Q(0)}{4\pi l c^2} \times \int_{-\infty}^{\infty} \frac{dk}{\omega^2 - \Omega^2} \frac{H_0(Rk) \sin (Z \zeta)}{(\omega^2 - \Omega^2) \zeta \cos \zeta + \delta \omega^2 \sin \zeta},$$

(19)

where $H_0(Rk)$ is the Hankel function of first kind, $R = l^{-1}r$.

It follows from the spectral properties of our system, that the denominator of the integrand, considered as a function of $\omega^2$, has a series of isolated simple zeroes:

$$F(\omega, k) = \left[\left(\omega^2 - \Omega^2\right) \zeta \cos \zeta + \delta \omega^2 \sin \zeta\right] \propto \prod_n \left[\omega^2 - \omega_n^2(k)\right],$$

(20)

where the index $n$ enumerates different polariton branches, $\omega_n^2(k)$. Since the frequency of the local state, $\omega$, lies in the polatiton gap, all $k$-zeros of $F(\omega, k)$ are removed from the $\text{Re } k$-axis. Closing the integration contour in Eq.(19) through the upper half-plane, we can calculate the integral by the method of residuals:

$$E(z, r) = \frac{i\alpha \omega^2 a^2 Q(0)}{2l c^2} \times \sum_{k_n} \text{Res} \left\{H_0(Rk) \sin (Z \zeta) [F(\omega, k)]^{-1}\right\}_{k_n};$$

(21)

where $k_n$ is a pole associated with $n$-th branch.

Near the bottom of the gap ($\omega \gtrsim \omega_{\text{max}}$) the factor presenting the lower polariton branch, $\omega^2 - \omega_0^2(k)$, is small for $k \sim k_{\text{max}}$. It suggests that the poles of $[F(\omega, k)]^{-1}$ corresponding to the lower branch are located in the vicinity of $k_{\text{max}}$. Using Eq.(11) one can find:

$$k_0^\pm = k_{\text{max}} \pm i \sqrt{\frac{\omega^2 - \omega_{\text{max}}^2}{3\beta^2}} = k_{\text{max}} \pm i \kappa.$$

(22)

Taking into account that $k_{\text{max}} \sim \eta \beta^{2/3} \gg \omega_{\text{max}} \sim \eta \beta$, and $\kappa \sim \eta \frac{k_{\text{max}}}{k_{\text{max}}} \ll k_{\text{max}}$ for a weak defect, one can obtain:
\[ \text{Res} \left[ F(\omega, k) \right]^{-1}_{k_0^+} = \left[ \frac{\partial F[\omega_0(k), k]}{\partial k} \right]^{-1}_{k_0^+} \approx \frac{\exp(-k_{\text{max}})}{3\beta^2 \kappa k_{\text{max}}}. \tag{23} \]

To evaluate zeroes of \( F(\omega, k) \) corresponding to the upper branches, we first impose the upper limitation on the width of the waveguide, such that \( \beta^{-2/3}a \ll l \ll \beta^{-1}a \). In this case, the phonon band lies well below the waveguide cut-off frequency and all high-order polariton branches are practically indistinguishable from the parent TE-modes. It gives us:

\[ k_n^+ = \pm i \sqrt{\omega_n^2 - \omega^2} \approx \pm i \omega_n, \tag{24} \]

and the corresponding residuals:

\[ \text{Res} \left[ F(\omega, k) \right]^{-1}_{k_0^+} \approx \frac{i \cos \omega_n}{\delta \omega^2}. \tag{25} \]

Finally, using Eqs.(21-26), we can find the large-distance asymptote of the electric field:

\[ E(z, r) \propto \mathcal{R}^{-1/2} \left[ \frac{\exp(-\mathcal{R} \kappa - zk_{\text{max}})}{6 \beta^2 \kappa k_{\text{max}}^{3/2}} + \sum_{n=1}^{\infty} \frac{\exp(-\mathcal{R} \omega_n) \sin(Z \omega_n) \cos \omega_n}{\delta \omega^2 \omega_n^{1/2}} \right]. \tag{26} \]

This result presents the field as a sum of contributions from all branches. However, because \( \omega_n \) lie close to \( \pi (n - \frac{1}{2}) \) for the upper branches, the only several terms corresponding to the low-located branches can be retained in the sum.

Equation (26) explicitly demonstrates the localization of the field in radial directions, whereas, the \( z \)-profile of the field depends on the defect strength, \( \kappa \). For a weak defect, when \( \kappa \ll \omega_1 \) (localization length is much greater than \( l \)), the first term of Eq. (26) dominates at \( \mathcal{R} \gg \kappa^{-1} \) and confines the field within the \( k_{\text{max}}^{-1} \)-wide layer around the dielectric sheet. However, in the kernel of the local state, within the distance \( \mathcal{R} \lesssim \kappa^{-1} \) from the defect, next terms of Eq.(26) begin to compete with the lower band contribution. This leads to “delocalization” of the field in \( z \)-direction within the kernel.

Upon increase of the defect strength, the local state moves away from \( \omega_{\text{max}} \) and the localization length, \( \kappa^{-1} \), decreases. When we are reaching \( \overline{\kappa}_{\text{cr}} \sim \omega_1/k_{\text{max}} \), the first term of Eq.(25) is no longer dominating in \( E(z, r) \). In this case, despite the localization of the field within the \( l \)-range in radial directions, its \( z \)-confinement completely disappears. Such a state can be qualified as a local waveguide mode. Analysis of the structure and properties of these states requires one to consider waveguides with \( l \sim \beta^{-2/3}a \), where \( k_{\text{max}} \sim k_0 \). Recalling the result concerning the energy distribution for 3D local polaritons \[ W_{\text{field}}/W_{\text{mech}} \sim (k_0/k_{\text{max}})^4, \] we can expect the energy equipartition in the local waveguide modes. Also, since \( k_{\text{max}} \sim \omega_1 \) in the sufficiently narrow waveguides, there the local modes can be provided by defects with \( \kappa \lesssim 1 \), such as isotop impurities or local structural defects. A more detailed analysis of this case will be presented elsewhere.
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