Going beyond the Cumulant Approximation: Power Series Correction to the Single-Particle Green's Function in the Holstein System

Bipul Pandey[*] and Peter B. Littlewood
James Franck Institute and Department of Physics, University of Chicago, Chicago, Illinois 60637, USA

(Received 22 January 2022; accepted 5 September 2022; published 21 September 2022)

In the context of a single-electron two orbital Holstein system coupled to dispersionless bosons, we develop a general method to correct the single-particle Green's function using a power series correction (PSC) scheme. We outline the derivations of various flavors of cumulant approximation through the PSC scheme explaining the assumptions and approximations behind them. Finally, we compare the PSC spectral function with cumulant and exact diagonalized spectral functions and elucidate three regimes of this problem—two where the cumulant explains and one where the cumulant fails. We find that the exact and the PSC spectral functions match within spectral broadening across all three regimes.

DOI: 10.1103/PhysRevLett.129.136401

Overview.—Electrons and holes in materials undergo numerous complex interactions among themselves and the external fields, as well as the constituent atomic lattice. The strength of such many-body interactions depends on various factors, like electronic configuration of the host material, presence of doping, defects, lattice vibrations, etc. Such factors manifest as bosonic collective excitations that renormalize the particle states (electrons and holes) into quasiparticle states with different energy and lifetime, and even mix quasiparticle states depending on the interaction strength. Alongside the quasiparticle features in photoemission spectra, these collective excitations show up as "shakeoff" features that can be loosely separated into sharp satellites emerging from bosonic collective modes (such as plasmons and optical phonons) and continua arising from nonzero-momentum particle-hole excitations (including excitons) [1–3].

In experiments, the interaction strengths between collective excitations and particles are achieved by modulating doping and defects [4,5]. In calculations, this interaction is modeled as tunable electron-boson coupling parameters. Although at very weak coupling the quasiparticle renormalization due to the collective modes is negligible, with stronger coupling a proportional renormalization of the quasiparticle occurs. As an example, in photoemission spectra of strontium titanate, this coupling manifests as a significant shift in quasiparticle energy, significant decrease in lifetime and intensity of quasiparticle features, and strong shakeoff features, as well as a strong carrier mass enhancement [6–10]. Strong electron-phonon coupling is also visible in electronic spectra in metallic cuprates [11,12], the metal-insulator transition in undoped cuprates [13], and other correlated metals, for example, FeSe/$SrTiO_3$ epitaxial layers [14].

At extreme values of the coupling constant, strong electron-boson coupling can completely self-trap and localize electrons creating polaronic states. This severely modifies carrier mobility in the material and is of particular interest in material design for photovoltaics and electronics [15–17]. Finally, in the presence of multiple boson species, competition between their effect on the carrier can create novel phase crossovers in materials [18]. A proper understanding and quantification of the effects of collective modes on charge carriers is vital in understanding and designing novel material with useful engineering applications.

The "*GW*" approximation [19], although expensive, is at the heart of understanding internal energy landscape as well as transport properties of materials. Computing the single-particle Green's function and charged excitation spectra from *GW* approximation is also a fundamental first step in understanding optical properties of material using the Bethe-Salpeter equation (BSE) [20], which is even more expensive. Inventing accurate approximations while taming computational cost is an important active research topic for both *GW* and BSE [21–27]. Therefore, a relatively inexpensive method to improve charged excitation spectra computed from simpler *GW* approximations in postprocessing is of great value for both material study and more sophisticated method development. In this Letter, we construct a method that systematically generalizes existing cumulant-expansion-based improvements to the *GW* spectra [28] to describe single-particle dynamics of a system with multiple electronic levels interacting through common boson baths. Our Letter is a stepping stone toward incorporating all four kinds of boson mediated losses shown in Fig. 1 in the charged excitation (*GW*) spectra of materials. Our inexpensive and simple method can improve the *GW* spectra by incorporating the self-correction along with interorbital transition or interband vertical transition, depending on the nature of problem (orbitals vs flat bands). Through our formalism, we also inexpensively solve for the photoemission spectra of a simple two-level Holstein system, elucidate its various regimes as we tune

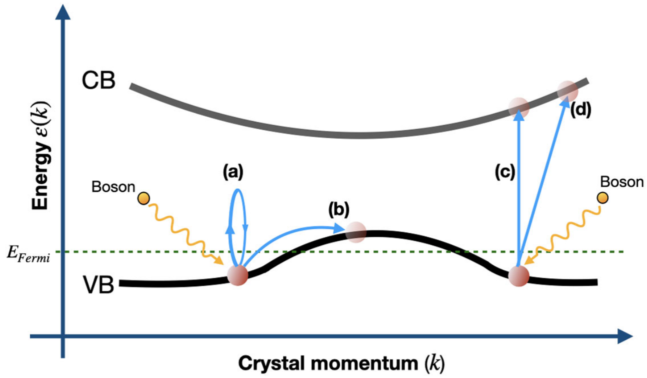

FIG. 1. Schematic of different kinds of boson mediated losses that create shakeoffs in excitation spectra of material. (a) Self transitions, (b) interorbital transition, (c) interband vertical transition, and (d) interband nonvertical transition between conduction and valence band (CB and VB).

the boson energy, and outline where and how cumulant-based methods lose their validity.

The Letter is organized as follows. In Sec. II, we introduce the model problem and concepts of the electron Green's function, electron self-energy, and Dyson's equation. In Sec. III, we briefly introduce existing methods and their major drawbacks. In Sec. IV, we develop our correction scheme and physically motivate the assumptions used to simplify the equations. In Sec. V, we outline the derivation of various flavors of cumulants through our method and clarify the implicitly made but vaguely understood assumptions behind these approximations. Finally, in Sec. VI, we identify three important regimes of the problem by comparing performances of the cumulant and the power series method with results from exact diagonalization of this problem in the finite boson basis.

Introduction to the problem.—We consider a model Hamiltonian for a single-electron two orbital Holstein system with bonding and antibonding energy $\varepsilon_+/\varepsilon_-$ such that their difference is Δ. This system is kept in baths of two dispersionless boson species ($\pm$) of energy ω_o. Interaction of electron with ($+/-$) bosons causes the electron's self- and interorbital transition.

The electron-boson interaction strength is controlled by a coupling constant g. The fermionic ladder operators are $c_+/c_+^\dagger$ and $c_-/c_-^\dagger$ for bonding and antibonding orbitals, respectively. The bosonic ladder operators for ($\pm$) bosons are $b_\pm/b_\pm^\dagger$. This Hamiltonian has three distinct pieces—the noninteracting part, H_+ that has ($+$) bosons that cause self-transition, and H_- that has ($-$) bosons that cause interorbital transitions,

$$\begin{aligned} H &= \sum_{i=\pm} \varepsilon_\pm c_i^\dagger c_i + H_+ + H_-, \quad \text{where} \\ H_+ &= \omega_o b_+^\dagger b_+ + g(c_+^\dagger c_+ + c_-^\dagger c_-)(b_+^\dagger + b_+), \\ H_- &= \omega_o b_-^\dagger b_- + g(c_+^\dagger c_- + c_-^\dagger c_+)(b_-^\dagger + b_-). \end{aligned} \tag{1}$$

This Hamiltonian describes the physics of a model of the dihydrogen cation (H_2^+)—two hydrogen nuclei and a single electron. Historically, this problem was approached with clamped nuclei approximation. This crude approach completely neglects the vibronic coupling between the electron and vibrational modes of nuclei (optical phonons in crystalline structures), which becomes crucial when $\Delta \approx \omega_o$. Vibronic couplings in this regime can cause interorbital transitions and severely renormalize the molecular energy levels [29–31]. Furthermore, no exact analytical solution exists and existing approximate methods either give incorrect boson satellites (GW) or are *ad hoc* and incorrect at strong coupling (cumulant) [32]. Hence, this is a good model to build and test our approximation scheme due to its simplicity and the failures of existing methods. The retarded-time (RT) formalism is better suited to handle electron-hole problems because it treats both of them in equal footing as particles [33]. For the Holstein problem (1) with Fock vacuum $|0\rangle$ as the ground state and $\{,\}/[,]$ as the anticommutator-commutator, the electron Green's function $G(n,t)$ for each orbital [33] n and the boson Green's function $\mathcal{D}(N,t)$ for each boson species N in RT formalism is [34]

$$\begin{aligned} G(n=\pm,t) &= -i\theta(t)\langle 0|\{c_n(t), c_n^\dagger\}|0\rangle, \\ \mathcal{D}(N=\pm,t) &= -i\theta(t)\langle 0|[b_N(t), b_N^\dagger]|0\rangle. \end{aligned} \tag{2}$$

For noninteracting ($g=0$) electrons and dispersionless bosons with energy ω_o, the bare electron Green's function G_o and a bare boson Green's function $\mathcal{D}$ are

$$G_o(\pm,t) = -i\theta(t)e^{-i\varepsilon_\pm t}, \qquad \mathcal{D}(\pm,t) = -i\theta(t)e^{-i\omega_o t}. \tag{3}$$

The quasiparticle energies, lifetimes, and boson satellites show up as complex poles of $G(n,\omega)$, where ω is the frequency. The frequency axis spectral function $A(m,n;\omega)$ (see Supplemental Material [35]) is defined as

$$A(m,n;\omega) = \frac{1}{\pi}|\mathrm{Im}G(m,n;\omega)|. \tag{4}$$

At zero coupling ($g=0$), the energy eigenvalues $\varepsilon_\pm$ of (1) are real and states have infinite lifetime, owing to the lack of interaction between the orbitals. However, upon switching on the boson mediated interaction ($g \neq 0$) between orbitals, the exchange of energy and momenta between states through boson exchange causes clumping of electrons and holes to form quasiparticles. Because of time-translational invariance, we can package this interaction information together and call it the self-energy,

$$-i\Sigma(t) = g^2 \sum_{N,n=\pm} \mathcal{D}(N,t)G(n,t) = g^2 \sum_{n=\pm} -i\Sigma(n,t). \tag{5}$$

Each orbital's self-energy $\Sigma(n,t)$ is complex-valued, giving rise to spectral peak broadening—an indication of a finite quasiparticle lifetime. A proper self-energy also incorporates boson mediated transitions, produces satellite peaks at the correct energies, and redistributes the spectral weight from the quasiparticle to the satellites. Dyson's equation governs the evolution of G by repeated application of this self-energy,

$$G(n,\omega) = G_o(n,\omega) + G_o(n,\omega)\Sigma(\omega)G(n,\omega). \quad (6)$$

GW, cumulant expansion, and their drawbacks.—Unlike fully self-consistent $GW\Gamma$ [19], the simpler GW approximations sacrifice self-consistency through abrupt truncation of Dyson's equation for reasonable computing cost. Although these approximations give a reasonably good description of quasiparticle properties at weak coupling, the plasmon satellites are averaged and misplaced at some incorrect average energy [28]. At strong coupling, due to the lack of self-consistency, even the quasiparticle properties can be incorrect.

For a single band of electron in a dispersionless boson bath [28], the following exact Green's function exists:

$$G(k,t) = G_o(k,t)e^{C(k,t)}. \quad (7)$$

The cumulant C is calculated by linearizing and comparing Eqs. (7) and (6). Cumulant-generated satellites manifest as a Poisson series of peaks, plasma frequency apart in spectral function. In real systems, although not all assumptions of the above model hold true, an approximate cumulant correction can be found using the same recipe as above on a GW self-energy [36]. Recently, interest in cumulant approximation has resurged [33,37–39] enabled by increases in computational ability to perform GW and inspired by experiments (e.g., [40]).

The cumulant has the considerable merit of giving near-exact spectra for weak electron-boson coupling: $\Delta \gg \omega_o$ and/or $g \ll 1$. However, at strong coupling and in the presence of multiple electronic levels, the bosons significantly affect quasiparticle properties in ways not reflected in the cumulant approximation. The cumulant is also not systematically improvable by design and lacks proper accounting of interband scattering owing to the absence of self-consistency.

Theoretical framework.—The power series ansatz: Rather than assuming an exponential correction, we assume a power series correction $\mathcal{P}_n(t)$ in the powers of g^2 to the nth orbital's bare Green's function $G_o(n,t)$ due to interaction with bosons for time duration t. By construction, the interacting system smoothly maps to the noninteracting system as g^2 goes to zero,

$$G(n,t) = G_o(n,t)\mathcal{P}_n(t) = G_o(n,t)\sum_{X=0}^{\infty} g^{2X}C_X(n,t). \quad (8)$$

Here $C_0 = 1$ and all other C_X are distinct correction functions of different orders that are zero when $t < 0$. This makes physical sense because in the RT framework the particle does not exist for $t < 0$. This, just like the cumulant, is still a diagonal approximation [33] to the Green's function matrix because, by construction, only those corrections in which a particle eventually returns back to its initial state n are accounted for.

Temporal contraction relation: Because of the boundary value dependence on time of the Green's function, $\mathcal{P}_n(t)$ has the following temporal contraction property:

$$\mathcal{P}_n(t_f - t_i) = \mathcal{P}_n(t_f - t_o)\mathcal{P}_n(t_o - t_i); \quad t_i < t_o < t_f. \quad (9)$$

This property is central in generating cumulant diagrams and does not apply between power series pieces of different orbitals.

Assumption on electron self-energy: To properly construct the electron self-energy, rather than replacing G by G_o inside the self-energy as in GW or cumulant expansions, we replace it by power series ansatz and reintroduce self-consistency,

$$\begin{aligned} -i\Sigma(t) &= g^2 \sum_{N,n=\pm} \mathcal{D}(N,t)G_o(n,t)\mathcal{P}_n(t) \\ &= g^2 \sum_{n=\pm} -i\Sigma_o(n,t)\mathcal{P}_n(t). \end{aligned} \quad (10)$$

Here, the nth orbital's self-energy $\Sigma_o(n,t)$ is computed using G_o. The introduction of power series in Σ through G produces corrections due to the particle's eventual return to the initial state after scattering through other possible states. Including these cyclic scattering contributions in the Green's function matrix's diagonal makes the diagonal exact.

Correction scheme: We take the temporal Dyson's equation for the mth band and replace G and Σ by their power series corrected versions from (8) and (10). We then use temporal limits enforced by the RT bare Green's function (3) and simplify the equation using the temporal contraction property from Eq. (9),

$$G(m,t-t_0) = G_o(m,t-t_0) + \iint dt_1 dt_2 G_o(m,t-t_2) \times \Sigma(t_2 - t_1)G(m,t_1 - t_0).$$

Setting $t_0 = 0$ and $t_2 - t_1 = \tau$, and simplifying, we get

$$\mathcal{P}_m(t) = 1 + (-ig^2)\sum_{n=\pm}\int_0^t dt_2 \times \int_0^{t_2} d\tau\, e^{i\epsilon_m\tau}\Sigma_o(n,\tau)\mathcal{P}_n(\tau)\mathcal{P}_m(t_2-\tau).$$

The integral in this equation contains two distinct correction contributions. The self-correction (P_m^{SC}) contains interactions within same orbital ($n = m$) on the right side of this equation, which will be merged by contraction property (9). The interband scattering term (P_m^{IC}) occurs when different orbitals interact ($n \neq m$) and therefore the contraction property is invalid,

$$\therefore \mathcal{P}_m(t) = 1 + P_m^{SC} + P_m^{IC}, \quad \text{where}$$

$$P_m^{SC} = -ig^2 \int_0^t dt_2 \int_0^{t_2} d\tau\, e^{i\varepsilon_m \tau} \Sigma_o(m,\tau) \mathcal{P}_m(t_2),$$

$$P_m^{IC} = -ig^2 \int_0^t dt_2 \int_0^{t_2} d\tau\, e^{i\varepsilon_m \tau} \Sigma_o(n,\tau) \mathcal{P}_n(\tau) \mathcal{P}_m(t_2 - \tau). \tag{11}$$

For numerical solution, we start with an initial guess of $\mathcal{P} = 1$ on the right and self-consistently compute better values for $\mathcal{P}$ on the left until it converges.

Derivation of various cumulant schemes.—We validate our method by deriving the exact result for the core-hole problem with a single orbital of bare energy ε_o in a bath of dispersionless plasmons of frequency ω_o [28]. The Hamiltonian in this case is

$$H = \varepsilon_o c^\dagger c + \omega_o b^\dagger b + g(b^\dagger + b)(c^\dagger c - 1).$$

This is an idealization of an isolated electron energy level ε_o deep under the Fermi level being probed using x-ray photoemission [41]. The energetic electron exiting the system leaves behind a hole and the electron cloud responds to this imbalance of Coulomb forces by undergoing quantized long range oscillations (plasmons) at multiples of ω_o. The corrected self-energy for this case is

$$\Sigma(t) = g^2 \Sigma_o(t) \mathcal{P}(t) = g^2[-ie^{-i(\varepsilon_o - \omega_o)t} \theta(t)] \mathcal{P}(t).$$

For a single energy level, there is no interband scattering correction in Eq. (11),

$$\mathcal{P}(t) = 1 + \left[-ig^2 \int_0^t dt_2 \int_0^{t_2} d\tau\, e^{i\varepsilon_o \tau} \Sigma_o(\tau) \mathcal{P}(t_2)\right].$$

Expanding power series on both sides and comparing terms of the same order in g^2, we generate the following higher order corrections:

$$C_m(t) = \frac{C(t)^m}{m!} \quad \text{and} \quad C(t) = \left[\frac{e^{i\omega_o t} - i\omega_o t - 1}{\omega_o^2}\right].$$

Summing all of these corrections results in the exact result from Eq. (7).

The time-ordered cumulant expression in [36–39] was derived assuming that the nth orbital's cumulant $C(n,t)$ depends only on the nth orbital's self-energy $\Sigma(n,t)$, thereby neglecting boson mediated interband scattering effects. In power series language for the Holstein model, this means that the band gap $\Delta \gg \omega_o$ and each orbital essentially is an independent core-hole problem with $P_I C$ set to zero and the corrections governed by P^{SC} alone.

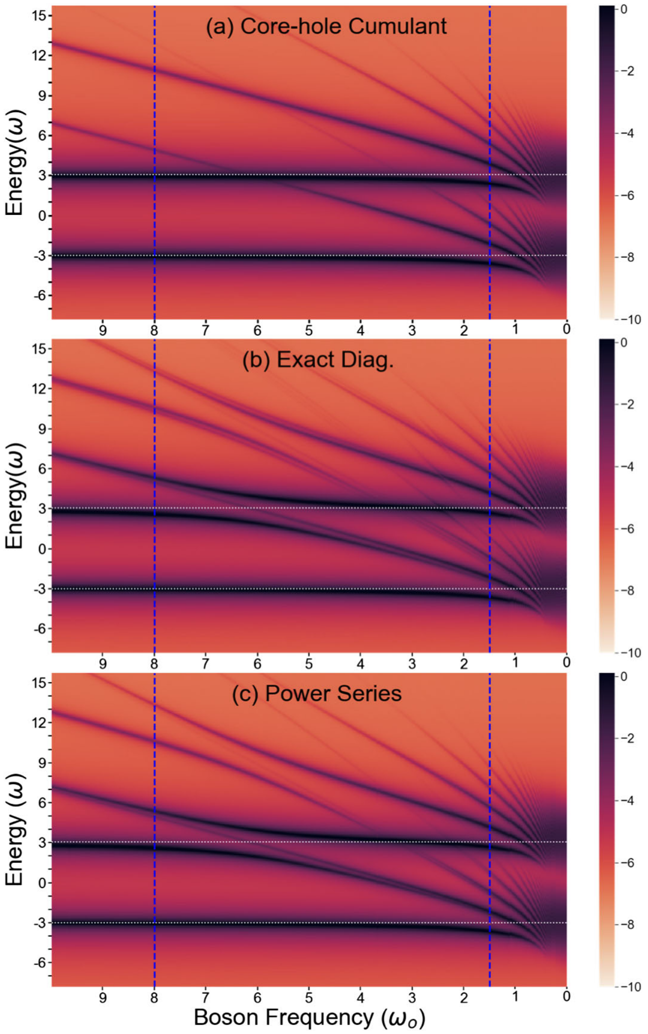

FIG. 2. Natural log of spectral function from (a) core-hole cumulant, (b) exact diagonalization, and (c) power series for $\varepsilon_\pm = \mp 3$ (horizontal white dotted lines) and ω_o between 10 and 0.1. Blue vertical lines separate the three distinct regions.

In the other limit ($\Delta \ll \omega_o$), the satellites are so far away that they do not modify the quasiparticle appreciably. Hence, both P^{SC} and P^{IC} are small and scale roughly equally, so they can be approximated as being independent of the orbital index in (11). This orbital independence lets us use the temporal contraction (9) for P^{IC} regardless of orbital identity, thereby giving retarded-cumulant correction [33,40,42]. The details of both derivations are in the Supplemental Material to this Letter [35].

Comparison between methods.—We numerically compute and compare the spectral functions from power series, exact diagonalization ($N = 50$ boson basis), and core-hole cumulant for problem (1) with $\varepsilon_\mp = \pm 3$, ω_o from 10 to 0.1, spectral broadening of 0.1, and a strong coupling parameter of $g = 1$ in Fig. 2. Depending on the magnitude of ω_o/Δ, Fig. 2 separates into three distinct regions roughly demarcated by dashed blue lines.

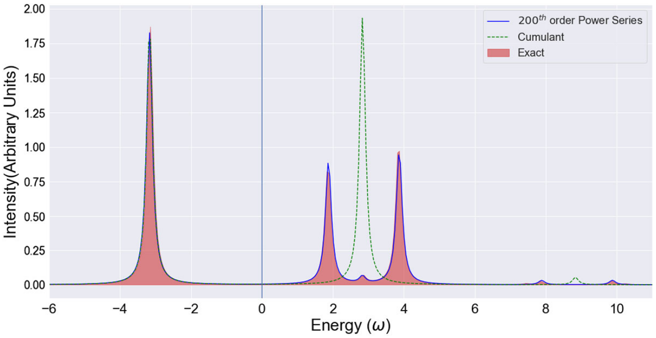

FIG. 3. Spectral function with $g = 1$, $\varepsilon_\pm = \mp 3$, and $\omega_o = 6$ from three methods. The power series, unlike the cumulant, captures the antibonding orbital splitting.

The first region is the weak coupling regime of $\omega_o \gg \Delta$—here $\omega_o > 8$. Here, both ($\pm$) plasmon satellites are far away from quasiparticles and therefore their effect on the quasiparticle energy and weight is negligible. This is most prominently seen from the negligible change in quasiparticle energies from noninteracting energies $\varepsilon_\pm$. Here, the cumulant adequately captures all the exact spectral features correctly.

The second region has $\omega_o \approx \Delta$—here $8 > \omega_o > 1.5$. A huge shift of spectral weight occurs from bonding to antibonding orbital, effectively splitting the antibonding orbital as well as the respective shakeoffs into two (between ω_o of four and seven). These are captured exactly by power series Fig. 3 but not by cumulant because they lack proper accounting of interband interactions.

The third region is when $\omega_o \ll \Delta$—here $\omega_o < 1.5$. Here bosonic events are extremely localized around the noninteracting energy and (+) bosons dominate the process. Therefore, interband correction is vanishingly small and the solution is dominated by self-correction, i.e., core-hole-like cumulant. We observe this in all three spectral functions, although both exact and power series solutions become computationally expensive—the former due to large boson number being necessary and the latter due to large convergence order.

Conclusion.—In this Letter, we derived a general power-series-based method that mitigates all the problems of cumulant methods, is practical to implement, and reproduces the exact result in a finite basis for this problem within the spectral broadening used. We also identified three important regimes of this problem as a function of boson energy and elucidated where the cumulant works, why the cumulant works, and when it fails. Recently, new works highlighting the importance of self-consistency in cumulant approximation [43,44] have emerged. Hence, we hope to extend this Letter to real multielectron systems with strong plasmon resonances, a sketch of which is provided in the Supplemental Material [35].

We heartily thank Paul J. Robinson and Professor David Reichman for useful discussions regarding the validity of our method as well as suggestions for the future applicability of our work.

---

[*] bipulpandey2004@uchicago.edu

[1] E. Goulielmakis, Z.-H. Loh, A. Wirth, R. Santra, N. Rohringer, V. S. Yakovlev, S. Zherebtsov, T. Pfeifer, A. M. Azzeer, M. F. Kling, S. R. Leone, and F. Krausz, Nature (London) **466**, 739 (2010).

[2] C. Lemell, S. Neppl, G. Wachter, K. Tőkési, R. Ernstorfer, P. Feulner, R. Kienberger, and J. Burgdörfer, Phys. Rev. B **91**, 241101(R) (2015).

[3] S. Neppl, R. Ernstorfer, A. L. Cavalieri, C. Lemell, G. Wachter, E. Magerl, E. M. Bothschafter, M. Jobst, M. Hofstetter, U. Kleineberg, J. V. Barth, D. Menzel, J. Burgdörfer, P. Feulner, F. Krausz, and R. Kienberger, Nature (London) **517**, 342 (2015).

[4] Z. Wang *et al.*, Nat. Mater. **15**, 835 (2016).

[5] M. Kang, S. W. Jung, W. J. Shin, Y. Sohn, S. H. Ryu, T. K. Kim, M. Hoesch, and K. S. Kim, Nat. Mater. **17**, 676 (2018).

[6] J. L. M. van Mechelen, D. van der Marel, C. Grimaldi, A. B. Kuzmenko, N. P. Armitage, N. Reyren, H. Hagemann, and I. I. Mazin, Phys. Rev. Lett. **100**, 226403 (2008).

[7] J. T. Devreese, S. N. Klimin, J. L. M. van Mechelen, and D. van der Marel, Phys. Rev. B **81**, 125119 (2010).

[8] Z. Wang *et al.*, Nat. Mater. **15**, 835 (2016).

[9] A. G. Swartz, H. Inoue, T. A. Merz, Y. Hikita, S. Raghu, T. P. Devereaux, S. Johnston, and H. Y. Hwang, Proc. Natl. Acad. Sci. U.S.A. **115**, 1475 (2018).

[10] A. Edelman and P. B. Littlewood, arXiv:2111.03138.

[11] O. Rösch, O. Gunnarsson, X. J. Zhou, T. Yoshida, T. Sasagawa, A. Fujimori, Z. Hussain, Z.-X. Shen, and S. Uchida, Phys. Rev. Lett. **95**, 227002 (2005).

[12] A. Damascelli, Z. Hussain, and Z.-X. Shen, Rev. Mod. Phys. **75**, 473 (2003).

[13] E. Baldini, M. A. Sentef, S. Acharya, T. Brumme, E. Sheveleva, F. Lyzwa, E. Pomjakushina, C. Bernhard, M. v. Schilfgaarde, F. Carbone, A. Rubio, and C. Weber, Proc. Natl. Acad. Sci. U.S.A. **117**, 6409 (2020).

[14] S. Yang, J. A. Sobota, D. Leuenberger, A. F. Kemper, J. J. Lee, F. T. Schmitt, W. Li, R. G. Moore, P. S. Kirchmann, and Z.-X. Shen, Nano Lett. **15**, 4150 (2015).

[15] M. Mohamed, M. M. May, M. Kanis, M. Brützam, R. Uecker, R. v. d. Krol, C. Janowitz, and M. Mulazzi, RSC Adv. **9**, 15606 (2019).

[16] X.-C. Ma, Y. Dai, L. Yu, and B.-B. Huang, Light Sci. Appl. **5**, e16017 (2016).

[17] I. N. Hulea, S. Fratini, H. Xie, C. L. Mulder, N. N. Iossad, G. Rastelli, S. Ciuchi, and A. F. Morpurgo, Nat. Mater. **5**, 982 (2006).

[18] J. M. Riley, F. Caruso, C. Verdi, L. B. Duffy, M. D. Watson, L. Bawden, K. Volckaert, G. van der Laan, T. Hesjedal, M. Hoesch, F. Giustino, and P. D. C. King, Nat. Commun. **9**, 2305 (2018).

[19] L. Hedin, Phys. Rev. **139**, A796 (1965).

[20] E. E. Salpeter and H. A. Bethe, Phys. Rev. **84**, 1232 (1951).

[21] J. Deslippe, G. Samsonidze, D. A. Strubbe, M. Jain, M. L. Cohen, and S. G. Louie, Comput. Phys. Commun. **183**, 1269 (2012).

[22] M. Govoni and G. Galli, J. Chem. Theory Comput. **11**, 2680 (2015).

[23] Z. Li, G. Antonius, M. Wu, F. H. da Jornada, and S. G. Louie, Phys. Rev. Lett. **122**, 186402 (2019).

[24] S. Acharya, D. Pashov, B. Cunningham, A. N. Rudenko, M. Rösner, M. Grüning, M. van Schilfgaarde, and M. I. Katsnelson, Phys. Rev. B **104**, 155109 (2021).

[25] H.-Y. Chen, D. Sangalli, and M. Bernardi, Phys. Rev. Lett. **125**, 107401 (2020).

[26] D. Foerster, P. Koval, and D. Sánchez-Portal, J. Chem. Phys. **135**, 074105 (2011).

[27] M. Marsili, E. Mosconi, F. De Angelis, and P. Umari, Phys. Rev. B **95**, 075415 (2017).

[28] D. C. Langreth, Phys. Rev. B **1**, 471 (1970).

[29] E. J. Heller, E. B. Stechel, and M. J. Davis, J. Chem. Phys. **71**, 4759 (1979).

[30] E. J. Heller, E. B. Stechel, and M. J. Davis, J. Chem. Phys. **73**, 4720 (1980).

[31] J. Ranninger and U. Thibblin, Phys. Rev. B **45**, 7730 (1992).

[32] O. Gunnarsson, V. Meden, and K. Schönhammer, Phys. Rev. B **50**, 10462 (1994).

[33] J. J. Kas, J. J. Rehr, and L. Reining, Phys. Rev. B **90**, 085112 (2014).

[34] G. L. Goodvin, M. Berciu, and G. A. Sawatzky, Phys. Rev. B **74**, 245104 (2006).

[35] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.129.136401 for brief description of electron and boson Green’s function, definition of electron spectral function, and the derivation of various cumulant approximation to the electron green's function in Holstein two site problem.

[36] F. Aryasetiawan, L. Hedin, and K. Karlsson, Phys. Rev. Lett. **77**, 2268 (1996).

[37] B. Gumhalter, V. Kovač, F. Caruso, H. Lambert, and F. Giustino, Phys. Rev. B **94**, 035103 (2016).

[38] F. Caruso, H. Lambert, and F. Giustino, Phys. Rev. Lett. **114**, 146404 (2015).

[39] J. Lischner, D. Vigil-Fowler, and S. G. Louie, Phys. Rev. Lett. **110**, 146801 (2013).

[40] M. Guzzo, J. J. Kas, L. Sponza, C. Giorgetti, F. Sottile, D. Pierucci, M. G. Silly, F. Sirotti, J. J. Rehr, and L. Reining, Phys. Rev. B **89**, 085425 (2014).

[41] F. Offi, W. S. M. Werner, M. Sacchi, P. Torelli, M. Cautero, G. Cautero, A. Fondacaro, S. Huotari, G. Monaco, G. Paolicelli, W. Smekal, G. Stefani, and G. Panaccione, Phys. Rev. B **76**, 085422 (2007).

[42] J. S. Zhou, M. Gatti, J. J. Kas, J. J. Rehr, and L. Reining, Phys. Rev. B **97**, 035137 (2018).

[43] P. J. Robinson, I. S. Dunn, and D. R. Reichman, Phys. Rev. B **105**, 224304 (2022).

[44] P. J. Robinson, I. S. Dunn, and D. R. Reichman, Phys. Rev. B **105**, 224305 (2022).