

Polarons in two-dimensional polar materials: All-coupling variational theory

A. Kudlis^{1,*}, V. Shahnazaryan^{2,†} and I. V. Tokatly^{3,4,5,‡}

¹Science Institute, *University of Iceland*, Dunhagi 3, IS-107 Reykjavik, Iceland

²*Abrikosov Center for Theoretical Physics, MIPT, Dolgoprudnyi, Moscow Region 141701, Russia*

³*Departamento de Polímeros y Materiales Avanzados: Física, Química y Tecnología, Universidad del País Vasco, Avenida Tolosa 72, E-20018 San Sebastián, Spain*

⁴*Donostia International Physics Center (DIPC), E-20018 Donostia-San Sebastián, Spain*

⁵*IKERBASQUE, Basque Foundation for Science, Plaza Euskadi 5, 48009 Bilbao, Spain*



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We present a detailed and self-contained theoretical study of polarons in two-dimensional (2D) polar materials, which extends the classical macroscopic theory of Fröhlich polarons to the 2D case. The theory is fully determined by experimentally accessible parameters, the static and optical 2D polarizabilities of a monolayer, the frequency of transverse optical phonons, and the effective mass of charge carriers. We define a single dimensionless parameter, which characterizes the coupling of electrons with longitudinal optical phonons, analyze both weak- and strong-coupling regimes, and adapt the Feynman variational path-integral approach for a high-quality interpolation between these limits. Our results provide insight into the ground-state energy and effective mass of polarons in modern 2D polar monolayers.

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I. INTRODUCTION

An electron moving in a polar solid interacts with ions in the lattice sites and locally polarizes the crystal. This leads to the formation of polaron, a quasiparticle composed of an electron surrounded by the local lattice deformation, as schematically shown in Fig. 1. Polarons play an essential role in elucidating the transport and optical properties of polar materials [1–3]. Originally, the idea of polaron was proposed by Landau and further elaborated by Pekar [4–6]. In the Landau-Pekar (LP) picture, the polar crystal is viewed as a polarizable medium that adiabatically follows the electron, forming a self-consistent polarization cloud around it. This reduces the electron energy and can lead to its self-trapping in a polarization well. In the 1950s, a complementary picture of an electron dressed by virtual longitudinal optical (LO) phonons was developed by Fröhlich based on a perturbative treatment of the problem [7], and somewhat later it was restated in a variational form by Lee, Low, and Pines (LLP) [8]. Shortly, Feynman made further significant progress in the theory of polarons by adopting for this purpose a path-integral variational formalism [9]. The adiabatic LP and the perturbative Fröhlich approaches describe, respectively, the regimes of strong and weak coupling of an electron to the lattice polarization. In

contrast, the Feynman theory is valid for arbitrary interaction strength, providing a high-quality interpolation between the weak- and strong-coupling limits. Despite its conceptual simplicity, the Feynman approach provides a remarkably accurate description of polarons [10], as has been confirmed by subsequent Monte Carlo calculations [11–13]. In fact, the Feynman theory of polarons is one of the most successful practical applications of the path-integral technique.

Due to their importance for physics of polar crystals, such as alkali halides and certain oxides, polaron effects have been extensively studied both theoretically and experimentally [14,15], providing a prototype for strong-coupling electron-phonon physics. Over the past decades, various theoretical techniques, including diagrammatic Monte Carlo [11–13] and first-principles DFT-based [16–18] methods, have been used to capture different aspects of polaron physics. An excellent account of the theory of polarons, including an extensive list of literature, can be found in the lecture notes of Ref. [3].

Starting from the earlier classical works by Landau, Pekar, Fröhlich, and Feynman, the formation of polarons has been mostly studied for three-dimensional (3D) polar crystals, first in the bulk and, beginning from the 1970s, on the surface [13,19–22]. Until very recently, the quasi-two-dimensional (2D) polarons, appearing due to the interaction with surface LO phonons on boundaries of 3D crystals, represented the only well-studied example of low-dimensional polarons. The studies of genuine 2D polarons remained relatively sparse, despite rapid progress in the discovery and synthesis of 2D materials [23]. Experimental reports on 2D polarons exist, for example, in hexagonal boron nitride (hBN) [24], MoS₂ monolayers [25], and SrTiO₃ surfaces [26,27].

A key feature of 2D polar materials, relevant for polaron physics, is their reduced, nonlocal dielectric screening [28–30]. This very special screening is responsible for the

*Contact author: andrewkudlis@gmail.com

†Contact author: vanikshahnazaryan@gmail.com

‡Contact author: ilya.tokatly@ehu.es

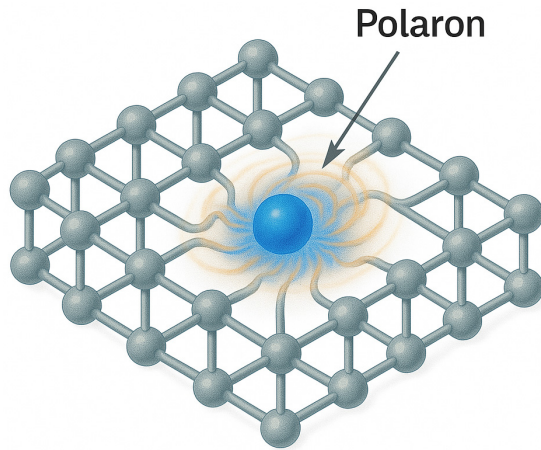


FIG. 1. Schematic illustration of a polaron in a polar lattice. The blue sphere represents an electron, whose interaction with the surrounding ions locally deforms the lattice (indicated by the orange halo), effectively dressing the electron with a phonon cloud.

nontrivial dispersion of long-wavelength LO phonons and fundamental modifications of electron-phonon interactions [31–35]. Some aspects of polaron physics in 2D polar monolayers have been studied within a first-principles extension of Landau-Pekar theory [36], and by perturbatively computing polaron shifts of Landau levels [37]. There are recent reports on 2D polarons in various material platforms [38–40]. In a recent work [41], we have proposed a macroscopic approach to optical phonon modes and electron-LO phonon coupling in 2D polar materials, which generalizes the classical textbook theory of 3D polar crystals [42,43]. Similarly to the 3D case, in this approach, the effective electron-phonon Hamiltonian is parametrized solely in terms of macroscopic experimentally accessible quantities—2D polarizabilities of the monolayer at low and high frequencies. In Ref. [41], the proposed theory of 2D polar material has been applied to study polarons and polaron-excitons within a variational scheme of LLP, which, strictly speaking, should be valid in the weak-coupling regime.

In the present paper, we combine the macroscopic framework of Ref. [41] with Feynman’s path-integral variational theory, known for its remarkable accuracy, to describe Fröhlich polarons in 2D polar crystals without any restriction to the strength of electron-phonon coupling. As a first step, we restate the 2D electron-phonon Hamiltonian derived in Ref. [41] in a form essentially identical to the standard 3D Fröhlich Hamiltonian. However, in the 2D case, the dimensionless Fröhlich constant $\alpha(q)$ and the frequency of LO phonons $\omega_{l,q}$ become functions of the wave vector q . This q dependence is encoded in the effective nonlocal 2D permittivities at high $\varepsilon_{\infty}(q)$ and low $\varepsilon_0(q)$ frequencies, reflecting the long-range nonlocality characteristic for the electrostatic response of 2D dielectrics [30]. The lack of a single Fröhlich constant in 2D makes characterization of the coupling strength not obvious. Here, we show that a properly defined upper bound α_m of the function $\alpha(q)$ provides a natural measure of electron-phonon coupling in 2D polar materials. With this formulation at hand, we derive an *all-coupling* Feynman’s variational functional that explicitly retains the effects of 2D nonlocal screening

and smoothly interpolates between the LLP/weak-coupling and LP/strong-coupling limits, while providing closed expressions for the ground-state energy and the polaron effective mass for arbitrary coupling. Therefore, the present work goes well beyond both Ref. [41], in which polarons are described within LLP scheme, and Ref. [36], where the adiabatic LP-type approximation is adopted. Using our all-coupling theory for realistic sets of material parameters, we identify a robust crossover between the weak (LLP)- and the strong (LP)-coupling regimes at the effective coupling constant of $\alpha_m^* \approx 5$. Moreover, we show that all currently known monolayers are on the weak-to-intermediate (LLP) side of the crossover, while the LP regime is never realized.

The paper is organized as follows. To make the paper self-contained and to restate the theory in a form suitable for a subsequent application of the path-integral technique, in Sec. II we derive the classical Lagrangian and Hamiltonian describing TO and LO phonons in 2D polar crystals, obtain the dispersion relation of LO phonons, and discuss its interpretation as a nonlocal Lyddane-Sachs-Teller (LST) relation. The electron-phonon coupling terms, in both the Lagrangian and Hamiltonian settings, are derived in Sec. III. Here, we also discuss relevant dimensionless parameters and introduce the effective coupling constant that measures the strength of electron-LO phonon interaction in 2D crystals. In Sec. IV, we consider the polaron ground state in the weak- and strong-coupling limits. In Sec. V, the Feynman variational path-integral approach is adopted for our case of 2D polar crystals. We show how it interpolates between the limits of weak and strong coupling, providing quantitative estimates of both the polaron binding energy and effective mass. The results of numerical calculations within the Feynman approach are presented in Sec. VI. We discuss applications to specific materials and provide a general analysis of the crossover between the weak- and strong-coupling regimes. The main results of this work are summarized in Sec. VII, with a discussion of the broader implications for transport and optical properties of 2D polar materials.

II. MACROSCOPIC THEORY OF 2D OPTICAL PHONONS

Let us consider a generic 2D polar crystal containing sublattices of positive and negative ions. The macroscopic theory of long-wavelength optical phonons in the 3D case dates back to the works of Born and Huang Kun [42], and by now constitutes a classical part of solid-state physics (see, for example, Ref. [43]). A proper 2D generalization of this macroscopic approach has been developed in our previous work [41]. Here, to make the paper self-contained, we present the main results of that theory and adapt its formulation for the specific purpose of this work.

A. Construction of Lagrangian density

Our main goal is to describe long-wavelength dynamics of the relative displacement field $\mathbf{S}$,

$$\mathbf{S} = \mathbf{U}_+ - \mathbf{U}_-, \quad (1)$$

where $\mathbf{U}_+$ and $\mathbf{U}_-$ are the displacement vectors of positive and negative ions, respectively. For further convenience, we define

the mass-weighted displacement field ξ as follows:

$$\xi = S\sqrt{nM}, \quad (2)$$

with $n = N/A$ being the surface density of unit cells, where A is the normalization area, and M the reduced ionic mass,

$$M = \frac{m_+ m_-}{m_+ + m_-}, \quad (3)$$

with $m_{\pm}$ denoting the respective ionic masses. In the long-wavelength limit, that is, assuming that the displacement varies on the scale much larger than the interatomic distance and the thickness of 2D layer, we write the Lagrangian density of the system as

$$\mathcal{L} = \left(\frac{1}{2} \dot{\xi}^2 - W^{2D} \right) \delta(z) + \epsilon_0 \epsilon \frac{\mathbf{E}^2}{2}, \quad (4)$$

where the first two terms (in parentheses) represent the kinetic and potential energy contributions (per unit area) localized in the plane that hosts the lattice ($z = 0$). The last term is the usual density of electromagnetic energy in the surrounding medium with the dielectric constant ϵ (ϵ_0 stands for the vacuum permittivity). It should be emphasized that while the moving ions are bound to the 2D plane, they produce an electric field throughout the 3D space, which is crucial for a correct description of dielectric screening in 2D materials [30]. The potential energy density W^{2D} in Eq. (4) accounts for the elastic energy of ionic displacements and their interaction with the electric field $\mathbf{E}$. Assuming for simplicity macroscopic isotropy of the 2D layer, we write

$$W^{2D} = \frac{1}{2} \omega_t^2 \xi^2 - \gamma \xi \cdot \mathbf{E}(\mathbf{r}, z=0) - \frac{\alpha_{\infty}}{2} \mathbf{E}^2(\mathbf{r}, z=0), \quad (5)$$

which is analogous to the corresponding expression in the 3D case [43]. Here, ω_t is the transverse optical (TO) phonon frequency, γ parametrizes a coupling between ξ and $\mathbf{E}$, and α_{∞} is the high-frequency (optical) 2D polarizability of a free-standing layer, which accounts for the electronic contribution to the electric response [44]. By definition, the resulting two-dimensional polarization field is

$$\mathbf{P}^{2D} = -\frac{\partial W^{2D}}{\partial \mathbf{E}} = \gamma \xi + \alpha_{\infty} \mathbf{E}. \quad (6)$$

Taking the derivative with respect to ξ gives

$$\frac{\partial W^{2D}}{\partial \xi} = \omega_t^2 \xi - \gamma \mathbf{E}. \quad (7)$$

In equilibrium (static limit $\omega = 0$), we must have

$$\omega_t^2 \xi_0 - \gamma \mathbf{E} = 0, \quad (8)$$

which leads to the following relations:

$$\xi_0 = \frac{\gamma}{\omega_t^2} \mathbf{E}, \quad \mathbf{P}_0^{2D} = \gamma \xi_0 + \alpha_{\infty} \mathbf{E} = \alpha_0 \mathbf{E}. \quad (9)$$

Thus, the full static 2D polarizability is identified as

$$\alpha_0 = \alpha_{\infty} + \frac{\gamma^2}{\omega_t^2}, \quad (10)$$

which eventually allows us to express the coupling parameter γ in Eq. (5) in terms of experimentally accessible quantities,

the static and optical 2D polarizabilities, and the frequency of TO phonons,

$$\gamma = \omega_t \sqrt{\alpha_0 - \alpha_{\infty}}. \quad (11)$$

B. Dispersion of LO phonons in 2D

The Lagrangian in Eq. (4) generates the following equation of motion for the mass-weighted displacement:

$$\ddot{\xi} = -\omega_t^2 \xi + \gamma \mathbf{E}(\mathbf{r}, z=0). \quad (12)$$

Another equation of motion corresponds to the Gauss law, which is obtained by defining $\mathbf{E} = -\nabla\varphi$ in Eqs. (4) and (5) and minimizing the Lagrangian with respect to the electrostatic potential φ . This equation relates the divergence of the electric field to the induced charge density,

$$\nabla \cdot \mathbf{E}(\mathbf{r}, z) = \frac{\rho}{\epsilon \epsilon_0} \equiv -\frac{1}{\epsilon \epsilon_0} \nabla \cdot \mathbf{P}^{2D} \delta(z). \quad (13)$$

Hence, once ξ is specified, the electrostatic potential can be determined.

By substituting $\mathbf{E} = -\nabla\varphi$ into the equations of motion, we have

$$\ddot{\xi} = -\omega_t^2 \xi - \gamma \nabla\varphi|_{z=0}, \quad (14)$$

$$\nabla^2 \varphi(\mathbf{r}, z) = \frac{1}{\epsilon \epsilon_0} \nabla \cdot (\gamma \xi - \alpha_{\infty} \nabla\varphi|_{z=0}) \delta(z). \quad (15)$$

Assuming plane-wave normal-mode solutions with in-plane wave vector $\mathbf{q}$, we perform the Fourier transform and obtain the following result for the Fourier component of the electrostatic potential:

$$\varphi_{\mathbf{q}, k_z} = -\frac{1}{\epsilon \epsilon_0} \frac{1}{q^2 + k_z^2} (i\gamma \mathbf{q} \cdot \xi_{\mathbf{q}} + \alpha_{\infty} q^2 \varphi_{\mathbf{q}}^{2D}). \quad (16)$$

Here, $q = |\mathbf{q}|$, and the following conventions for the Fourier representation are used. In the plane, we define

$$\xi_{\mathbf{q}} = \int d^2r \xi(\mathbf{r}) \frac{e^{-i\mathbf{q} \cdot \mathbf{r}}}{\sqrt{A}}, \quad \xi(\mathbf{r}) = \sum_{\mathbf{q}} \xi_{\mathbf{q}} \frac{e^{i\mathbf{q} \cdot \mathbf{r}}}{\sqrt{A}}, \quad (17)$$

while in the z direction the Fourier transform reads

$$g_{k_z} = \int_{-\infty}^{\infty} dz e^{-ik_z z} g(z), \quad g(z) = \int_{-\infty}^{\infty} \frac{dk_z}{2\pi} e^{ik_z z} g_{k_z}. \quad (18)$$

We also introduce the following notation for the in-plane slice of the potential:

$$\varphi_{\mathbf{q}}^{2D} = \frac{1}{2\pi} \int_{-\infty}^{\infty} dk_z \varphi_{\mathbf{q}, k_z} \equiv \varphi_{\mathbf{q}}(z=0). \quad (19)$$

The explicit integration with $\varphi_{\mathbf{q}, k_z}$ of Eq. (16) gives

$$\varphi_{\mathbf{q}}^{2D} = -\frac{\gamma \xi_{\mathbf{q}}^l}{2\epsilon \epsilon_0 [1 + \alpha_{\infty} q / (2\epsilon \epsilon_0)]}, \quad (20)$$

where we defined the longitudinal component of the displacement as $\xi_{\mathbf{q}}^l = i\mathbf{q} \cdot \xi_{\mathbf{q}}/q$.

Substituting $\nabla\varphi|_{z=0} = i\mathbf{q} \varphi_{\mathbf{q}}^{2D}$ into Eq. (14), and assuming a harmonic solution $\xi_{\mathbf{q}} \sim e^{-i\omega t}$, we find

$$\omega^2 \xi_{\mathbf{q}}^l = \omega_t^2 \xi_{\mathbf{q}}^l - \gamma q \varphi_{\mathbf{q}}^{2D}. \quad (21)$$

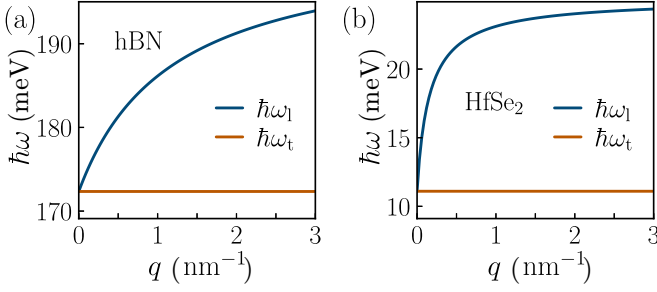


FIG. 2. Long-wavelength LO-phonon dispersion in two representative monolayers, illustrating the 2D nonlocal screening. Panels (a) (hBN) and (b) (HfSe₂) show the transverse optical mode (flat line, $\hbar\omega_t$, orange) and the longitudinal optical mode (solid line, $\hbar\omega_{l,q}$, blue) as functions of in-plane wave vector q . In both cases, $\omega_l(q)$ starts from ω_t at $q \rightarrow 0$ and increases monotonically with q , approaching the asymptote $\omega_t\sqrt{r_0/r_\infty} = \omega_t\sqrt{\sigma_0}$ at large q , in accordance with Eq. (22). The strength and curvature of the upshift are material dependent and governed by the ratio σ_0 of screening lengths.

Finally, using Eq. (20) and $\gamma^2 = \omega_t^2(\alpha_0 - \alpha_\infty)$, we recover the dispersion relation for the LO phonon mode, obtained recently in Ref. [41],

$$\omega_{l,q}^2 = \omega_t^2 \frac{1 + \alpha_0 q / (2\epsilon_0)}{1 + \alpha_\infty q / (2\epsilon_0)} = \omega_t^2 \frac{1 + r_0 q}{1 + r_\infty q}, \quad (22)$$

with 2D screening lengths, $r_\infty = \alpha_\infty / (2\epsilon_0)$, $r_0 = \alpha_0 / (2\epsilon_0)$, related to optical and static polarizabilities [45]. The dispersion relation of Eq. (22) agrees with earlier considerations supported by first-principles calculations [31,32,34,35]. Recently, it has been observed experimentally [46]. A typical dispersion of optical phonon modes for two representative materials is shown in Fig. 2.

The nontrivial dispersion of LO phonons in 2D polar insulators is a direct consequence of their nonlocal dielectric screening [30]. An interesting observation is that Eq. (22) can be interpreted as a 2D form of the LST relation. In fact, the quantity $\epsilon(q) = \epsilon + \alpha q / 2\epsilon_0$ plays a role of an effective dielectric constant in 2D, and therefore, Eq. (22) can naturally be represented in the form

$$\omega_{l,q}^2 = \omega_t^2 \frac{\epsilon_0(q)}{\epsilon_\infty(q)}. \quad (23)$$

It is worth noting that this simple form of the nonlocal LST-type relation is a consequence of the assumed macroscopic isotropy of the crystal, which is fulfilled in many known monolayers [32]. In low-symmetry 2D materials, by analogy with 3D case, one naturally expects that Eq. (23) will be replaced by a q -dependent version of the generalized LST relation [47,48].

III. ELECTRON-PHONON LAGRANGIAN

Having in mind the subsequent application to the path-integral formulation of the polaron problem, we now cast the phonon Lagrangian in the form of a sum over independent normal modes. All quantities in the following are expressed in the momentum representation introduced in the previous section. That is, the in-plane wave vectors, which count normal modes, assumed discrete (attributed to a large, but finite

normalization area A), while the out-of-plane wave vectors k_z are continuous and integration over k_z is assumed everywhere.

A. Normal-mode phonon Lagrangian

The original Lagrangian density of Eq. (4) explicitly reads as follows:

$$\mathcal{L} = \left(\frac{1}{2} \dot{\xi}^2 - \frac{\omega_t^2}{2} \xi^2 + \gamma \xi \cdot \mathbf{E}(\mathbf{r}, z=0) + \frac{\alpha_\infty}{2} \mathbf{E}^2(\mathbf{r}, z=0) \right) \delta(z) + \epsilon_0 \epsilon \frac{\mathbf{E}^2}{2}, \quad (24)$$

resulting in the Lagrangian

$$L = \int \mathcal{L} d^2r dz. \quad (25)$$

In order to obtain the Lagrangian in terms of normal modes, we should substitute the electric field $\mathbf{E} = -\nabla\varphi$ in the Fourier representation. Using the relation of Eq. (16), we find the result

$$\begin{aligned} \nabla\varphi(\mathbf{r}, z) = & -\frac{i\gamma}{\epsilon_0\epsilon\sqrt{A}} \sum_{\mathbf{q}} \frac{q \xi_{\mathbf{q}}^1 e^{i\mathbf{q}\cdot\mathbf{r}}}{1 + r_\infty q} \\ & \times \int_{-\infty}^{\infty} \frac{dk_z}{2\pi} \frac{\mathbf{q} + \hat{\mathbf{z}}k_z}{q^2 + k_z^2} e^{ik_z z}, \end{aligned} \quad (26)$$

which, at $z=0$ (the location of the 2D crystal), takes the following form:

$$\nabla\varphi(\mathbf{r}, 0) = -\frac{i\gamma}{2\epsilon_0\epsilon\sqrt{A}} \sum_{\mathbf{q}} \frac{\mathbf{q}}{1 + r_\infty q} \xi_{\mathbf{q}}^1 e^{i\mathbf{q}\cdot\mathbf{r}}. \quad (27)$$

After separating the longitudinal and transverse components of the displacement as $\xi_{\mathbf{q}} = -i\frac{\mathbf{q}}{q}\xi_{\mathbf{q}}^1 + \xi_{\mathbf{q}}^\perp$, using Eq. (27), and evaluating the contribution of each term in Eqs. (24) and (25), we find the Lagrangian of optical phonons in the following form (see Appendix A for the details):

$$L = L_{\text{TO}} + L_{\text{LO}}, \quad (28)$$

where, as expected, the contributions of the transverse and longitudinal modes read

$$L_{\text{TO}} = \frac{1}{2} \sum_{\mathbf{q}} (|\dot{\xi}_{\mathbf{q}}^\perp|^2 - \omega_t^2 |\xi_{\mathbf{q}}^\perp|^2), \quad (29)$$

$$L_{\text{LO}} = \frac{1}{2} \sum_{\mathbf{q}} (|\dot{\xi}_{\mathbf{q}}^1|^2 - \omega_{l,q}^2 |\xi_{\mathbf{q}}^1|^2). \quad (30)$$

Apparently, each mode $\xi_{\mathbf{q}}^1$ and $\xi_{\mathbf{q}}^\perp$ behaves as an independent harmonic oscillator with natural frequencies $\omega_{l,q}$ and ω_t , respectively.

B. Electron-phonon interaction in the Lagrangian framework

Finally, to include interaction with a charge carrier (electron or hole), one notices that a charge $-e$ located at a point $\mathbf{r}_e$ couples to LO optical phonons via the electrostatic potential, yielding the following correction to the Lagrangian:

$$\Delta L = e \int d^2r \delta(\mathbf{r} - \mathbf{r}_e) \varphi(\mathbf{r}, 0) = e \varphi(\mathbf{r}_e, 0). \quad (31)$$

Using the representation $\varphi(\mathbf{r}, 0) = A^{-1/2} \sum_{\mathbf{q}} \varphi_{\mathbf{q}}^{2D} e^{i\mathbf{q}\cdot\mathbf{r}}$ together with Eq. (20) for $\varphi_{\mathbf{q}}^{2D}$, we obtain

$$\varphi(\mathbf{r}, 0) = -\frac{\gamma}{2\epsilon_0\epsilon\sqrt{A}} \sum_{\mathbf{q}} \frac{\xi_{\mathbf{q}}^1}{1+r_{\infty}q} e^{i\mathbf{q}\cdot\mathbf{r}}, \quad (32)$$

and therefore

$$\begin{aligned} \Delta L &= -\frac{e\gamma}{2\epsilon_0\epsilon\sqrt{A}} \sum_{\mathbf{q}} \frac{\xi_{\mathbf{q}}^1 e^{i\mathbf{q}\cdot\mathbf{r}_e}}{1+r_{\infty}q} \\ &= \frac{1}{2} \sum_{\mathbf{q}} g_q (e^{i\mathbf{q}\cdot\mathbf{r}_e} \xi_{\mathbf{q}}^1 + e^{-i\mathbf{q}\cdot\mathbf{r}_e} \xi_{-\mathbf{q}}^1), \end{aligned} \quad (33)$$

with a real coupling constant

$$g_q = -\frac{e\gamma}{2\epsilon_0\epsilon\sqrt{A} [1+r_{\infty}q]}. \quad (34)$$

The overall electron-LO phonon Lagrangian is then

$$\begin{aligned} L_{e-ph} &= \frac{m_e \dot{\mathbf{r}}_e^2}{2} + \frac{1}{2} \sum_{\mathbf{q}} (|\dot{\xi}_{\mathbf{q}}^1|^2 - \omega_{l,q}^2 |\xi_{\mathbf{q}}^1|^2) \\ &\quad + \frac{1}{2} \sum_{\mathbf{q}} g_q (e^{i\mathbf{q}\cdot\mathbf{r}_e} \xi_{\mathbf{q}}^1 + e^{-i\mathbf{q}\cdot\mathbf{r}_e} \xi_{-\mathbf{q}}^1). \end{aligned} \quad (35)$$

Since TO phonons do not produce a long-range electric field, they do not interact electrostatically with electrons and are therefore irrelevant for the Fröhlich polaron problem. We therefore ignore the L_{TO} part of the Lagrangian in the following consideration. In Sec. V, the Lagrangian (35) will be used within the path-integral framework to develop a 2D generalization of Feynman's all-coupling variational theory of the Fröhlich polaron.

C. Hamiltonian formulation

It is also useful to restate the electron-LO phonon problem in the Hamiltonian formalism. In particular, we need it for the next section, where the weak-coupling limit will be analyzed within the perturbation theory.

First, within the standard canonical quantization, we express the coordinate and momentum operators for LO modes in terms of the corresponding phonon creation and annihilation operators. For each in-plane momentum $\mathbf{q}$, we have

$$\xi_{\mathbf{q}}^1 = \sqrt{\frac{\hbar}{2\omega_{l,q}}} (\hat{a}_{\mathbf{q}} + \hat{a}_{-\mathbf{q}}^{\dagger}), \quad (36)$$

$$\hat{\pi}_{\mathbf{q}} = \dot{\xi}_{\mathbf{q}}^1 = -i\sqrt{\frac{\hbar\omega_{l,q}}{2}} (\hat{a}_{\mathbf{q}} - \hat{a}_{-\mathbf{q}}^{\dagger}), \quad (37)$$

with $[\hat{a}_{\mathbf{q}}, \hat{a}_{\mathbf{q}'}^{\dagger}] = \delta_{\mathbf{q},\mathbf{q}'}$. The phonon Hamiltonian is then obtained via the Legendre transform,

$$\hat{H}_{ph} = \sum_{\mathbf{q}} \hbar\omega_{l,q} \hat{a}_{\mathbf{q}}^{\dagger} \hat{a}_{\mathbf{q}}. \quad (38)$$

By expressing $\xi_{\mathbf{q}}^1$ in ΔL in terms of $\hat{a}_{\mathbf{q}}, \hat{a}_{\mathbf{q}}^{\dagger}$, we get the 2D Fröhlich interaction, and eventually the electron-phonon Hamiltonian

$$\hat{H}_{e-ph} = \frac{\hat{\mathbf{p}}_e^2}{2m_e} + \hat{H}_{ph} + \sum_{\mathbf{q}} V_q (e^{i\mathbf{q}\cdot\mathbf{r}_e} \hat{a}_{\mathbf{q}} + e^{-i\mathbf{q}\cdot\mathbf{r}_e} \hat{a}_{\mathbf{q}}^{\dagger}), \quad (39)$$

with the following coupling amplitude:

$$V_q = -g_q \sqrt{\frac{\hbar}{2\omega_{l,q}}} = \frac{e\omega_l}{2\epsilon_0\epsilon} \sqrt{\frac{\hbar(\alpha_0 - \alpha_{\infty})}{2A\omega_{l,q}}} \frac{1}{1+r_{\infty}q}. \quad (40)$$

This Hamiltonian is the starting point for the perturbative weak-coupling and variational-strong coupling analyses in Sec. IV, while the Lagrangian form above is convenient for the path-integral approach, which we adopt in Sec. V.

Interestingly, the coupling amplitude V_q of Eq. (40) can be represented in a form similar to the standard Fröhlich interaction with 3D bulk LO phonons [7,8,43], and with dispersionless 2D surface phonons at surfaces of 3D polar crystals [13,19–22]. To this end, we introduce an effective *nonlocal* dimensionless Fröhlich constant $\alpha(q)$:

$$\alpha(q) = \frac{e^2}{4\pi\epsilon_0} \frac{1}{\hbar\omega_{l,q}} \sqrt{\frac{m_e\omega_{l,q}}{2\hbar}} \left[\frac{1}{\epsilon_{\infty}(q)} - \frac{1}{\epsilon_0(q)} \right], \quad (41)$$

where $\epsilon_{0,\infty}(q) = \epsilon(1+r_{0,\infty}q)$ are the effective 2D dielectric “constants”, and the frequency of LO phonons satisfies the *nonlocal* Lyddane-Sachs-Teller relation of Eq. (23):

$$\omega_{l,q} = \omega_l \sqrt{\frac{\epsilon_0(q)}{\epsilon_{\infty}(q)}}. \quad (42)$$

Then, the amplitude of 2D electron-phonon coupling in Eq. (40) can be represented as follows:

$$V_q = \left[\frac{\pi\alpha(q)\hbar}{Aq} \hbar\omega_{l,q} \sqrt{\frac{2\hbar\omega_{l,q}}{m_e}} \right]^{1/2}, \quad (43)$$

which is exactly the form commonly used for the coupling to quasi-2D surface phonons [13,19–22]. However, in the present case of genuine 2D phonons, both the Fröhlich coupling constant and the frequency of LO phonons become q dependent. This dependence, reflecting the long-range nonlocality of 2D dielectric screening, is a unique feature of the Fröhlich polaron problem in 2D, which makes it substantially different from the corresponding problems both in the bulk and on the surface of 3D polar materials.

D. Characterization of the coupling strength in 2D

In the cases of 3D crystals, both in the bulk and on the surface, the polaron problem is fully determined by a single dimensionless parameter, the coupling constant α . In 2D, the effective Fröhlich constant Eq. (41) acquires the wave vector dependence, and the very characterization of the coupling strength becomes nontrivial.

In general, the electron-phonon problem of Eqs. (39) and (40) contains four length scales, the screening lengths, r_{∞} and r_0 , the effective Bohr radius a_B , and the oscillator length r_l related to the frequency of TO phonons:

$$a_B = \frac{4\pi\epsilon_0\epsilon\hbar^2}{e^2m_e}, \quad r_l = \sqrt{\frac{\hbar}{2m_e\omega_l}}. \quad (44)$$

Three independent ratios of these lengths determine three independent dimensionless parameters that fully characterize the problem of the 2D Fröhlich polaron.

TABLE I. Comparison of key parameters of the polaron problem in 3D and 2D polar materials. Here, $1/\varepsilon^* = 1/\varepsilon_\infty - 1/\varepsilon_0$ and $1/\varepsilon^*(q) = 1/\varepsilon_\infty(q) - 1/\varepsilon_0(q)$.

Quantity		3D	2D
Material parameters	High-frequency screening	ε_∞	$\varepsilon_\infty(q) = \varepsilon(1 + r_\infty q)$
	Static screening	ε_0	$\varepsilon_0(q) = \varepsilon(1 + r_0 q)$
	TO phonon frequency	ω_{TO}	ω_t
LO phonon dispersion	Lyddane-Sachs-Teller relation	$\omega_{\text{LO}} = \omega_{\text{TO}} \sqrt{\varepsilon_0/\varepsilon_\infty}$	$\omega_{l,q} = \omega_t \sqrt{\varepsilon_0(q)/\varepsilon_\infty(q)}$
Fröhlich coupling constant		$\alpha = \frac{e^2}{4\pi\epsilon_0\varepsilon^*} \sqrt{\frac{m_e}{2\hbar^2\omega_{\text{LO}}}}$	$\alpha(q) = \frac{e^2}{4\pi\epsilon_0\varepsilon^*(q)} \sqrt{\frac{m_e}{2\hbar^2\omega_{l,q}}}$
Electron-phonon coupling		$V_q = -\frac{i}{q} \left[\frac{\alpha\hbar}{V} \hbar\omega_{\text{LO}} \sqrt{\frac{\hbar\omega_{\text{LO}}}{2m_e}} \right]^{\frac{1}{2}}$	$V_q = \left[\frac{\pi\alpha(q)\hbar}{Aq} \hbar\omega_{l,q} \sqrt{\frac{2\hbar\omega_{l,q}}{m_e}} \right]^{\frac{1}{2}}$
Length scales	Polaron radius	$l_e = \sqrt{\frac{\hbar}{2m_e\omega_{\text{LO}}}}$	$l_t = \sqrt{\frac{\hbar}{2m_e\omega_t}}$
	Bohr radius	$a_B = 4\pi\epsilon_0 \frac{\hbar^2}{e^2 m_e}$	$a_B = 4\pi\epsilon_0 \varepsilon \frac{\hbar^2}{e^2 m_e}$
	High-frequency screening length		r_∞
Effective coupling strength	Static screening length		r_0
		$\alpha = \frac{l_e}{a_B} \left[\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0} \right]$	$\alpha_m = \frac{r_l}{a_B} \frac{\sqrt{r_0} - \sqrt{r_\infty}}{\sqrt{r_0} + \sqrt{r_\infty}} > \alpha(q)$

Our choice of these three parameters is based on the following observation. By inspecting the q dependence of the coupling constant in Eq. (41), we notice that $\alpha(q)$ is bounded from above. First, the frequency of LO phonons always satisfies the inequality $\omega_{l,q} \geq \omega_t$. Second, the factor $\varepsilon_\infty^{-1}(q) - \varepsilon_\infty^{-1}(q)$ vanishes in the limits $q \rightarrow 0$ and $q \rightarrow \infty$, and has a single maximum at $q = q_m = 1/\sqrt{r_0 r_\infty}$. Therefore, by replacing $\omega_{l,q} \rightarrow \omega_t$ and evaluating $\varepsilon_\infty^{-1}(q) - \varepsilon_0^{-1}(q)$ at $q = q_m$, we find the following upper bound for $\alpha(q)$:

$$\alpha(q) < \alpha_m = \frac{r_l}{a_B} \frac{\sqrt{r_0} - \sqrt{r_\infty}}{\sqrt{r_0} + \sqrt{r_\infty}}. \quad (45)$$

In the following, we choose the value of α_m as the first dimensionless parameter characterizing the system, while the other two are defined by the ratios

$$\sigma_0 = \frac{r_0}{r_\infty} > 1, \quad \sigma_t = \frac{r_l}{r_\infty}. \quad (46)$$

On physical grounds, it is natural to expect that α_m indeed provides a proper measure of the coupling strength, which generalizes the notion of Fröhlich constant to the case of 2D polar materials [49]. This expectation is confirmed in the following sections by explicitly analyzing the polaron problem. We will see that the polaron energy depends most strongly on α_m with the value of $\alpha_m \sim 5$ separating the weak- and strong-coupling regimes. Moreover, for any value of α_m , the dependency on the other parameters is moderate.

We conclude this section by collecting in Table I a concise side-by-side list of key differences between the electron-LO-phonon problems in 3D and 2D polar crystals.

IV. POLARON PROBLEM IN WEAK- AND STRONG-COUPLING LIMITS

A. Weak-coupling regime: second-order perturbation theory

In the limit $\alpha_m \ll 1$, we adopt the perturbation theory, assuming the unperturbed ground state, which consists of an electron with zero momentum and the phonon vacuum, $|0\rangle = |\mathbf{P}=0\rangle \otimes |0\rangle_{\text{ph}}$. Since the interaction vertex $V_q(e^{i\mathbf{q}\cdot\hat{\mathbf{r}}_e}\hat{a}_{\mathbf{q}} - e^{-i\mathbf{q}\cdot\hat{\mathbf{r}}_e}\hat{a}_{\mathbf{q}}^\dagger)$ changes the phonon number by ± 1 , the first-order correction to the energy vanishes. The leading energy shift

appears in the second order,

$$\Delta E_0 = - \sum_{n \neq 0} \frac{|\langle n | \hat{H}_{\text{int}} | 0 \rangle|^2}{E_n^{(0)} - E_0^{(0)}}, \quad (47)$$

where the intermediate states $|n\rangle$ contain one LO phonon with the wave vector $\mathbf{q}$ and the electron recoiling with momentum $-\mathbf{q}$. Evaluating the matrix elements, we get the following result:

$$\Delta E_0 = - \sum_{\mathbf{q}} \frac{|V_q|^2}{\frac{\hbar^2 q^2}{2m_e} + \hbar\omega_{l,q}}. \quad (48)$$

After replacing summation by integration $\sum_{\mathbf{q}} \rightarrow \frac{A}{(2\pi)^2} \int d^2 q$, using Eq. (43) to express V_q in terms of $\alpha(q)$, the perturbative correction to the energy reduces to the following form:

$$\Delta E_0 = -\hbar \int_0^\infty dq \frac{\alpha(q) \hbar\omega_{l,q}}{\frac{\hbar^2 q^2}{2m_e} + \hbar\omega_{l,q}} \sqrt{\frac{\hbar\omega_{l,q}}{2m_e}}, \quad (49)$$

where we performed a trivial angular integration. Finally, we change the integration variable $p = r_\infty q$ and express the polaron energy shift in terms of dimensionless parameters introduced in Sec. III D,

$$E_0 = \Delta E_0 = -\alpha_m \hbar\omega_t \mathcal{I}_0(\sigma_0, \sigma_t), \quad (50)$$

where the dimensionless integral coefficient reads

$$\mathcal{I}_0(\sigma_0, \sigma_t) = \int_0^\infty \frac{\sigma_t(\sqrt{\sigma_0} + 1)^2 p dp}{(1+p)(1+\sigma_0 p)(\sigma_t^2 p^2/\Omega_p + 1)}, \quad (51)$$

with $\Omega_p = \sqrt{(1+\sigma_0 p)/(1+p)}$ being the dimensionless frequency of LO phonons.

It is clear from Eq. (50) that the parameter α_m indeed plays the same role as the coupling constant in the 3D Fröhlich problem. The remaining integral coefficient defined by Eq. (51) turns out to be a well-behaved dimensionless function of the order of unity in a wide range of its arguments. This allows us to roughly estimate the polaron binding energy as $E_b \approx \alpha_m \hbar\omega_t$.

Figure 3 displays a color map of $\mathcal{I}_0(\sigma_0, \sigma_t)$ over a range of material parameters σ_0 and σ_t that vary by an order of

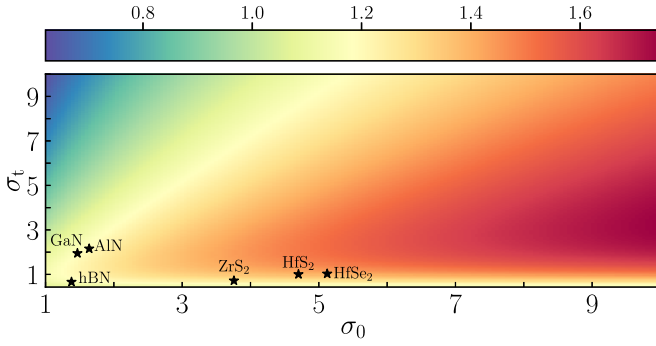


FIG. 3. Colour map of the dimensionless weak-coupling integral $I_0(\sigma_0, \sigma_t)$ defined in Eq. (51). The horizontal axis is the ratio σ_0 of static to high-frequency screening lengths; the vertical axis is σ_t . Stars indicate the realistic parameter sets for hBN, GaN, AlN, ZrS₂, HfS₂, and HfSe₂. The weak dependence on both variables (the color scale spans only a factor of 2) justifies treating α_m as the principal measure of the electron-phonon coupling in the perturbative regime.

magnitude. The stars indicate the positions of the specific 2D materials analyzed in Table II. Throughout the region, the function $I_0(\sigma_0, \sigma_t)$ varies only moderately, taking values of order unity, which confirms the interpretation of α_m as the effective coupling constant controlling the overall magnitude of the polaron energy shift. We will see that this conclusion is valid for any coupling strength.

It is well known that for 3D polar crystals, the polaron energy shift calculated as the lowest-order perturbative correction is exactly reproduced within the so-called intermediate coupling variational theory by Lee, Low, and Pines [8]. In 2D, the situation is the same. In fact, the perturbative result of Eq. (50) exactly coincides with the polaron energy obtained in Ref. [41] within the variational LLP approach. The variational derivation of the perturbative result explains why the linear dependence of the polaron energy extends to quite large coupling both in 3D and in 2D, as we will see in Sec. VI.

B. Strong-coupling limit (Landau-Pekar model)

In the strong-coupling regime $\alpha_m \gg 1$, the adiabatic approximation for the phonon subsystem is justified, and the

polaron binding energy can be estimated using the Landau-Pekar energy functional (see, e.g., Ref. [3]):

$$E_e^{\text{LP}} = \langle \psi | \frac{\hat{p}_e^2}{2m_e} | \psi \rangle - \sum_{\mathbf{k}} \frac{|V_{\mathbf{k}}|^2 |g_{\mathbf{k}}|^2}{\hbar \omega_{\mathbf{k}}}. \quad (52)$$

Here, $\psi(r)$ is the wave function of the self-trapped electron, and the corresponding form factor is defined as

$$g_{\mathbf{k}} = \langle \psi | e^{i\mathbf{k} \cdot \mathbf{r}} | \psi \rangle. \quad (53)$$

1. Hydrogenlike trial wave function

For the simplest and frequently used hydrogenic trial wave function [36] in 2D,

$$\psi_H = \sqrt{\frac{2}{\pi}} \frac{1}{a} e^{-r/a}, \quad (54)$$

the kinetic energy is

$$\langle \psi_H | \frac{\hat{p}_e^2}{2m_e} | \psi_H \rangle = \frac{\hbar^2}{2m_e a^2}, \quad (55)$$

and the density form factor reads

$$g_q = \frac{1}{(1 + (aq)^2/4)^{3/2}}. \quad (56)$$

Hence, the Landau-Pekar energy as a function of the variational parameter a is given as follows:

$$E_{\text{LP}}^H(a) = \frac{\hbar^2}{2m_e a^2} - \sum_{\mathbf{q}} \frac{\pi \hbar \alpha(q)}{A q} \sqrt{\frac{2 \hbar \omega_{\mathbf{q}}}{m_e}} \frac{1}{[1 + (aq)^2/4]^3}, \quad (57)$$

where we used Eq. (43) and expressed V_q in terms of the nonlocal coupling constant $\alpha(q)$ and the frequency $\omega_{\mathbf{q}}$. Following the same steps as in the previous subsection, we convert sum to integral, express everything in terms of the three dimensionless parameters, and arrive at the final result

$$E_{\text{LP}}^H(a) = \frac{\hbar^2}{2m_e a^2} - \alpha_m \hbar \omega_t \mathcal{I}_{\text{LP}}^H(a, \sigma_0, \sigma_t), \quad (58)$$

TABLE II. Polaron binding energies and effective masses for selected monolayers. For each carrier (electron or hole), we list the binding energy obtained in the weak-coupling approximation (E_0), the strong-coupling/Landau-Pekar approximation with a *Gaussian* trial state (E_{LP}), and the Feynman variational result (E_F), all in meV. The effective masses are in units of free electron mass m_0 . Material parameters taken from experimental and first-principles calculation data for a set of 2D polar materials: TO phonon energy $\hbar \omega_t$, screening lengths r_0 , r_∞ ^a, and electron and hole effective masses m_e , m_h (taken from Ref. [36]).

Mat.	E_{LP}^e	E_0^e (meV)	E_F^e	E_{LP}^h	E_0^h (meV)	E_F^h	m_e	m_0^*	m_e^F	m_h	m_0^*	m_h^F	α_m^e	α_m^h	$\hbar \omega_t$ (meV)	r_0 (nm)	r_∞ (nm)
hBN	15.00	129.3	131.6	9.433	116.3	118.2	0.83	1.00	1.04	0.65	0.77	0.80	0.65	0.56	172.3 [32]	1.076 [32]	0.780 [32]
GaN	0.647	55.16	56.4	44.78	132.0	138.4	0.24	0.29	0.30	1.35	2.02	2.49	0.64	1.51	73.30 [50]	1.109	0.756
AlN	12.52	105.4	110.1	80.98	186.5	201.4	0.51	0.71	0.83	1.49	2.53	3.99	1.19	2.03	74.15 [51]	0.763	0.466
HfSe ₂	71.55	89.56	101.6	81.82	98.51	112.2	0.18	0.43	1.26	0.23	0.57	1.78	5.75	6.50	11.10 [52]	21.75	4.246
HfS ₂	92.04	123.2	137.7	127.9	155.1	174.6	0.24	0.54	1.32	0.44	1.07	3.02	5.00	6.77	17.81 [53]	13.98	2.974
ZrS ₂	89.92	142.9	153.2	80.58	133.7	143.2	0.31	0.58	0.93	0.26	0.48	0.74	3.80	3.48	29.82 [54]	10.62	2.826

^aThe screening lengths are extracted from Ref. [36] as $r_{0[\infty]} = \epsilon_{0[\infty]} d/2$, where $\epsilon_{0[\infty]}$ is the static (high-frequency) dielectric constant and d is the monolayer thickness.

where the dimensionless integral in the second term reads

$$\mathcal{I}_{\text{LP}}^{\text{H}} = \int_0^\infty \frac{\sigma_t(\sqrt{\sigma_0} + 1)^2 dp}{(1+p)(1+\sigma_0 p)[1+(pa/2r_\infty)^2]^3}. \quad (59)$$

Minimizing the function Eq. (57) [or, equivalently, Eq. (58)] with respect to a gives the polaron binding energy in the strong-coupling limit. The above Landau-Pekar type theory with the hydrogenic trial function can be directly compared with similar calculations in Ref. [36] based on a simplified model of electron-LO phonon coupling that neglects the phonon dispersion. As we have demonstrated recently [41], for sufficiently strong coupling, the correct treatment of the dispersion and the related nonlocality of the coupling constant becomes highly important (see the Supplemental Material in Ref. [41]).

2. Gaussian trial wave function

Another popular variational ansatz, which typically gives lower energy, employs the normalized Gaussian that in 2D reads

$$\psi_{\text{G}}(\mathbf{r}) = \sqrt{\frac{1}{\pi a^2}} e^{-r^2/2a^2}. \quad (60)$$

The corresponding expectation value of the kinetic energy coincides with the one for hydrogenic ansatz,

$$\langle \psi_{\text{G}} | \frac{\hat{p}_e^2}{2m_e} | \psi_{\text{G}} \rangle = \frac{\hbar^2}{2m_e a^2}, \quad (61)$$

while the form factor is now

$$g_q(a) = e^{-a^2 q^2/4}. \quad (62)$$

Substituting Eqs. (61) and (62) in the Landau-Pekar functional and expressing the coupling through the nonlocal Fröhlich constant, we obtain the following variational energy:

$$E_{\text{LP}}^{\text{G}}(a) = \frac{\hbar^2}{2m_e a^2} - \sum_q \frac{\pi \hbar \alpha(q)}{A q} \sqrt{\frac{2\hbar\omega_{\text{L},q}}{m_e}} e^{-a^2 q^2/2}, \quad (63)$$

which eventually reduces to the form similar to Eq. (58),

$$E_{\text{LP}}^{\text{G}}(a) = \frac{\hbar^2}{2m_e a^2} - \alpha_m \hbar \omega_{\text{L}} \mathcal{I}_{\text{LP}}^{\text{G}}(a, \sigma_0, \sigma_t), \quad (64)$$

where for the Gaussian trial function, the dimensionless integral reads

$$\mathcal{I}_{\text{LP}}^{\text{G}} = \int_0^\infty \frac{\sigma_t(\sqrt{\sigma_0} + 1)^2 p}{(1+p)(1+\sigma_0 p)} \exp\left(-\frac{p^2 a^2}{2r_\infty^2}\right) dp. \quad (65)$$

By minimizing the energy of Eq. (64) with respect to the variational parameter a , we get the Landau-Pekar polaron energy in the Gaussian trial class. As we have already mentioned, the Gaussian ansatz yields a better energy estimate compared to the hydrogenic one (see Sec. IV and Fig. 5). However, most importantly, it corresponds to the exact strong-coupling limit of Feynman's variational theory, which we consider in the following sections.

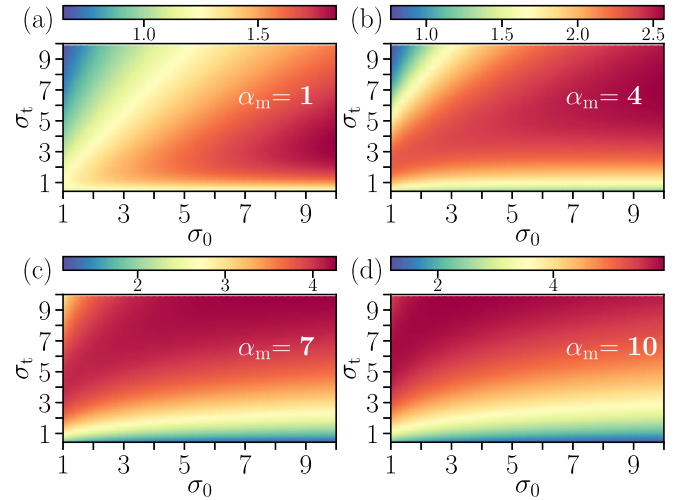


FIG. 4. Normalized energy $E_F^e/E_F^e(\sigma_0 = 1, \sigma_t = 0.5)$ for four values of the coupling prefactor α_m : (a) $\alpha_m = 1$, (b) $\alpha_m = 4$, (c) $\alpha_m = 7$, and (d) $\alpha_m = 10$. The color scale (different in each panel) indicates the magnitude of the normalized energy over the plane spanned by the ratios of characteristic length scales, $\sigma_0 = r_0/r_\infty$ and $\sigma_t = r_t/r_\infty$. For weak coupling, it remains close to unity, reproducing the perturbative behavior. Stronger coupling progressively amplifies the dependence on the material parameters, but it still remains moderate.

V. FEYNMAN VARIATIONAL SOLUTION OF THE POLARON PROBLEM IN 2D

Feynman's variational solution of the polaron problem, based on the path-integral formalism [9], is known to provide a high-quality all-coupling theory that smoothly interpolates between weak- and strong-coupling limits and perfectly agrees with essentially exact Monte Carlo results for both the bulk 3D and the surface quasi-2D polarons [11–13]. In this section, we adapt this approach to the case of genuine 2D polarons. As the standard derivation [9,55] of the corresponding variational problem assumes from the very beginning dispersionless phonons and q -independent Fröhlich coupling, its 2D generalization requires some care. Therefore, in the following, we carefully adapt the key steps of the Feynman derivation to our specific situation, avoiding unnecessary assumptions. This in particular allows us to keep the paper fully self-contained.

A. Effective action

Following Feynman's variational path-integral approach [9,55], in the 2D case, we start from the electron-phonon Lagrangian of Eq. (35) and rotate to imaginary time $t \rightarrow -iu$ to obtain the path-integral representation of the partition function. After that, the LO phonon coordinates appearing in Eq. (35) are integrated out, and Gaussian elimination yields a nonlocal Euclidean action for the electron trajectory $\mathbf{r}_e(u)$,

$$S_{\text{eff}}[\mathbf{r}_e] = \int_0^{\beta\hbar} du \frac{m_e}{2} \dot{\mathbf{r}}_e^2(u) - \frac{1}{2\hbar} \int_0^{\beta\hbar} dt \int_0^{\beta\hbar} ds \times \sum_{\mathbf{k}} |V_{\mathbf{k}}|^2 e^{-\omega_{\text{L},\mathbf{k}}|t-s|} e^{i\mathbf{k} \cdot [\mathbf{r}_e(t) - \mathbf{r}_e(s)]}, \quad (66)$$

where $|V_{\mathbf{k}}|^2$ and $\omega_{\text{L},\mathbf{k}}$ are, respectively, the 2D coupling amplitude and the frequency of LO phonons, defined by Eqs. (42)

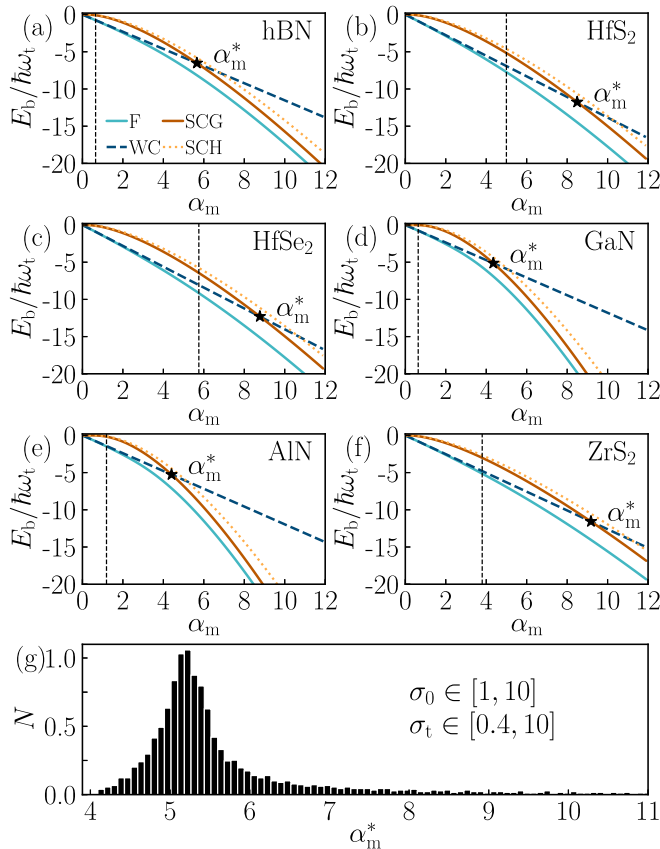


FIG. 5. (a)–(f) Binding energy as a function of the coupling constant α_m for six pairs of parameters (σ_0, σ_t) corresponding to indicated polar monolayers. The legend in panel (a) refers to all other panels, namely, solid turquoise (F): Feynman variational result; dashed blue (WC): weak-coupling approximation; solid orange (SCG): strong-coupling approximation with Gaussian trial function; dotted orange (SCH): strong-coupling approximation with hydrogenic trial function. The vertical dashed line indicates the physical value α_m^* for each material. The star marks the crossover α_m^* where the strong-coupling branch overtakes the weak-coupling one. (g) Histogram of α_m^* obtained from a random scan over the region $\sigma_0 \in [1, 10]$ and $\sigma_t \in [0.4, 10]$ in the (σ_0, σ_t) plane.

and (43) in Sec. III. Equation (66) is the starting point for the all-coupling variational analysis used below. The intermediate steps in the derivation of Eq. (66) are presented in Appendix B 1.

B. Upper bound for the ground-state energy

The ground-state energy E , corresponding to the action $S_{\text{eff}}[\mathbf{r}]$, can be obtained from the Feynman variational principle, which states that for a trial/reference action $S_0[\mathbf{r}]$ the following inequality holds true [55]:

$$E \leq E_0 + \lim_{\beta \rightarrow \infty} \frac{1}{\beta \hbar} \langle S_{\text{eff}} - S_0 \rangle_0, \quad (67)$$

where E_0 is the ground-state energy of the reference system, $\beta = 1/(k_B T)$ is the inverse temperature with k_B denoting the Boltzmann constant, and $\langle \cdots \rangle_0$ stands for the averaging with the reference action S_0 .

Here, we follow the original Feynman's proposal and use the quadratic reference action that preserves translational invariance and captures retardation,

$$S_0[\mathbf{r}_e] = \int_0^{\beta \hbar} du \frac{m_e}{2} \dot{\mathbf{r}}_e^2(u) + \frac{C}{2} \int_0^{\beta \hbar} du \int_0^{\beta \hbar} du' \times e^{-W|u-u'|} |\mathbf{r}_e(u) - \mathbf{r}_e(u')|^2, \quad (68)$$

characterized by two positive parameters C and W . It is customary to reparametrize the strength C of the effective harmonic interaction in Eq. (68), using the following combination:

$$V = \sqrt{W^2 + \frac{4C}{m_e W}} > W, \quad (69)$$

as the second independent parameter. In particular, in terms of the parameters V and W , the ground-state energy of the reference state takes the following simple form: $E_0 = \hbar(V - W)$ [55].

An explicit calculation of the right-hand side in Eq. (67) is presented in Appendix B 2. The final variational upper bound for the ground-state energy, expressed in terms of dimensionless variational parameters,

$$v = \frac{V}{\omega_t}, \quad w = \frac{W}{\omega_t}, \quad (70)$$

the effective 2D Fröhlich coupling constant α_m , and the pair of material parameters (σ_0, σ_t) , can be represented as follows:

$$E \leq \hbar\omega_t \frac{(v - w)^2}{2v} - \hbar\omega_t \alpha_m \mathcal{I}_F(\sigma_0, \sigma_t; v, w), \quad (71)$$

with the dimensionless function

$$\begin{aligned} \mathcal{I}_F(\sigma_0, \sigma_t; v, w) &= \int_0^\infty d\tau \int_0^\infty dp \frac{\sigma_t(\sqrt{\sigma_0} + 1)^2 p e^{-\tau}}{(1+p)(1+\sigma_0 p)} \\ &\times \exp \left\{ -\frac{\sigma_t^2 p^2}{\Omega_p} \left[\frac{w^2}{v^2} \tau + \Omega_p \frac{v^2 - w^2}{v^3} (1 - e^{-v\tau/\Omega_p}) \right] \right\}. \end{aligned} \quad (72)$$

The variational problem defined by Eqs. (71) and (72) generalizes the all-coupling Feynman solution of the polaron problem to the case of 2D polar materials. Minimization of the right-hand side with respect to the variational parameters v and w yields the Feynman estimate E_F of the ground-state energy of the 2D Fröhlich polaron.

It is easy to see that in the weak- and strong-coupling regimes, the Feynman solution recovers the results of Sec. IV. In the limit $\alpha_m \ll 1$, the right-hand side in Eq. (71) is dominated by the first term, which is minimized when $v = w$. The polaron energy is then given by the second term evaluated at this point. By setting $v = w$ and performing the trivial integration over τ in Eq. (72), we find

$$\mathcal{I}_F(\sigma_0, \sigma_t; v, v) = \mathcal{I}_0(\sigma_0, \sigma_t), \quad (73)$$

where $\mathcal{I}_0(\sigma_0, \sigma_t)$ is defined by Eq. (51), which is the weak-coupling result derived in Sec. IV A.

In the opposite, strong-coupling regime $\alpha_m \gg 1$, the phonon cloud is tightly bound to the electron. Variationally,

this corresponds to $w = 0$ with $v \gg 1$. In this case, the first term in Eq. (71) reduces to $\hbar\omega_t v/2$, whereas the integral in Eq. (72) simplifies as

$$\mathcal{I}_F \approx \int_0^\infty dp \frac{\sigma_t(\sqrt{\sigma_0} + 1)^2 p e^{-\frac{\sigma_t^2 p^2}{v}}}{(1+p)(1+\sigma_0 p)}. \quad (74)$$

After identification $v = \hbar/(m_e \omega_t a^2)$, we observe that in this limit the upper bound of Eq. (71) coincides with the Landau-Pekar functional (64) obtained in Sec. IV B for the Gaussian trial function.

Apparently, for intermediate couplings $0 < \alpha_m < \infty$, Eqs. (71) and (72) provide a smooth interpolation between the above two limits. In general, the effective 2D coupling constant α_m defined in Sec. III D enters the energy of Eq. (71) only as a prefactor in front of a dimensionless integral $\mathcal{I}_F(v, w)$ that depends on the variational parameters v and w . However, in contrast to the 3D case, the integral $\mathcal{I}_F$ defined in Eq. (72) also contains the remaining two material parameters σ_0 and σ_t , which is a specific 2D feature.

In Sec. III A, we have demonstrated that the strongest dependence on the material-specific parameters is encoded in the effective coupling constant α_m , whereas the variation of polaron energy over the (σ_0, σ_t) plane is moderate. This tendency is preserved for larger coupling. Figure 4 illustrates the dependence of the polaron energy obtained from Eq. (71) on σ_0 and σ_t . Each panel displays a color map of the ratio $E_F/E_F(\sigma_0 = 1, \sigma_t = 0.5)$ for four representative coupling strengths $\alpha_m = 1, 4, 7$, and 10. We observe that all panels look very similar, with a slight tendency of increasing the variations over the (σ_0, σ_t) plane at larger couplings. In all panels in Fig. 4 throughout the shown region, the polaron energy changes at most by a factor of 2. On the other hand, it increases by more than an order of magnitude when α_m goes from 1 to 10 (see the next section). Therefore, even well beyond the weak-coupling regime, the effective coupling constant α_m alone provides a good overall characterization of the coupling strength. However, for the quantitative description of the 2D polaron, the dependence on the other parameters becomes important, especially when the strong-coupling physics comes into play.

C. Renormalization of the polaron mass

To obtain the dynamical mass of the polaron, we use Feynman's uniform-drift construction [9]. The system is set to a steady motion with velocity $\mathbf{U}$ by shifting the electron trajectory as $\mathbf{r}_e(u) \rightarrow \mathbf{r}_e(u) + \mathbf{U}u$. Then, we compute the leading correction $\sim U^2$ to the energy and identify the corresponding coefficient with the effective mass. The above trajectory shift, first, generates the trivial correction $m_e U^2/2$ to the kinetic energy and, second, multiplies each interaction vertex by the phase factor $\exp[i\mathbf{k} \cdot \mathbf{U}(u - u')]$. The detailed calculation of the modified upper bound as a function of velocity, including the explicit expansion in powers of U , is given in Appendix B 3. The result up to the second order in U reads as follows [cf. Eq. (71)]:

$$E(U) \leq \frac{m_e^* U^2}{2} + \hbar\omega_t \frac{(v - w)^2}{2v} - \hbar\omega_t \alpha_m \mathcal{I}_F, \quad (75)$$

where $\mathcal{I}_F$ is defined by Eq. (72), and the renormalized electron mass is $m_e^* = m_e + \delta m$ with polaron shift

$$\delta m(v, w) = \alpha_m m_e \mathcal{M}_F(\sigma_0, \sigma_t; v, w), \quad (76)$$

where

$$\begin{aligned} \mathcal{M}_F(\sigma_0, \sigma_t; v, w) &= \int_0^\infty d\tau \int_0^\infty dp \frac{\sigma_t^3(\sqrt{\sigma_0} + 1)^2 p^3 \tau^2 e^{-\tau}}{(1 + \sigma_0 p)^2} \\ &\times \exp \left\{ -\frac{\sigma_t^2 p^2}{\Omega_p} \left[\frac{w^2}{v^2} \tau + \Omega_p \frac{v^2 - w^2}{v^3} (1 - e^{-v\tau/\Omega_p}) \right] \right\}. \end{aligned} \quad (77)$$

The value of δm evaluated at the optimal point (v^*, w^*) that minimizes the energy bound in Eq. (71) determines the effective mass of the Fröhlich polaron in 2D crystals.

In the weak-coupling regime $v^* = w^*$, and the integral over τ in Eq. (B32) can be easily calculated. By performing the integration, we recover the mass renormalization obtained recently using the variational approach of LLP [41]. That is, $\mathcal{M}_F(\sigma_0, \sigma_t; v^*, w^*) = \mathcal{M}_{\text{LLP}}(\sigma_0, \sigma_t)$, where

$$\mathcal{M}_{\text{LLP}}(\sigma_0, \sigma_t) = \int_0^\infty \frac{\sigma_t^3(\sqrt{\sigma_0} + 1)^2 p^3 dp}{(1 + \sigma_0 p)^2 (1 + \sigma_t^2 p^2 / \Omega_p)^3}. \quad (78)$$

At stronger couplings, the effective mass increases and, similarly to the binding energy, the growth of m_e^* is governed primarily by α_m , with a moderate dependence on σ_0 and σ_t .

VI. NUMERICAL RESULTS

The variational formalism developed in Sec. V can be applied to real materials, provided that the parameters characterizing a polar monolayer—the energy $\hbar\omega_t$ of TO phonons, the static and high-frequency screening lengths r_0 and r_∞ , and the electron (or hole) mass $m_{e(h)}$ —are specified. In Table II, we summarize the input data collected from the literature for a representative set of 2D polar materials, together with the corresponding results for the energies and effective masses of polarons. For each material, we show the binding energy of the electron and hole polarons in three approximations: the weak-coupling limit ($E_0^{e(h)}$), which gives results identical to LLP approximation [41], the strong-coupling/Landau-Pekar limit ($E_{\text{LP}}^{e(h)}$), and the full Feynman variational result ($E_F^{e(h)}$). The same hierarchy is reported for the effective masses: the band mass $m_{e(h)}$, the polaron mass calculated within the weak-coupling/LLP approach $m_{e(h)}^{\text{LLP}}$, and the variational mass $m_{e(h)}^F$. All strong coupling results in Table II are obtained with the Gaussian trial wave function because, compared to the hydrogenic ansatz, it systematically produces lower variational energies and thus stronger binding. In particular, the ratio $E_{\text{LP}}^G/E_{\text{LP}}^H$ equals 1.248 for hBN, 1.121 for HfS₂, 1.117 for HfSe₂, 1.408 for GaN, 1.304 for AlN, and 1.123 for ZrS₂ (see also Fig. 5 and discussion below).

The results presented in Table II show that for materials with a small effective coupling constant, such as hBN and GaN in which $\alpha_m \lesssim 1$, the Feynman energies are within a few percent of the weak-coupling predictions, while the Landau-Pekar approximation dramatically, up to an order of magnitude, underestimates the binding energy. Materials with

a larger difference between the static and high-frequency screening lengths, and softer phonons, such as HfSe_2 , HfS_2 , and ZrS_2 , have a larger effective coupling constant $\alpha_m \sim 4-5$. In these materials, the accuracy of the weak-coupling results worsens, being within 5%–10% of the Feynman estimate. On the other hand, the quality of the Landau-Pekar energy improves, but for all studied materials the weak-coupling/LLP gives better results than the adiabatic strong-coupling theory. In all cases, including weakly coupled materials, the weak-coupling/LLP results for the polaron mass are less accurate than those for the binding energy. In fact, the deviation of the LLP and Feynman estimates for the effective mass ranges from 4% in hBN to more than a factor of 3 in HfS_2 .

To illustrate the dependence of the polaron binding energy on the effective coupling constant α_m , we take six pairs of parameters (σ_0, σ_t) corresponding to specific materials from Table II. Then, for each pair, we plot the polaron energy, obtained from the accurate all-coupling variational theory of Sec. V, as a function of α_m , together with the results of the weak- and strong-coupling approximations. Figure 5 summarizes these results for the case of electron polaron. Panels (a)–(f) correspond to the pairs (σ_0, σ_t) of screening parameters for the materials indicated on the panel. The vertical dashed line shows the physical α_m for each material.

The solid turquoise line is the full result of the Feynman-type variational theory, developed in Sec. V. The dashed blue line is the weak-coupling approximation of Sec. III A, which can also be restated in a variational form [41]. Finally, the solid orange line shows the strong-coupling Landau-Pekar energy obtained with the Gaussian trial wave function. For comparison, the dotted orange line shows the Landau-Pekar energy computed with the hydrogenic trial function. Apparently, the Gaussian ansatz in the Landau-Pekar functional is always superior over the hydrogenic one. In the following, when referring to the strong coupling approximation, we always mean the Gaussian trial function. The asterisk in panels (a)–(f) of Fig. 5 indicates the intersection of the weak- and strong-coupling curves, while α_m^* is the crossover value of the coupling constant above which the strong-coupling ansatz variationally wins over the weak-coupling approximation and becomes closer to apparently the best result of Feynman's theory. The lower panel shows the distribution of the crossover coupling constant α_m^* obtained by scanning over the broad ranges of screening parameters $\sigma_0 \in [1, 10]$ and $\sigma_t \in [0.4, 10]$ in the (σ_0, σ_t) plane. The distribution clearly shows that most of the systems cluster around $\alpha_m^* \approx 5$. Remarkably, this value is about twice the smaller than the crossover value of $\alpha_{3D}^* = 3\pi \approx 10$ for bulk 3D polarons, but somewhat larger than the threshold coupling constant $\alpha_{\text{surf}}^* = 4$ for surface polarons. This last value can be deduced as the crossover value between the strong- and weak-coupling limits of the polaron binding energy [20]. Therefore, in a sense, the Fröhlich polarons in genuine 2D materials take an intermediate position between the bulk and surface polarons in 3D polar crystals.

VII. CONCLUSION

We presented a macroscopic theory of Fröhlich polarons in strictly 2D polar crystals. By construction, in our approach, the quantitative description of polaron physics requires only

the knowledge of experimentally accessible macroscopic polarizabilities and effective masses. The required material parameters can also be obtained routinely using standard first-principles methods.

Starting from a long-wavelength continuum description of optical phonons in 2D, we derived an effective Lagrangian of LO phonons, interacting with electrons. It turns out that in 2D polar materials, the electron-LO phonon interaction is characterized by three independent dimensionless parameters. We have identified one of them, α_m in Eq. (45), as an effective coupling constant that provides a natural measure of interaction strength and can be viewed as a 2D generalization of the standard Fröhlich constant. The dependence on the remaining two parameters appears to be moderate, whereas it becomes quantitatively more important at stronger coupling.

Aimed at a high-quality description at arbitrary coupling strength, we adapt and apply Feynman's variational path-integral framework to the case of genuine 2D polarons; we derived the two-parameter variational functional for the polaron energy and the generalization of Feynman result for the polaron mass. By analyzing the dependence of the polaron energy on the effective coupling strength, we identified the value of $\alpha_m^* \approx 5$ above which the weak-coupling regime crosses over into the regime of strongly coupled polarons. However, none of the 2D materials currently known and typically discussed in the literature [18,41] has the coupling constant that exceeds the strong-to-weak coupling threshold. In most cases, the weak-coupling approach [41] provides reasonable estimates of the polaron energies, close to the Feynman variational results. However, the polaron mass shows a much stronger dependence on the coupling strength. In fact, for some materials studied in this work, we observed that the weak-coupling approximation gives a polaron mass that is up to 3–4 times smaller compared to the accurate variational estimate.

In conclusion, the present framework provides a transparent and accurate tool for analyzing charge transport, optical response, and many-body phenomena in polar monolayers. By construction, the present macroscopic approach is restricted to long-wavelength optical phonons, which is sufficient to capture polaron effects in most realistic situation. In addition, for the sake of simplicity, we considered the case of macroscopically isotropic/cubic materials with a single LO phonon mode. This limitation can be easily relaxed along the same lines as in the 3D case (see, for example, Ref. [47]). Extensions to finite-temperature, low-symmetry materials, multiple optical branches, and more complex quasiparticles, such as exciton-polarons or trions dressed by LO phonons, will be the subject of future work.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

APPENDIX A: LO PHONON DISPERSION

The Lagrangian of optical phonons is

$$L = \int \mathcal{L} d^2r dz, \quad (\text{A1})$$

where $\mathcal{L}$ is the Lagrangian density defined by Eq. (24). We separate the longitudinal and transverse components of the displacement as $\xi_{\mathbf{q}} = -i\frac{q}{q}\xi_{\mathbf{q}}^{\parallel} + \xi_{\mathbf{q}}^{\perp}$. Using Eq. (27) for the electric field in the plane, we integrate each term separately and get, one by one, the following results:

$$\frac{1}{2} \int \dot{\xi}^2 \delta(z) d^2r dz = \frac{1}{2} \sum_{\mathbf{q}} (|\dot{\xi}_{\mathbf{q}}^{\parallel}|^2 + |\dot{\xi}_{\mathbf{q}}^{\perp}|^2), \quad (\text{A2})$$

$$\frac{\omega_t^2}{2} \int \xi^2 \delta(z) d^2r dz = \frac{\omega_t^2}{2} \sum_{\mathbf{q}} (|\xi_{\mathbf{q}}^{\parallel}|^2 + |\xi_{\mathbf{q}}^{\perp}|^2), \quad (\text{A3})$$

$$\begin{aligned} \gamma \int \xi \cdot \nabla \varphi(\mathbf{r}, 0) \delta(z) d^2r dz \\ = -\omega_t^2 \sum_{\mathbf{q}} \frac{(r_0 - r_{\infty})q}{1 + r_{\infty}q} |\xi_{\mathbf{q}}^{\parallel}|^2, \end{aligned} \quad (\text{A4})$$

$$\begin{aligned} \frac{\alpha_{\infty}}{2} \int [\nabla \varphi(\mathbf{r}, 0)]^2 \delta(z) d^2r dz \\ = \frac{\omega_t^2}{2} \sum_{\mathbf{q}} \frac{(r_0 - r_{\infty})q r_{\infty}q}{(1 + r_{\infty}q)^2} |\xi_{\mathbf{q}}^{\parallel}|^2, \end{aligned} \quad (\text{A5})$$

$$\frac{\varepsilon \epsilon_0}{2} \int (\nabla \varphi)^2 d^2r dz = \frac{\omega_t^2}{2} \sum_{\mathbf{q}} \frac{\varepsilon (r_0 - r_{\infty})q}{(1 + r_{\infty}q)^2} |\xi_{\mathbf{q}}^{\parallel}|^2. \quad (\text{A6})$$

By collecting all terms together in Eq. (24), we arrive at the following result for the Lagrangian of optical phonons:

$$L = L_{\text{TO}} + L_{\text{LO}}, \quad (\text{A7})$$

where

$$L_{\text{TO}} = \frac{1}{2} \sum_{\mathbf{q}} (|\dot{\xi}_{\mathbf{q}}^{\perp}|^2 - \omega_t^2 |\xi_{\mathbf{q}}^{\perp}|^2), \quad (\text{A8})$$

$$L_{\text{LO}} = \frac{1}{2} \sum_{\mathbf{q}} (|\dot{\xi}_{\mathbf{q}}^{\parallel}|^2 - \omega_{l,q}^2 |\xi_{\mathbf{q}}^{\parallel}|^2). \quad (\text{A9})$$

APPENDIX B: DETAILS OF DERIVATION OF FEYNMAN VARIATIONAL SOLUTION TO THE POLARON PROBLEM IN 2D

This Appendix provides details of calculations needed for adapting the Feynman variational approach to the case of 2D Fröhlich polarons. Starting from the real-time Lagrangian in Eq. (35), we perform the Wick rotation and integrate out LO phonons to obtain the nonlocal Euclidean action. Then, for a

quadratic reference action with parameters (C, W) (with $V = \sqrt{W^2 + 4C/(m_e W)}$), compute the Gaussian form factor and the displacement correlator, and eventually express the Feynman upper bound in terms of dimensionless parameters $v = V/\omega_t$, $w = W/\omega_t$, α_m , σ_0 , and σ_t , recovering Eqs. (71)–(72). We also implement a uniform drift to extract the polaron mass function $\mathcal{M}_F$ of Eq. (77). Technical conventions (Fourier, continuum, $\beta \rightarrow \infty$) are stated succinctly, and the resulting formulas underpin the trends summarized in Fig. 4 and used in Sec. IV.

1. Effective action

The first step is to cast the electron-phonon problem in imaginary time, starting from the real-time Lagrangian of Eq. (35) derived in Sec. III B. The corresponding real-time action reads

$$\begin{aligned} S = \int_{t_i}^{t_f} dt \left[\frac{m_e}{2} \dot{\mathbf{r}}_e^2 + \frac{1}{2} \sum_{\mathbf{q}} (\dot{\xi}_{\mathbf{q}}^{\parallel} \xi_{-\mathbf{q}}^{\parallel} - \omega_{l,q}^2 \xi_{\mathbf{q}}^{\parallel} \xi_{-\mathbf{q}}^{\parallel}) \right. \\ \left. + \frac{1}{2} \sum_{\mathbf{q}} g_{\mathbf{q}} (e^{i\mathbf{q} \cdot \mathbf{r}_e} \xi_{\mathbf{q}}^{\perp} + e^{-i\mathbf{q} \cdot \mathbf{r}_e} \xi_{-\mathbf{q}}^{\perp}) \right]. \end{aligned} \quad (\text{B1})$$

Introducing imaginary time u by the standard complex rotation, $t \rightarrow -iu$, $dt = -i du$, we generate the Euclidean action:

$$S \longrightarrow S_E = -iS = \int_0^{\beta\hbar} du \mathcal{L}_E, \quad \beta = \frac{1}{k_B T}, \quad (\text{B2})$$

where $\mathcal{L}_E$ is obtained from the real-time Lagrangian by simply changing the sign in front of kinetic energy terms, $(\dot{})^2 \rightarrow -(\dot{})^2_u$.

The phase factor e^{iS} in the real-time propagator, after the above Wick rotation, becomes a Boltzmann weight in the path-integral representation of partition function,

$$\mathcal{Z} = \int \mathcal{D}\mathbf{r}_e(u) \mathcal{D}\xi_{\mathbf{q}}^{\parallel}(u) e^{-S_E[\mathbf{r}_e, \xi^{\parallel}]/\hbar}. \quad (\text{B3})$$

Performing the Gaussian integration over the phonon coordinates, one obtains an effective action that involves only the electron trajectory,

$$\begin{aligned} S_{\text{eff}}[\mathbf{r}_e] = \int_0^{\beta\hbar} du \frac{m_e \dot{\mathbf{r}}_e^2(u)}{2} - \frac{1}{2\hbar} \int_0^{\beta\hbar} \int_0^{\beta\hbar} dt ds \\ \times \sum_{\mathbf{k}} |V_{\mathbf{k}}|^2 e^{-\omega_{l,k}|t-s|} e^{i\mathbf{k} \cdot [\mathbf{r}_e(t) - \mathbf{r}_e(s)]}. \end{aligned} \quad (\text{B4})$$

This action is used to define the upper bound of the polaron energy within the Feynman variational principle.

2. The upper bound of the polaron ground-state energy

The starting point here is the following relation between the ground-state energy E and the effective action:

$$E = \lim_{\beta \rightarrow \infty} \left[-\frac{1}{\beta} \ln e^{-\beta F} \right], \quad (\text{B5})$$

where $e^{-\beta F} = \mathcal{Z}$ is defined via the path integral,

$$e^{-\beta F} = \int e^{-S_{\text{eff}}[\mathbf{r}_e]/\hbar} \mathcal{D}\mathbf{r}_e. \quad (\text{B6})$$

The Feynman variational principle is formulated as follows. Let us define a trial (reference) action $S_0[\mathbf{r}_e]$, and the corresponding ground-state energy,

$$E_0 = \lim_{\beta \rightarrow \infty} \left[-\frac{1}{\beta} \ln e^{-\beta F_0} \right], \quad (\text{B7})$$

$$e^{-\beta F_0} = \int e^{-S_0[\mathbf{r}_e]/\hbar} \mathcal{D}\mathbf{r}_e. \quad (\text{B8})$$

Then, the upper bound for the exact energy of the ground state is given by the inequality [55]

$$E \leq E_0 + \lim_{\beta \rightarrow \infty} \frac{1}{\beta \hbar} \langle S_{\text{eff}} - S_0 \rangle_0, \quad (\text{B9})$$

where the average with respect to the trial action S_0 is computed as follows:

$$\langle \mathcal{O} \rangle_0 = \frac{\int \mathcal{O} \exp[-S_0/\hbar] \mathcal{D}\mathbf{r}_e}{\int \exp[-S_0/\hbar] \mathcal{D}\mathbf{r}_e}. \quad (\text{B10})$$

The trial action should be defined with some variational freedom under the requirement that it does not restrict the electron motion too strongly, remaining a good variational model.

The Feynman classic approach employs a Gaussian trial action, which allows for an analytic treatment of the resulting path integrals and yields highly accurate results for the polaron energy, effective mass, and related characteristics. Specifically, the trial action is taken in the following form:

$$S_0 = \int_0^{\beta \hbar} \frac{m_e \dot{\mathbf{r}}_e^2(u)}{2} du + \frac{C}{2} \int_0^{\beta \hbar} \int_0^{\beta \hbar} du du' \times e^{-W|u-u'|} |\mathbf{r}_e(u) - \mathbf{r}_e(u')|^2, \quad (\text{B11})$$

where $[W] = \text{s}^{-1}$ and $[C] = \text{kg s}^{-3}$ are two variational parameters.

In order to evaluate the second term in the right-hand side in Eq. (B9), one needs two building blocks, first, the form factor,

$$I(\mathbf{k}; u, v) = \langle e^{i\mathbf{k} \cdot [\mathbf{r}_e(u) - \mathbf{r}_e(v)]} \rangle_0, \quad (\text{B12})$$

and, second, the displacement correlator, which can be found by differentiating $I(\mathbf{k}; u, u')$ with respect to $\mathbf{k}$:

$$\langle |\mathbf{r}_e(u) - \mathbf{r}_e(v)|^2 \rangle_0 = -\nabla_{\mathbf{k}}^2 I(\mathbf{k}; u, u')|_{\mathbf{k}=0}. \quad (\text{B13})$$

Explicitly, the form factor $I(\mathbf{k}; u, u')$ in the limit $\beta \rightarrow \infty$ reads

$$I(\mathbf{k}; u, u') = \exp \left[-\frac{k^2}{2} \left(\frac{W^2 \hbar}{V^2 m_e} |u - u'| + \frac{4C\hbar}{WV^3 m_e^2} [1 - e^{-V|u-u'|}] \right) \right], \quad (\text{B14})$$

where $V = \sqrt{W^2 + 4C/(m_e W)}$.

Having the expression for $I(\mathbf{k}; u, u')$ at hand, we can evaluate the average $\langle S_{\text{eff}} - S_0 \rangle_0$. Rewriting it in terms of $I(\mathbf{k}; u, u')$,

one finds

$$\begin{aligned} \langle S_{\text{eff}} - S_0 \rangle_0 &= - \sum_{\mathbf{k}} \frac{|V_{\mathbf{k}}|^2}{2\hbar} \int_0^{\beta \hbar} du \int_0^{\beta \hbar} du' e^{-\omega_{\mathbf{k}} |u-u'|} I(\mathbf{k}; u, u') \\ &\quad - \frac{C}{2} \int_0^{\beta \hbar} du \int_0^{\beta \hbar} du' e^{-W|u-u'|} [\nabla_{\mathbf{k}}^2 I(\mathbf{k}; u, u')]_{\mathbf{k}=0}, \end{aligned} \quad (\text{B15})$$

We now evaluate three contributions entering the bound of Eq. (B9): (1) the C term originating from S_0 ; (2) the reference ground-state energy E_0 ; and (3) the genuine electron-phonon contribution that contains $|V_{\mathbf{k}}|^2$.

a. The C term

Using the definition of the displacement correlator and the explicit form of Eq. (B14), one finds in the limit $\beta \rightarrow \infty$,

$$\frac{C}{2} \int_0^{\beta \hbar} du \int_0^{\beta \hbar} du' e^{-W|u-u'|} \left[-2 \frac{dI(k, u, u')}{dk^2} \right]_{\mathbf{k}=0} \quad (\text{B16})$$

$$= \frac{2C \beta \hbar^2}{m_e W V} = \hbar^2 \beta \frac{V^2 - W^2}{2V}, \quad (\text{B17})$$

where we expressed the parameter C in terms of V and W as $C = \frac{m_e W}{4} (V^2 - W^2)$.

b. Trial ground-state energy E_0

The free energy $F_0(C)$ of the Gaussian model follows from $\partial_C F_0 = \beta^{-1} \hbar^{-1} \langle \partial_C S_0 \rangle_0$. With the same Gaussian algebra that leads to Eq. (B17), we obtain $\partial_C E_0 = 2\hbar/(m_e W V)$. Integrating this derivative and using the condition $E_0(C=0) = 0$ gives the closed form of the reference energy,

$$E_0 = \hbar (V - W). \quad (\text{B18})$$

c. Electron-phonon part

Only this contribution depends on the phonon dispersion and electron-phonon coupling, and therefore carries all information about specific features of the 2D system. Keeping the explicit coupling amplitude $|V_{\mathbf{k}}|^2$, we define

$$\mathcal{F} \equiv \sum_{\mathbf{k}} \frac{|V_{\mathbf{k}}|^2}{2\hbar} \int_0^{\beta \hbar} du \int_0^{\beta \hbar} du' e^{-\omega_{\mathbf{k}} |u-u'|} I(\mathbf{k}; u, v). \quad (\text{B19})$$

Using the identity

$$\int_0^{\beta \hbar} \int_0^{\beta \hbar} du du' g(|u - u'|) \xrightarrow{\beta \rightarrow \infty} 2\beta \hbar \int_0^{\infty} du g(u) \quad (\text{B20})$$

and the fact that $I(\mathbf{k}; u, u')$ in Eq. (B14) depends on $|u - u'|$, we obtain

$$\begin{aligned} \mathcal{F} &= \beta \sum_{\mathbf{k}} |V_{\mathbf{k}}|^2 \int_0^{\infty} du \exp[-\omega_{\mathbf{k}} u] \\ &\quad \times \exp \left[-\frac{k^2}{2} \left(\frac{W^2 \hbar}{V^2 m_e} u + \frac{4C\hbar}{WV^3 m_e^2} [1 - e^{-Vu}] \right) \right]. \end{aligned} \quad (\text{B21})$$

Collecting Eqs. (B17), (B18), and (B21) in the right-hand side of Eq. (B9), we arrive at the upper bound that depends on

two dimensionless variational parameters $v = V/\omega_t$ and $w = W/\omega_t$,

$$E \leq \hbar\omega_t \frac{(v-w)^2}{2v} - \int_0^\infty du \sum_{\mathbf{k}} \frac{\pi\alpha(k)}{A k} \hbar\omega_{l,k} \sqrt{\frac{2\hbar\omega_{l,k}}{m_e}} e^{-\omega_{l,k}u} \times \exp \left[-\frac{\hbar k^2}{2m_e\omega_t} \left(\frac{w^2}{v^2} u\omega_t + \frac{v^2 - w^2}{v^3} [1 - e^{-v u\omega_t}] \right) \right]. \quad (\text{B22})$$

After replacing the sum over momenta by integral, and a q -dependent rescaling of the time variable $u \rightarrow \tau \omega_{l,q}$, we arrive at the following final result for the variational upper bound:

$$E \leq \hbar\omega_t \frac{(v-w)^2}{2v} - \hbar\omega_t \alpha_m \mathcal{I}_F(\sigma_0, \sigma_t; v, w), \quad (\text{B23})$$

where we introduced the dimensionless function

$$\mathcal{I}_F(\sigma_0, \sigma_t; v, w) = \int_0^\infty d\tau \int_0^\infty dp \frac{\sigma_t(\sqrt{\sigma_0} + 1)^2 p e^{-\tau}}{(1+p)(1+\sigma_0 p)} \times \exp \left\{ -\frac{\sigma_t^2 p^2}{\Omega_p} \left[\frac{w^2}{v^2} \tau + \Omega_p \frac{v^2 - w^2}{v^3} (1 - e^{-v\tau/\Omega_p}) \right] \right\}. \quad (\text{B24})$$

The last two equations provide the required generalization of the Feynman's variational theory of Fröhlich polarons to the case of 2D polar materials. Equations (B23) and (B24) above coincide, respectively, with Eqs. (71) and (72).

3. Renormalization of the polaron mass

In order to obtain the dynamical mass of the polaron, we follow the method introduced in Ref. [9]. We set the whole electron-phonon complex in motion at a small constant velocity $\mathbf{U}$ and determine the quadratic increase in energy with U . A rigid drift is implemented by shifting the electron path according to $\mathbf{r}_e(u) \rightarrow \mathbf{r}_e(u) + \mathbf{U}u$. Under this transformation, the Euclidean action S_{eff} is modified in three ways: (1) the electron kinetic term acquires the classical shift $m_e U^2/2$; (2) every vertex factor gets an extra phase $\exp[i\mathbf{k} \cdot \mathbf{U}(u - u')]$; and (3) the same phase appears in the trial action S_0 , so that all Gaussian averages can still be performed analytically.

With the drift included, the form factor introduced above in Eq. (B12)

$$I_U(\mathbf{k}; u, u') = I(\mathbf{k}; u, u') \exp[i\mathbf{k} \cdot \mathbf{U}(u - u')], \quad (\text{B25})$$

where $I(\mathbf{k}; u, v)$ is given by Eq. (B14). Expanding the additional phase to the second order in $\mathbf{U}$ and noticing that the linear term vanishes after angular integration over $\mathbf{k}$, one finds that only the U^2 contribution appears in the variational bound. Re-evaluating $\langle S_{\text{eff}} - S_0 \rangle_0$ with the modified I_U yields three pieces: the C -term stemming from the trial action, the ground-state energy E_0 of the trial model, and the genuine

electron-phonon part that contains the explicit coupling $|V_k|^2$. The contributions that originate solely from the trial action are identical to those in Ref. [9]:

$$\begin{aligned} \frac{C}{2} \int_0^{\beta\hbar} \int_0^{\beta\hbar} du du' e^{-W|u-u'|} (-\nabla_{\mathbf{k}}^2 I_U(\mathbf{k}; u, u'))|_{\mathbf{k}=0} \\ = \frac{2C\beta\hbar^2}{m_e W V} + \frac{C U^2}{2} \int_0^{\beta\hbar} \int_0^{\beta\hbar} du du' (u - u')^2 e^{-W|u-u'|} \\ = \frac{2C\beta\hbar^2}{m_e W V} + \frac{2C\beta\hbar U^2}{W^3}, \quad (\beta \rightarrow \infty), \end{aligned} \quad (\text{B26})$$

so that $\partial E_0/\partial C = 2\hbar/(m_e W V) + 2U^2/W^3$ and, after integrating with the condition $E_0(C=0) = \frac{1}{2} m_e U^2$, one obtains

$$E_0 = \hbar(V - W) + \frac{U^2}{2} \left(m_e + \frac{4C}{W^3} \right). \quad (\text{B27})$$

This is the only contribution that depends on the phonon dispersion and the electron-phonon coupling

$$\begin{aligned} \mathcal{F}_U = \beta \sum_{\mathbf{k}} |V_k|^2 \int_0^\infty du e^{-\omega_{l,k}u} \\ \times I(\mathbf{k}; u, 0) \left(1 - \frac{(\mathbf{k} \cdot \mathbf{U})^2 u^2}{2} \right) \\ = \mathcal{F} - \beta\hbar \frac{\delta m U^2}{2}, \end{aligned} \quad (\text{B28})$$

where we expanded $\exp[i\mathbf{k} \cdot \mathbf{U}(t - s)]$ and taking into account carrying out the angular average over $\mathbf{k}$ kept only two first nontrivial terms. Collecting all contributions, we obtain

$$E(U) \leq \frac{m_e^* U^2}{2} + \hbar\omega_t \frac{(v-w)^2}{2v} - \frac{1}{\beta\hbar} \mathcal{F}. \quad (\text{B29})$$

The electron mass, renormalized by polaron effects, can be read out of the first term,

$$m_e^* = m_e + \delta m, \quad (\text{B30})$$

where the mass correction is

$$\delta m(v, w) = \alpha_m m_e \mathcal{M}_F(\sigma_0, \sigma_t; v, w), \quad (\text{B31})$$

with

$$\begin{aligned} \mathcal{M}_F(\sigma_0, \sigma_t; v, w) \\ = \int_0^\infty d\tau \int_0^\infty dp \frac{\sigma_t^3(\sqrt{\sigma_0} + 1)^2 p^3 \tau^2 e^{-\tau}}{(1 + \sigma_0 p)^2} \\ \times \exp \left\{ -\frac{\sigma_t^2 p^2}{\Omega_p} \left[\frac{w^2}{v^2} \tau + \Omega_p \frac{v^2 - w^2}{v^3} (1 - e^{-v\tau/\Omega_p}) \right] \right\}. \end{aligned} \quad (\text{B32})$$

Upon identification $\frac{1}{\beta\hbar} \mathcal{F} = \hbar\omega_t \alpha_m \mathcal{I}_F$, Eq. (B29) gives Eq. (75), while Eq. (B32) coincides with Eq. (77). Evaluation of δm in Eq. (B31) at the optimal parameters (v^*, w^*) minimizing the upper bound of Eq. (B23) determines the polaron shift of the electron mass.

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