

# Nonperturbative Self-Consistent Electron-Phonon Spectral Functions and Transport

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Electron-phonon coupling often dominates the electron spectral functions and carrier transport properties. However, studies of this effect in real materials have largely relied on perturbative one-shot methods due to the lack of a first-principles theoretical and computational framework. Here, we present a self-consistent theory and implementation for the nonperturbative calculations of spectral functions and conductivity due to electron-phonon coupling. Applying this method to monolayer InSe, we demonstrate that self-consistency qualitatively affects the spectral function and transport properties compared to state-of-the-art one-shot calculations and allow one to reconcile calculations with angle-resolved photoemission experiments. The developed method can be widely applied to materials with dominant electron-phonon coupling at moderate computational cost.

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Spectral functions are essential in many-body physics, linking theory and experiment. Experimentally, spectral functions can be extracted using high-resolution angle-resolved photoemission spectroscopy (ARPES) [1]. Theoretically, spectral functions can be calculated from Green's functions, including many-body correlation via the self-energy. The coupling between electrons and phonons, the collective vibration of the lattice, plays a crucial and often dominant role in shaping the electron spectral function [2]. The electron-phonon (*e-ph*) interaction leads to the broadening and renormalization of the quasiparticle peak [3–7], photoemission kinks in metals [8,9], and additional structures such as satellite peaks [10–15]. Understanding and predicting these effects is the foundation of electron-phonon physics.

*Ab initio* calculation of the *e-ph* self-energy and spectral functions is usually limited to perturbation theory. Standard approaches use the perturbative Fan-Migdal (FM) self-energy [16,17], solving the Dyson equation [10] or applying the cumulant expansion [15,18–26] for spectral functions. However, these approaches often fail, causing divergent quasiparticle dispersion [26–28] or unphysical spectral properties [26]. Notably, this problem is not limited to strong *e-ph* coupling: the negative curvature in the quasiparticle dispersion of the Fröhlich model [27] and the divergent linewidth of piezoelectric materials [29] illustrate failures at all coupling strengths. Advanced methods incorporating higher-order *e-ph* interactions [30–44] are limited to model Hamiltonians. Nonperturbative methods

using molecular dynamics [45,46] or special-displacement supercells [47,48] are difficult to converge [46,49], and cannot capture nonadiabatic *e-ph* coupling, crucial in polar materials [28,50].

The electron spectral function also governs charge transport. Within the Green-Kubo theory [51,52], neglecting current vertex corrections allows direct computation of conductivity from spectral functions [53,54]. This approach, known as the bubble approximation, has the advantage of including broadening and satellites beyond the quasiparticle approximation [24,46,55–57].

A material of particular interest for nonperturbative *e-ph* coupling is monolayer InSe. While known for high electron mobility [58,59], its valence band features a flat, Mexican-hat-like dispersion [60–66], leading to strong *e-ph* interaction. In particular, state-of-the-art one-shot calculations predicted a dynamical splitting of the valence band into two peaks in the spectral function with a gap larger than 150 meV [67]. The hole mobility of monolayer InSe has been studied using the Boltzmann transport equation (BTE) [68–70] and the bubble approximation with the one-shot spectral function [67]. Yet, given the strong renormalization, nonperturbative corrections are expected to be essential in monolayer InSe.

In this Letter, we present a nonperturbative *ab initio* calculation of the *e-ph* spectral function and conductivity. Our approach, the self-consistent  $GD_0$  (sc $GD_0$ ) method [36–44], extends the FM self-energy to include the self-consistently renormalized spectral functions, analogously to the sc $GW_0$  method for electron-electron interactions [71]. Using self-consistent spectral functions, we compute conductivity using the bubble approximation. We find that self-consistency significantly affects the spectral function

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and conductivity of monolayer InSe. This effect is present in any materials and can be explored with the  $\text{scGD}_0$  method.

The most common approximation for the  $e$ -ph self-energy is the FM formula [2]

$$\Sigma_{n\mathbf{k}}^{G_0D_0\text{-FM}}(\varepsilon) = \sum_{\nu m} \int \frac{d\mathbf{q}}{V_{\text{BZ}}} |g_{m\nu}(\mathbf{k}, \mathbf{q})|^2 \times \sum_{\pm} \frac{f^{\pm}(\varepsilon_{m\mathbf{k}+\mathbf{q}}) + n_{\mathbf{q}\nu}}{\varepsilon - \varepsilon_{m\mathbf{k}+\mathbf{q}} \pm \omega_{\mathbf{q}\nu} + i\eta}, \quad (1)$$

where  $V_{\text{BZ}}$  is the Brillouin zone volume,  $g_{m\nu}(\mathbf{k}, \mathbf{q})$  the  $e$ -ph matrix element,  $\varepsilon_{n\mathbf{k}}$  and  $\omega_{\mathbf{q}\nu}$  the electron and phonon energies,  $\eta$  is a positive infinitesimal broadening,  $f^+(\varepsilon)$  the Fermi-Dirac distribution,  $f^-(\varepsilon) = 1 - f^+(\varepsilon)$ , and  $n_{\mathbf{q}\nu}$  the Bose-Einstein distribution. We neglect the band off-diagonal self-energy, which is a valid approximation if the bands do not hybridize [72], as in InSe. We refer to Eq. (1) as the  $G_0D_0$ -FM self-energy, corresponding to a one-loop diagram [see Fig. S1(a) of Supplemental Material (SM) [73]] with bare electron and phonon Green's functions,  $G_0$  and  $D_0$ , as in the  $G_0W_0$  approximation [93]. In this Letter, we assume that  $G_0$  is obtained from Kohn-Sham density functional theory and that  $D_0$  and  $e$ -ph coupling come from adiabatic density functional perturbation theory [2,94,95]. We neglect the renormalization of the phonon Green's function since we focus on the intrinsic and low doping regime where the phonon renormalization is small [2]. The total  $G_0D_0$  self-energy also contains the Debye-Waller (DW) term [3–5],

$$\Sigma_{n\mathbf{k}}^{G_0D_0}(\varepsilon) = \Sigma_{n\mathbf{k}}^{G_0D_0\text{-FM}}(\varepsilon) + \Sigma_{n\mathbf{k}}^{\text{DW}}. \quad (2)$$

In this Letter, we compute the DW term within the rigid-ion approximation [96].

Using the  $G_0D_0$  self-energy, the renormalized electron dispersion can be computed within RS perturbation theory [2,23,28,97,98] as

$$E_{n\mathbf{k}}^{\text{RS}} = \varepsilon_{n\mathbf{k}} + \Sigma_{n\mathbf{k}}^{G_0D_0}(\varepsilon_{n\mathbf{k}}). \quad (3)$$

The energy-dependent spectral function is computed by solving the Dyson equation

$$A_{n\mathbf{k}}^{G_0D_0}(\varepsilon) = -\frac{1}{\pi} \text{Im} G_{n\mathbf{k}}^{G_0D_0}(\varepsilon), \quad (4)$$

$$G_{n\mathbf{k}}^{G_0D_0}(\varepsilon) = \frac{1}{\varepsilon - \varepsilon_{n\mathbf{k}} + \mu - \Sigma_{n\mathbf{k}}^{G_0D_0}(\varepsilon)}, \quad (5)$$

with  $\mu$  the chemical potential.

To capture higher-order  $e$ -ph coupling, one often uses the cumulant approximation. It replaces the Dyson equation with the expansion

$$G_{n\mathbf{k}}^{\text{Cum}}(\varepsilon) = -i \int dt e^{i\varepsilon t} \Theta(t) e^{-i\varepsilon_{n\mathbf{k}} t} e^{C_{n\mathbf{k}}(t)}, \quad (6)$$

where  $\Theta(t)$  is the Heaviside function and  $C_{n\mathbf{k}}(t)$  the cumulant function. We adopt the retarded cumulant method [24,99,100] as we deal with finite temperatures. The cumulant function is approximated via second-order perturbation theory [21,99,100],

$$C_{n\mathbf{k}}(t) \approx \frac{1}{\pi} \int d\varepsilon |\text{Im} \Sigma_{n\mathbf{k}}^{G_0D_0}(\varepsilon + \varepsilon_{n\mathbf{k}})| \frac{e^{-i\varepsilon t} + i\varepsilon t - 1}{\varepsilon^2}. \quad (7)$$

Unlike the  $G_0D_0$  approximation, which only produces a single satellite [15], the cumulant approach can generate multiple satellite peaks, as seen in experiments [11–14]. However, it has been shown that the cumulant approximation can lead to unphysically large renormalizations for  $\mathbf{k}$  points away from the band edge [26,44], similar to the RS approximation [27].

To incorporate nonperturbative  $e$ -ph effects, we adopt the  $\text{scGD}_0$  method, where the self-energy is obtained by replacing the bare electron Green's function in the  $G_0D_0$  approximation with the dressed one [see Fig. S1(b) in SM [73]]. The imaginary part of the  $\text{scGD}_0$  self-energy is given by [56]

$$\begin{aligned} \text{Im} \Sigma_{n\mathbf{k}}^{\text{scGD}_0}(\varepsilon) &= -\pi \int \frac{d\mathbf{q}}{V_{\text{BZ}}} \sum_{m\nu} |g_{m\nu}(\mathbf{k}, \mathbf{q})|^2 \sum_{\pm} [f^{\pm}(\varepsilon \pm \omega_{\mathbf{q}\nu}) + n_{\mathbf{q}\nu}] \\ &\times A_{m\mathbf{k}+\mathbf{q}}^{\text{scGD}_0}(\varepsilon \pm \omega_{\mathbf{q}\nu}). \end{aligned} \quad (8)$$

The real part is obtained using the Kramers-Kronig relation, with the addition of the real-valued DW term,

$$\text{Re} \Sigma_{n\mathbf{k}}^{\text{scGD}_0}(\varepsilon) = \frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} d\varepsilon' \frac{\text{Im} \Sigma_{n\mathbf{k}}^{\text{scGD}_0}(\varepsilon')}{\varepsilon' - \varepsilon} + \Sigma_{n\mathbf{k}}^{\text{DW}}, \quad (9)$$

where  $\mathcal{P}$  denotes the principal value. The spectral function in Eq. (8) is obtained via the Dyson equation

$$A_{n\mathbf{k}}^{\text{scGD}_0}(\varepsilon) = -\frac{1}{\pi} \text{Im} G_{n\mathbf{k}}^{\text{scGD}_0}(\varepsilon), \quad (10)$$

$$G_{n\mathbf{k}}^{\text{scGD}_0}(\varepsilon) = \frac{1}{\varepsilon - \varepsilon_{n\mathbf{k}} + \mu - \Sigma_{n\mathbf{k}}^{\text{scGD}_0}(\varepsilon)}. \quad (11)$$

Solving Eqs. (8)–(11) self-consistently yields the  $\text{scGD}_0$  self-energy and spectral function. We also iteratively adjust the temperature-dependent chemical potential to match the total electron occupation.

We remark that it is advantageous to first compute the imaginary part and then use the Kramers-Kronig relation to obtain the real part. The relation between the real parts of  $G$  and  $\Sigma$  involves a  $\mathbf{q}$ -dependent energy integral [56].

Separating Eqs. (8) and (9) enables performing the  $\mathbf{q}$  integration for the imaginary part first, followed by a  $\mathbf{q}$ -independent energy integral.

In practical *ab initio* calculations, several hundred bands per atom are needed to converge the real part of the FM self-energy [49]. Calculating this via the Kramers-Kronig transformation is impractical, as it demands evaluating  $\text{Im} \Sigma(\varepsilon)$  up to  $\varepsilon \sim 300$  eV. We simplify the calculation by treating the contribution of high-energy bands as a static correction. We thus self-consistently compute  $\text{Im} \Sigma_{nk}^{\text{scGD}_0}(\varepsilon)$  only for the states inside the active space window  $[\varepsilon^{\min}, \varepsilon^{\max}]$ , and add the static correction as

$$\begin{aligned} \text{Re} \Sigma_{nk}^{\text{scGD}_0}(\varepsilon) = & \frac{1}{\pi} \mathcal{P} \int_{\varepsilon^{\min}}^{\varepsilon^{\max}} d\varepsilon' \frac{\text{Im} \Sigma_{nk}^{\text{scGD}_0}(\varepsilon')}{\varepsilon' - \varepsilon} \\ & + \text{Re} \Sigma_{nk}^{G_0D_0\text{-FM-rest}} + \text{Re} \Sigma_{nk}^{\text{DW}}. \end{aligned} \quad (12)$$

Here,  $\text{Re} \Sigma_{nk}^{G_0D_0\text{-FM-rest}}$  is the one-shot FM self-energy [Eq. (1)] from states outside the active space,  $\varepsilon_{m\mathbf{k}+\mathbf{q}} \notin [\varepsilon^{\min}, \varepsilon^{\max}]$ , evaluated at the bare eigenvalue  $\varepsilon_{n\mathbf{k}}$ . This term can be efficiently computed by solving the linear Sternheimer equation [97,98] and interpolating using Wannier function perturbation theory [101]. The static corrections in the second line of Eq. (12) are always included during the self-consistent calculation of the  $\text{scGD}_0$  self-energy, i.e., Eq. (12) replaces Eq. (9). (See Sec. S3 of SM [73] for additional details.)

In practice, we initialize the  $\text{scGD}_0$  self-energy as a constant  $-i\eta$  with  $\eta = 5$  meV. We do not use any artificial broadening during the  $\text{scGD}_0$  iteration, and the choice of  $\eta$  does not affect the converged  $\text{scGD}_0$  self-energy. We iterate until the maximum change of  $\Sigma_{nk}^{\text{scGD}_0}(\varepsilon)$  is below 0.1 meV. The  $\text{scGD}_0$  method is implemented as an extension of our ElectronPhonon.jl package [102], which uses quantities

computed from Quantum ESPRESSO [103], Wannier90 [104], and EPW [105,106]. Computational details are provided in Sec. S4 of SM [73].

The  $\text{scGD}_0$  method, also known as the self-consistent Migdal, self-consistent first-Born, or the noncrossing approximation, has been applied to the Holstein [36–38,40–44,107,108] and Anderson-Holstein models [39]. This nonperturbative method sums the infinite series of noncrossing rainbow diagrams [see Fig. S1(c) of SM [73]], including those with an arbitrary number of  $e$ -ph vertices. It is expected to be valid from weak to intermediate coupling but will fail in the strong coupling regime, as in the case of self-trapped small polarons [109]. State-of-the-art *ab initio* calculations [56,57] used a one-shot calculation of Eqs. (8) and (9) (without  $\Sigma_{nk}^{\text{DW}}$ ) with the spectral function obtained from dynamical mean-field theory to compute the  $e$ -ph self-energy. Here, we self-consistently solve Eqs. (8)–(12) to include nonperturbative  $e$ -ph effects on the spectral function.

We apply the  $\text{scGD}_0$  method to monolayer InSe, which has a Mexican-hat-like valance band [60–62,64]. This dispersion leads to strong  $e$ -ph effects in hole-doped InSe monolayers: the one-shot  $G_0D_0$  spectral functions show a dynamic splitting of 150 meV or larger [67]. We study whether this large renormalization persists under a self-consistent update of the spectral function.

Figures 1(a)–1(d) show the valence band spectral functions of monolayer InSe at 100 K, compared to the ARPES photointensity [110] measured at the same temperature. The RS dispersion has several kinks with inverted curvature near the valence band maximum (VBM), an artifact also observed for the Fröhlich model [27]. The cumulant spectral function shows a single broad peak with no satellites and a prominent dip at  $\Gamma$ . The one-shot  $G_0D_0$  method yields two dynamically split bands with a splitting of 170 meV, consistent with Ref. [67]. However, this large

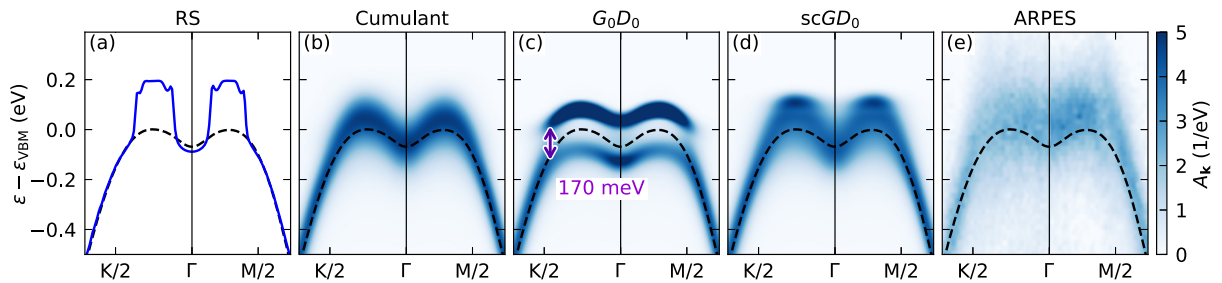


FIG. 1. (a)–(d) Renormalized dispersion and spectral functions of monolayer InSe at  $T = 100$  K computed using the Rayleigh-Schrödinger (RS) perturbation theory [Eq. (3)], cumulant approximation [Eqs. (6) and (7)], one-shot  $G_0D_0$  [Eqs. (4) and (5)], and  $\text{scGD}_0$  [Eqs. (8)–(12)] methods. The dashed black curves represent the bare electron band. Results in (a)–(c) are computed with an artificial broadening of  $\eta = 5$  meV but the results are qualitatively the same with larger values; see Sec. S5 of SM [73]. The  $\text{scGD}_0$  result in (d) does not use any artificial broadening. To account for the experimental resolution, we apply a Gaussian filter with widths of  $0.03 \text{ \AA}^{-1}$  and 20 meV [110] on the calculated spectral functions in (b)–(d). This filter does not qualitatively impact the results, as shown in Fig. S7 of SM [73] for the results without the filter. (e) Experimental ARPES photointensity reported in Ref. [110], with the background value (average of the data inside the band gap) subtracted, and the data at each  $\mathbf{k}$  normalized to satisfy  $\int A_{\mathbf{k}}(\omega) d\omega = 1$ . The data is vertically shifted to align the intensity peaks with the bare bands at  $\varepsilon = -0.4$  eV ( $\varepsilon = 0$  is 2.234 eV below the experimental Fermi level).

splitting vanishes under self-consistency. The  $scGD_0$  spectral function shows a broadened quasiparticle peak with multiple satellites, with a distance between the peaks of 30–50 meV, comparable to the longitudinal optical (LO) phonon energy of 27 meV. An analysis of the difference between  $G_0D_0$  and  $scGD_0$  self-energies is provided in the End Matter (Fig. 4). Our  $scGD_0$  result is consistent with the absence of a 150 meV scale gap in the ARPES photo-intensity [64,110]. Although both the  $scGD_0$  and cumulant spectral functions are consistent with the experimental observation within the experimental resolution, higher-resolution ARPES may observe the satellites predicted by our  $scGD_0$  calculation.

Beyond ARPES predictions that can sometimes be challenging to verify experimentally, the electron spectral function can be directly used to compute carrier conductivity, a simpler quantity to measure. Neglecting the current vertex corrections, we calculate the conductivity in the bubble approximation, which is given by [53,54]

$$\begin{aligned} \text{Re}\sigma_{\alpha\alpha}^{\text{Bubble}}(\Omega) &= \frac{\pi}{V_{\text{uc}}} \sum_{mn} \int \frac{d\mathbf{k}}{V_{\text{BZ}}} |v_{m\mathbf{k}}^\alpha|^2 \\ &\times \int d\epsilon \frac{f^+(\epsilon) - f^+(\epsilon + \Omega)}{\Omega} \\ &\times A_{m\mathbf{k}}(\epsilon) A_{n\mathbf{k}}(\epsilon + \Omega). \end{aligned} \quad (13)$$

The corresponding mobility is  $\mu_{\alpha\alpha}(\Omega) = \text{Re}\sigma_{\alpha\alpha}(\Omega)/n^c$  with  $n^c$  the carrier concentration. Equation (13) has been used to calculate the conductivity due to electron-electron interaction [54],  $e$ -ph interaction [24,46,55], or both [56,57]. Here, we investigate how the self-consistent renormalization of the spectral function affects the dc and ac mobility of monolayer InSe.

To study transport in the intrinsic limit, we set a small carrier concentration, placing the chemical potential within the band gap. In this nondegenerate regime, mobility should be independent of carrier concentration [111,112]. However, as detailed in the End Matter, we find that both the  $G_0D_0$  and cumulant spectral functions yield strongly  $n^c$ -dependent mobility (Fig. 5). We attribute this artifact to the slow  $1/\epsilon^4$  decay of the spectral function above the band edge, which induces a large number of carriers on the tail of the spectral function. To address this problem, we developed a truncation scheme that removes the tail above a cutoff energy (see Sec. S2 of SM [73]), which results in a  $n^c$ -independent  $G_0D_0$  mobility. However, this scheme cannot be applied to the cumulant spectral function of InSe due to its large broadening. The  $scGD_0$  method does not suffer from this problem and yields an  $n^c$ -independent mobility.

Figure 2(a) shows the intrinsic dc mobility of InSe. While the BTE and self-energy relaxation time approximation (SERTA) mobilities [113] are nearly identical and agree with the results from Refs. [69,70], the bubble

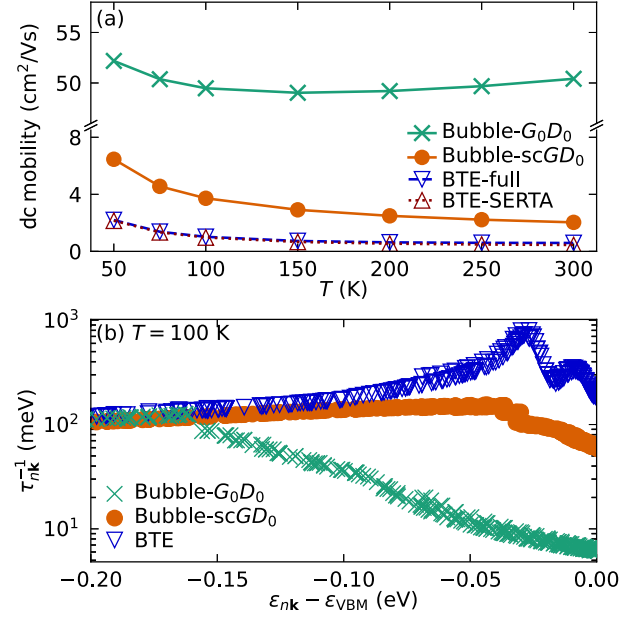


FIG. 2. (a) Intrinsic dc mobility of monolayer InSe obtained within the bubble approximation with the one-shot (Bubble- $G_0D_0$ ) or the self-consistent (Bubble- $scGD_0$ ) spectral function, the full Boltzmann transport equation (BTE-full), and the BTE with the self-energy relaxation time approximation (BTE-SERTA). (b) Effective scattering rates at  $T = 100$  K, calculated as the imaginary part of the self-energy evaluated at the bare or renormalized energy [Eqs. (14)–(16)].

mobility with the  $scGD_0$  spectral function is 3 to 4 times larger. This suggests that the quasiparticle approximation significantly underestimates the mobility. Using the one-shot  $G_0D_0$  spectral function instead of the  $scGD_0$  one leads to an overestimation by more than an order of magnitude, and the mobility strongly depends on the artificial broadening  $\eta$ , varying inversely with  $\eta$  (see Fig. S9 of SM [73]). These results can be understood in terms of the effective electron scattering rates shown in Fig. 2(b), which are inversely proportional to the mobility. The SERTA and BTE mobilities are linked to the imaginary part of the  $G_0D_0$  self-energy at the bare band energy,

$$(\tau_{nk}^{\text{BTE}})^{-1} = 2|\text{Im}\Sigma_{n\mathbf{k}}^{G_0D_0}(\epsilon_{n\mathbf{k}})|. \quad (14)$$

For the spectral functions, we define the effective scattering rates using the self-energy at the renormalized quasiparticle energy,

$$(\tau_{nk}^{\text{Bubble-}G_0D_0})^{-1} = 2|\text{Im}\Sigma_{n\mathbf{k}}^{G_0D_0}(E_{n\mathbf{k}}^{G_0D_0})|, \quad (15)$$

$$(\tau_{nk}^{\text{Bubble-}scGD_0})^{-1} = 2|\text{Im}\Sigma_{n\mathbf{k}}^{scGD_0}(E_{n\mathbf{k}}^{scGD_0})|. \quad (16)$$

The renormalized quasiparticle energies,  $E_{n\mathbf{k}}^{G_0D_0}$  and  $E_{n\mathbf{k}}^{scGD_0}$ , are the energies where the spectral functions  $A_{n\mathbf{k}}^{G_0D_0}(\epsilon)$  and  $A_{n\mathbf{k}}^{scGD_0}(\epsilon)$  are maximal. Near the VBM, the BTE scattering

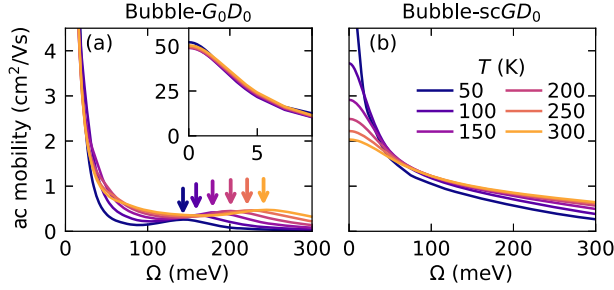


FIG. 3. Intrinsic ac mobility of monolayer InSe computed using the bubble approximation [Eq. (13)] with the (a)  $G_0D_0$  and (b)  $scGD_0$  spectral functions. The arrows indicate peaks at finite frequency. The inset of panel (a) is an enlargement of the low-frequency Drude peak.

rates exhibit a peak, while the  $scGD_0$  result shows a smoother variation and smaller values due to the self-consistent broadening. For the one-shot self-energy, the scattering rates near the VBM are tiny because the renormalized valence bands lie within the bare band gap where the imaginary part of the  $G_0D_0$  self-energy is small. This behavior is an artifact of the one-shot approximation.

Finally, we show in Fig. 3 the ac mobility calculated using the  $G_0D_0$  and  $scGD_0$  spectral functions. For the one-shot spectral function, we observe a peak around 150–250 meV, corresponding to the gap shown in Fig. 1(c), as in Ref. [67]. This feature persists with a larger artificial broadening (see Fig. S10 of SM [73]). However, with the self-consistent spectral function, the unphysical peaks are smoothed out due to the reduced splitting and the broader Drude peak.

The self-consistent framework developed in this Letter offers a versatile tool for analyzing the nonperturbative effect of  $e$ -ph coupling with moderate computational effort, bridging the gap between one-shot approximations and numerically exact methods. While we have focused on the spectral functions and conductivity, this framework can be extended to other properties such as superconductivity and indirect absorption [114,115]. Our Letter also lays the groundwork for applying diagrammatic many-body approaches to  $e$ -ph systems. The  $scGD_0$  method could be extended to explore contributions beyond the Fan-Migdal diagram. Notably, the development of a fully self-consistent  $GD$  scheme using the renormalized phonon Green's function  $D$  [36,37,116], the inclusion of the dynamical screening [117–119], and the consideration of  $e$ -ph vertex correction [40,42,120] is a promising avenue for future work. Combining the  $scGD_0$  formalism with dynamical mean-field theory [121,122] and its diagrammatic extensions [123] to study transport from the interplay of electron-electron [54] and  $e$ -ph coupling [124] beyond the one-shot approximation [56,57] is another interesting direction for future research.

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**Data availability**—The data that support the findings of this Letter are openly available [125].

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## End Matter

*Appendix A: Comparison of the self-energies*—Figure 4 compares the  $G_0D_0$  and  $scGD_0$  self-energies at 100 K and 300 K. The one-shot  $G_0D_0$  self-energy shows a sharp peak in the imaginary part at  $\varepsilon = \varepsilon_{\text{VBM}} - \omega_{\text{LO}}$ , where  $\omega_{\text{LO}} \sim 27$  meV is the LO phonon frequency. This peak, associated with phonon emission, is significantly damped and smeared in the  $scGD_0$  self-energy due to the self-consistent broadening of the spectral function. This self-consistent regularization becomes more pronounced at higher temperatures.

At 300 K, the  $scGD_0$  self-energy reaches around 0.1 eV. While this value is smaller than the full bandwidth of the valence band of InSe (1.2 eV; see Fig. S4 of SM [73]), it is larger than the width of the Mexican-hat feature at the

valence band edge (0.07 eV). Thus, the self-energy leads to a significant renormalization near the valence band edge, qualitatively altering the spectral function.

*Appendix B: Tail of the spectral functions*—Compared to the  $G_0D_0$  and cumulant spectral functions, the  $scGD_0$  spectral functions show a clear qualitative difference in insulators and nondegenerate semiconductors: the former exhibits an unphysically long tail in the midgap region, which is absent in the latter. Figures 5(a) and 5(b) show the density of states (DOS),

$$\text{DOS}(\varepsilon) = \sum_{n\mathbf{k}} A_{n\mathbf{k}}(\varepsilon), \quad (\text{B1})$$

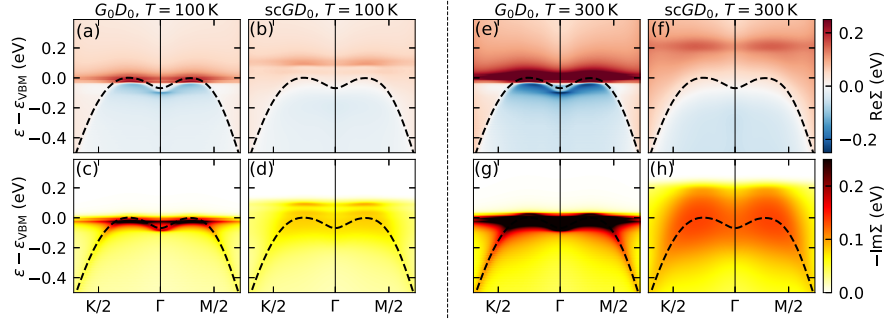


FIG. 4. (a),(b) Real and (c),(d) imaginary parts of the  $G_0D_0$  and  $scGD_0$  self-energies of monolayer InSe at  $T = 100$  K. (e)–(h) Same for  $T = 300$  K. The dashed black curves represent the bare electron band.

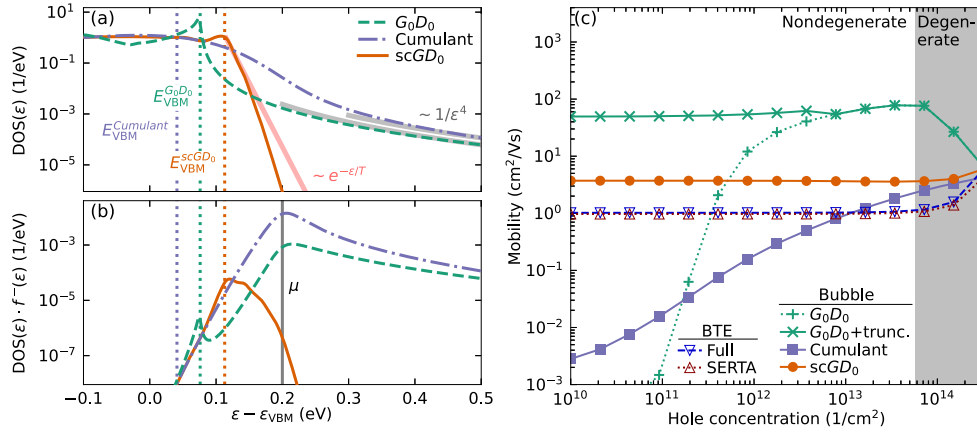


FIG. 5. (a) Density of states (DOS) and (b) hole DOS of monolayer InSe at  $T = 100$  K with a fixed chemical potential of 0.2 eV. The vertical dotted lines indicate the renormalized band edge, defined as the peak of the spectral function at the valence band maximum (VBM). The shaded curves in (a) show the power law and exponential decay derived in Sec. S2 of SM [73]. Both  $G_0D_0$  and cumulant spectral functions exhibit a slow  $1/\epsilon^4$  decay in the midgap region, leading to an unphysical peak at  $\mu$  in the hole DOS. In contrast, the  $scGD_0$  spectral function decays faster than  $e^{-\epsilon/T}$ , with the hole DOS peaking at the renormalized band edge as expected. (c) Doping dependence of the hole mobility of monolayer InSe at  $T = 100$  K. The white (gray) background indicates the nondegenerate (degenerate) semiconductor regime, where the BTE chemical potential is above (below) the bare VBM. Significant doping dependence is observed for the  $G_0D_0$  (green plus markers) and cumulant (blue squares) mobilities, while the mobility is doping-independent in the nondegenerate regime for the  $scGD_0$  (orange circles) and BTE results (blue and red triangles).

and the hole DOS [ $f^-(\epsilon)\text{DOS}(\epsilon)$ ]. In Fig. 5(a), the DOS from the  $G_0D_0$  and the cumulant methods shows a slow  $1/\epsilon^4$  decay (gray shaded curves). This is a result of the Lorentzian broadening of the bare Green's function (see Sec. S2 of SM [73] for a detailed derivation.) Importantly, reducing the artificial broadening  $\eta$  reduces the amplitude of the tail but does not change the  $1/\epsilon^4$  scaling. When multiplied by the hole occupation factor, which increases exponentially with energy up to the chemical potential, the hole DOS continues to increase above the renormalized band edge, peaking at the chemical potential [Fig. 5(b)]. As a result, in nondegenerate semiconductors, the  $G_0D_0$  and cumulant methods incorrectly predict an unphysically large amount of carriers in the midgap region, where the spectral function is small. In contrast, the  $scGD_0$  spectral function decays faster than  $e^{-\epsilon/T}$  (red shaded

curve), and the hole DOS peaks at the renormalized band edge as expected.

*Appendix C: Doping dependence of the conductivity—*Figure 5(c) shows the doping dependence of the hole mobility of monolayer InSe at 100 K. Surprisingly, the  $G_0D_0$  (green plus markers) and cumulant mobility (blue squares) show significant changes as a function of hole concentration. This result contrasts sharply with what is typically observed in nondegenerate semiconductors with dominant  $e$ -ph scattering [111,112]. We attribute this artifact to the unphysical tail in the spectral functions, which places most of the holes near the chemical potential, where the actual spectral function is small. Because of this strong doping dependence, the intrinsic (low doping limit) mobility cannot be properly calculated using the  $G_0D_0$  and cumulant approximations.

To alleviate this problem, we developed a truncation scheme [see Eq. (S19) of SM [73]] that removes the tail of the spectral function beyond the local minima in the hole DOS. Then, we obtain a doping-independent Bubble- $G_0D_0$  mobility (green crosses). However, this truncation could not be applied to the cumulant spectral functions of InSe due to its large broadening.

In contrast, the  $scGD_0$  mobility (orange circles) as well as the BTE and SERTA mobility (blue and red

triangles) remain doping-independent in the nondegenerate regime (white background). Doping dependence is observed only in the metallic regime (gray background), where the chemical potential lies within the valence band. Therefore, it is highly desirable to use the  $scGD_0$  method, rather than the  $G_0D_0$  or cumulant approximations, for studying transport in nondegenerate semiconductors.