Polarons in highly doped atomically thin graphitic materials.

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Polaron spectral functions are computed for highly doped graphene-on-substrate and other atomically thin graphitic systems using the diagrammatic Monte Carlo technique. The specific aim is to investigate the effects of interaction on spectral functions when the symmetry between sub-lattices of a honeycomb lattice has been broken by the substrate or ionicity, inducing a band gap. Introduction of electron-phonon coupling leads to several polaronic features, such as band-flattening and changes in particle lifetimes. At the K point, differences between energies on each sub-lattice increase with electron-phonon coupling, indicating an augmented transport gap, while the spectral gap decreases slightly. Effects of phonon dispersion and long-range interactions are investigated, and found to lead to only quantitative changes in spectra.

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INTRODUCTION

The graphene hexagonal lattice leads to exceptional electronic properties: a zero bandgap semiconductor with very high mobilities, and essentially massless Dirac fermions [1]. This has already led to a number of applications, but transistors for digital applications remain elusive because they require a gap. While a gap cannot be induced in suspended monolayer graphene, recent experimental work using angle resolved photo-emission spectroscopy (ARPES) has shown that there may be bandgaps in graphene on certain substrates: ARPES measurements have found a gap in graphene on a monolayer of intercalated gold on ruthenium [2] and there has been significant debate regarding whether a gap is present in monolayer graphene on silicon carbide [3, 4].

The debate about the existence of the gap seen for graphene on SiC relates to the interpretation of ARPES measurements [5]. The authors of Ref. [6] claim that there is a gap of around 0.26eV because of modulation of the potential due to the substrate, however it has been claimed that this gap is a mis-interpretation of other excitations such as polarons and polaritons [3, 4]. ARPES measurements are carried out using either doping or gating to move the system away from half filling. For example, in Ref. [3] the Fermi energy, $E_F$, is shifted by around $0.4eV$. Bostwick et al. [4] consider a system where $E_F$ is shifted by 0.45eV. Where evidence has been found for the opening of the gap in the ruthenium system due to a breaking of the symmetry of the two carbon sub-lattices in graphene, the Fermi energy was reported to be around 0.15eV below the Dirac point [2]. Therefore, the effects of doping on spectral functions are of interest.

An alternative material with a honeycomb lattice and a bandgap resulting from a modulated potential is boron nitride (BN), which can be mechanically exfoliated in atomically thin layers [7]. Atomically thick hexagonal BN (h-BN) bonds through $sp^3$ hybridization (just as in graphene) and has itinerant electrons in $\pi$ orbitals [8]. In BN, ionicity means that $\pi$ orbitals on N sites are shifted up in energy by $+\Delta$, with a decrease in energy of $-\Delta$ on B sites, causing a gap of order $2\Delta$. Ab-initio simulations have established hopping in the BN monolayer to be $t = 2.33eV$ [9] and that the parameter, $\Delta = 1.96eV = 0.84t$. Experiments on monolayer h-BN find a gap of 5.56eV [10]. Longitudinal acoustic (LA) phonons energies peak at around 140meV at the M point, and transverse acoustic (TA) phonons at around 110meV at the K point. Optical phonon energies range between 160meV and 200meV [11], and strong coupling between electrons and phonons is expected because individual sites have an net charge in the ionic materials.

Several other atomically thin materials can be mechanically exfoliated, including SnS$_2$, CdI$_2$ and MoS$_2$, but these have the chalcogenide structure, rather than the graphene honeycomb lattice. Other layered materials that have a honeycomb structure include GaN, which has a bandgap of 2.15eV and can be grown in thin films [12]. The related AlN can also exist in a hexagonal structure with bandgap 6.28eV [13]. Following the discovery of silicene [14], it is possible that other III-V semiconductors can be encouraged to grow in thin hexagonal films.

Recently, I calculated that gaps caused by modulated potentials on honeycomb lattices may be enhanced by introducing strong electron-phonon coupling through a highly polarizable superstrate [15, 16]. Effective electron-electron interactions can be induced via a strong interaction between the electrons in a graphene monolayer and phonons in a strongly polarizable substrate because of limited out of plane screening, similar to that seen for quasi-2D materials [17]. Such interactions have been experimentally demonstrated between carbon nanotubes and a SiO$_2$ substrate [18], and are necessary to account for the lower mobilities of graphene on SiO$_2$ [19]. These interactions will form polaronic states and affect the overall electronic structure in the graphene monolayer. For the graphene-on-substrate systems with small gaps, devices could be covered with highly polarisable su-
The model Hamiltonian is introduced in Sec. IV. Details of the extensions to DQMC specific to graphene are explained in Sec. V. Sec. VI presents detailed spectral functions for a range of model parameters. A summary and conclusions are given in Sec. VII.

MODEL HAMILTONIAN

In this section, I introduce details of a model Hamiltonian for polaronic effects in gapped systems. Three components are required to examine polaronic interactions between graphene and surface phonons in a substrate (or superstrate): (a) Intersite hopping within the hexagonal plane, which is well known to properly account for the band structure of monolayer graphene. (b) An electron-phonon interaction to account for any polaronic effects from interaction with surface phonons in the substrate. (c) Direct Coulomb interaction between the electrons and substrate. The same components are valid for monolayers of ionic graphitic materials such as BN.

A complication of the Hamiltonian required to describe graphene, is that a basis of two atoms is needed to represent the honeycomb lattice. This leads to a Hamiltonian with the form,

$$H_{\text{total}} = \sum_k \left( \tilde{g} a_k^\dagger a_k + \tilde{g}^* c_k^\dagger c_k + \phi_k b_k^\dagger b_k + \phi_k^* d_k^\dagger d_k \right)$$

The Fröhlich form for the electron-phonon interaction has been demonstrated experimentally for carbon nanotubes on SiO₂ [13], is theoretically proposed for quasi-2D systems where out of plane hopping is low [17] and has also been found necessary to account for mobilities for graphene on substrate systems [19]. The interactions between electrons in the graphene and polarizable ions in the substrate are shown schematically in Fig. 1. There are only weak interactions between electrons and phonons within the graphene plane, accounting for the very high mobility of suspended graphene. In ionic materials such as BN, this problem is complicated by the need to account for large interactions between the graphene plane and substrate systems [19]. The interactions between electrons and substrate are shown schematically in Fig. 1.

The term $H_{\text{total}}$ describes the electron-phonon interaction. Phonons with momentum $q$ are created on A sites with $b_q^\dagger$ and on the B sublattice with $d_q^\dagger$. Thus, interactions between electrons and phonons on the same sub-lattices have magnitude $\tilde{g}_q$, and interactions between different sub-lattices have magnitude $\tilde{g}_q$. The Fröhlich form for the electron-phonon interaction has been demonstrated experimentally for carbon nanotubes on SiO₂ [13], is theoretically proposed for quasi-2D systems where out of plane hopping is low [17] and has also been found necessary to account for mobilities for graphene on substrate systems [19]. The interactions between electrons in the graphene and polarizable ions in the substrate are shown schematically in Fig. 1.
as BN, in plane interactions may be stronger. Two interactions are considered: a Holstein interaction, where the Fourier transform of the force function, is momentum independent and $\tilde{g} = 0$, and a Fröhlich interaction, where the Fourier transform, $g\tilde{q} = \sum_n g_0[n] e^{i\tilde{q} \cdot n}/\sqrt{N}$, of the force function is truncated at nearest neighbors so $g\tilde{q} \propto \exp(i\tilde{q} \delta_i)$.

$H_{ph}$ is the energy of the phonons in the substrate (treated as harmonic oscillators, and including both kinetic and potential energy of the ions), and the phonon dispersion is $\Omega_{q} = \Omega + \Delta \Omega (\cos(q_x\sqrt{3}) + 2 \cos(q_y\sqrt{3}/2) \cos(3q_x/2))$, where $\Delta\Omega$ controls the width of the phonon band. It is usual to define a dimensionless electron-phonon coupling, $\lambda = \Phi(0,0)/tM\Omega^2$, where the effective interaction $\Phi(n, n') = \sum_m g_m[n] g_m[n']/(2M\Omega)$, where $g_m[n] = \kappa/(1 + |n - m|^2)^{3/2}$ and $\kappa$ is a coupling constant. Typical effects of the electron-phonon interaction in conventional semiconductors are the generation of polarons, leading to changes in the bandstructure and thus the effective mass of the carriers, modification of the Landau levels and changes in the optical properties of the material such as absorption peaks in the mid infra-red.

To complete the model of graphene on a substrate, $H_{static}$ describes interaction between electrons in the monolayer and a static potential, $V(\mathbf{r}_i)$, induced by the substrate (where $\mathbf{r}_i$ are vectors to lattice sites). Here, a modulated potential is considered where A sites have energy $\Delta$ and B sites $-\Delta$ leading to breaking of the symmetry between A and B sub-lattices and giving rise to a gap. Such a form has been suggested to explain gaps in graphene on SiC [3] and graphene on rubidium [2], and is the standard form used in tight binding models of BN [9]. A similar electron-phonon Hamiltonian without the static potential was considered by Covaci and Berciu [20].

Solution of this Hamiltonian is extremely involved, and

FIG. 2. Image plots of the graphene spectral function across the Brillouin zone. Various $\Delta$ and $\lambda$ are shown. The non-interacting dispersion is overlaid. At $\Delta = 0$, A and B sub-lattices are symmetrical, leading to identical spectral functions, so only a single panel is shown for each $\lambda$. Comparison with the non-interacting dispersion shows that there is some flattening of the band close to the K point. The spectral function is sharp close to the K point. In the vicinity of the $\Gamma$ point, the spectral function is also sharp for states within an energy $\hbar \Omega$ of the bottom of the band. Quasi-particle lifetime (related to the inverse of the width) is greatly reduced close to the tops of the bands. Increase in $\Delta$ breaks the AB symmetry. Some band flattening is seen. There is also a weak excitation associated with B sites at higher energies, which touches the lower band. Panel (a) $\Delta = 0$, $\lambda = 0.7$, (b) $\Delta = 0$, $\lambda = 2.8$ where A and B site electrons are identical. (c) and (d) $\Delta = 0.5t$, $\lambda = 0.7$, and (e) and (f) $\Delta = t$, $\lambda = 0.7$, showing A and B site electrons respectively. There is only a single electron, so the chemical potential is at the bottom of the band, $E_0$. The origin is arbitrary, but for consistency has been taken as the point where the unperturbed bands cross.
complications arise because electron-phonon interactions are retarded. The next section describes how to solve the polaron problem for the graphene lattice using DQMC.

**METHOD: DIAGRAMMATIC QUANTUM MONTE CARLO**

I use the diagrammatic quantum Monte Carlo (DQMC) method to establish the properties of polarons on the graphene lattice [21, 22]. In order to take account of the bipartite lattice and AB modulated potential, the DQMC method has to be modified to include basis. This is achieved by considering the non-interacting Green function to have a matrix form. A slight complication is presented by the off-diagonal terms of this matrix, which are complex. This could in principle lead to a phase problem (which is a generalized sign problem). The complex phase is found to be very small when measuring the on-site Green functions of interest here. For imaginary time, $\tau_f > \tau_i$ and at absolute zero, the Green functions are defined as follows:

$$G = \begin{pmatrix} G_{AA} & G_{AB} \\ G_{BA} & G_{BB} \end{pmatrix}$$

$$= \begin{pmatrix} -\langle a(\tau_f) a^{\dagger}(\tau_i) \rangle & -\langle a(\tau_f) c^{\dagger}(\tau_i) \rangle \\ -\langle c(\tau_f) a^{\dagger}(\tau_i) \rangle & -\langle c(\tau_f) c^{\dagger}(\tau_i) \rangle \end{pmatrix} ,$$

where,

$$G_{AA} = \frac{[\exp(-E_B(\tau_f - \tau_i)) + \exp(-E_A(\tau_f - \tau_i))]^2}{2 \sqrt{\phi_k^2 + \Delta^2}} + \Delta [\exp(-E_A(\tau_f - \tau_i)) - \exp(-E_B(\tau_f - \tau_i))]$$

$$G_{BB} = \frac{[\exp(-E_B(\tau_f - \tau_i)) + \exp(-E_A(\tau_f - \tau_i))]^2}{2 \sqrt{\phi_k^2 + \Delta^2}} + \Delta [\exp(-E_B(\tau_f - \tau_i)) - \exp(-E_A(\tau_f - \tau_i))]$$

$$G_{AB} = \frac{\phi_k [\exp(-E_B(\tau_f - \tau_i)) - \exp(-E_A(\tau_f - \tau_i))]^2}{2 \sqrt{\phi_k^2 + \Delta^2}}$$

$$G_{BA} = \frac{\phi_k^* [\exp(-E_B(\tau_f - \tau_i)) - \exp(-E_A(\tau_f - \tau_i))]^2}{2 \sqrt{\phi_k^2 + \Delta^2}}.$$
stein interaction with local phonons have the form the electron propagator. The propagators for the Hol-
inserting and removing interaction lines into or from
the diagram.

\[ \int_{0}^{\tau} G(t) \right. \]

ordered between times \( 0 \) and \( \tau \)
contains a single electron. Since \( G \)
functions are zero valued because the polaron only con-

\[ \Delta = 0, \Delta = \frac{t}{2} \]

functions across the Brillouin zone for various \( \Delta \) and \( \lambda \). The gap opening is clearly visible.

The same data as Fig. 2 is shown with both \( A \) and \( B \) sub-
lattices superimposed on the same plot. From top to bottom,
the ability to construct spectral functions from contin-
uation at absolute zero where features can be very
sharp. Each configuration of the spectral func-
tion is weighted as

\[ \exp(-\omega_0 \tau) \delta_{XX} \]

where \( X \in \{A, B\} \) represents the sub-
lattice type at the end of the propagator and \( \delta_{XX} \)
is the Kronecker \( \delta \)-function. Sub-lattice type is fixed at
the ends of the whole diagram so that the dynamics of
each symmetry broken sub-lattice can be probed inde-
pendently.

The imaginary time Green function tails off exponen-
tially, and can vary by several orders of magnitude, which
makes direct measurement of the Green function his-
togram impossible within a reasonable time-scale. To
avoid this, a Wang-Landau algorithm is used to make
an initial guess for the histogram, so that all diagram
lengths, \( \tau \) are visited a similar number of times during
each simulation. The advantage of the Wang-Landau al-
gorithm is that it obtains the histogram extremely fast.
This histogram is not used directly for computation of
the Green function, because the bin size is finite lead-
ting to systematic errors. Rather, it is used as input for
a reweighting procedure so that all imaginary times are
visited \([22]\) and the \( \tau \) dependent Green function is calcu-
lated using the estimator given in Ref. \([22]\) which corrects
for finite histogram bin size. Proper choice of the pseudo
chemical potential, \( \mu \), speeds up the initialization of the
Wang-Landau algorithm.

It is worth noting that \( G_{AB} \) and \( G_{BA} \) have a com-
plex phase. A Monte Carlo procedure can be obtained
by keeping track of this phase \( e^{i \theta} \) such that averages of
an estimator \( O \) are given by \( \langle e^{i \theta} O \rangle / \langle e^{i \theta} \rangle \). For all cases
considered here, the average phase \( \langle e^{i \theta} \rangle \) is found to be
extremely close to 1, and no expectation values had a
complex component after averaging. There is no obvi-
ous reason why the phase should cancel (unlike in the
1D case where the signs exactly cancel \([23]\)) and for large
numbers of particles, the phase could become a problem.

In order to obtain spectral functions, stochastic analy-
lytic inference is used \([24]\). Green functions are built
up from \( \delta \)-functions that can be moved continuously in
frequency using a separate Monte Carlo update scheme.
The ability to construct spectral functions from contin-
uous frequencies is necessary to obtain reliable analytic
continuation at absolute zero where features can be very
sharply peaked. Each configuration of the spectral func-
tion is weighted as \( w \propto \exp(-\chi^2 / 2\alpha) \) and the factor \( \alpha \)
is reduced from a large value until the average \( \chi^2 < N_G \),
the number of points in the Green function. Averages are
then taken. Additional global updates (where all points
are shifted simultaneously) have been included in the
procedure to ensure that the algorithm is ergodic.

RESULTS

This section begins by examining how the opening of
a band-gap affects the spectral functions when the
electron-phonon coupling is switched on. The phonon
energy is set as \( h \Omega = t \) unless otherwise specified. This

\[ E_A = \mu + \sqrt{\Delta^2 + |\phi_k|^2} \]

and \( E_B = \mu - \sqrt{\Delta^2 + |\phi_k|^2} \). Here, the pseudo chemical potential \( \mu \) allows greater control of
the algorithm, but since a single particle is simulated, the
ture chemical potential lies at the bottom of the band.
Thus, the results are only accurate when the electron
density is low (i.e. the system is doped well away from
half-filling). For \( \tau_f < \tau_i \) and absolute zero, all Green
functions are zero valued because the polaron only con-
tains a single electron. Since \( G(\tau_f < \tau_i) = 0 \), vertices are
ordered between times \( 0 \) and \( \tau \), where \( \tau \) is the length of
the diagram.

In its most basic form, the algorithm proceeds by
inserting and removing interaction lines into or from
the electron propagator. The propagators for the Hol-
stein interaction with local phonons have the form

\[ \exp(-\omega_0 \tau) \delta_{XX} \]
very high value is chosen so that features relating to polarons can be distinguished easily. Note that phonons of this energy are still in the adiabatic regime at around one third of the half band width. Naturally, this energy is much higher than that of any phonons in graphene or BN, or of any surface phonons in the substrate, and smaller values will also be discussed later in the article. Values for $\Delta$ ranging from 0 to $t$ are also large for the same reason. For simplicity (unless specified) the out-of-plane interaction $g_{kq}$ is approximated to be momentum independent, and $\tilde{g} = 0$, leading to a local Holstein interaction.

Fig. 2 shows how the graphene spectral function, $A(E)$, changes across the Brillouin zone. Spectral functions are computed from the full Green functions, $G_{AA}(\tau)$ and $G_{BB}(\tau)$, with $G$ calculated on a logarithmic mesh with 500 points. Separate image plots can be seen for A and B type electrons to make the specific contributions from each sub-lattice clear, and the non-interacting band structure is superimposed for comparison. A moderate dimensionless electron-phonon coupling of $\lambda = 0.7$ is chosen, with the exception of panel (b), which shows spectral functions for the larger $\lambda = 2.8$. Panels (a) and (b) correspond to $\Delta = 0$, (c) and (d) to $\Delta = t/2$ and (e) and (f) to $\Delta = t$.

The main features of Fig. 2 are: (1) The quasi-particle peaks are sharp at low energies $E_0 < E < \hbar\Omega$ (where $E_0$ is the polaron ground state energy) but broaden significantly for higher energies. This is especially noticeable in panels (a), (d) and (f). (2) A clearly identifiable polaron band (split off from the main dispersion) can be seen for large $\lambda$ in panel (b). (3) Asymmetry between electrons on site A and site B increases with $\Delta$. (4) The spectral gap can be seen to increase with $\Delta$. The spectral gap is slightly smaller than $\Delta$ due to broadening of the quasiparticle peak. (5) The beginnings of a flat polaron band can be seen for $\Delta = t$ in panel (f) and is just visible in panel (d) for $\Delta = t/2$. Again, it should be noted that these results are for polarons (a single electron at the bottom of an empty band interacting with phonon modes). As the Fermi energy is changed, the spectral function close to the chemical potential may be modified significantly. Far from the chemical potential, differences should be less pronounced. Here, the spectral gap is defined as the distance between the peaks of the spectral function close to the Dirac point.

When $\Delta = 0$, A and B sites are symmetrical and the spectral function for each sub-lattice is identical, so re-
FIG. 6. (Color online) Graphene spectral function across Brillouin zone. $\Delta = t$ and various $\lambda$. Data is as Fig. 5 but with results for both sub-lattices plotted together. It can be seen that the gap is robust against increase in $\lambda$, although a number of additional excitations appear.

The spectral function is also sharply peaked close to the K point. By examining the zero gap states (Fig. 2(a) and (b)) it is possible to consider whether it is possible that polaronic states could simulate a gap. Examination of Fig. 2(a) shows that there is a flattening of the dispersion near the K point (highlighted by comparison with the non-interacting dispersion) accompanying a decrease in the width of the quasi-particle peak. The flattening of the dispersion close to the K point is associated with a steepening of the dispersion along the KM and KG lines, giving the dispersion the appearance of a waterfall. The waterfall-like features are at variance with the idea that polaronic features could look like a band gap [6], since emulation of a gap would require the dispersion to show the opposite shape to that seen here: a very steep band structure close to K with a rapid change of gradient leading to flattening between K and M. Panel (b) shows that the spectral weight gets broader with increased $\lambda$, indicating that it is also likely that finite quasi-particle lifetimes contribute to an obscuring of any gap rather than any apparent gap opening. Since the results are taken with the chemical potential at the bottom of the band, the results are speculative and further studies at higher densities would be required to reach a full conclusion.

A major feature in panel (b) for $\lambda = 2.8$ is the emergence of a flat polaron band, which separates off from the lower band. Such a feature typically corresponds to the energy required to excite a real phonon, and is singular in basic Migdal–Eliashberg theories of electron-phonon interactions (see e.g. Ref. 25). The large electron-phonon coupling leads to a significant decrease in the quasi-particle lifetime at large energies, however, the band flattening seen around the K point (which is unconnected to the polaron band) persists. The ground state polaron energy is lowered due to the polaron self-interaction (seen as the offset from the non-interacting band at the Γ point).

Increase in $\Delta$ breaks the symmetry between A and B sub-lattices, and this can be seen in Fig. 2 panels (c)-(f). A gap opens as $\Delta$ is increased. A band at high energies, roughly tracing the dispersion of the ungapped, non-interacting band, can be seen in panels (d) and (f), although its spectral weight is extremely small. The origins of this band are unclear. Increased $\Delta$ also leads to a flat polaron band, that separates from the main band (it can be seen as the lowest energy feature at the K point). This feature is only just visible in panel (d), but is clearly separated from the main band in panel (f). The appearance of this band at weak $\lambda$ is a consequence of polaron localization at large $\Delta$, which increases polaron self-interaction (i.e. the effective $\lambda$ is increased by the localization).
FIG. 7. As Fig. 5 with the leading corrections from Fröhlich interactions included. There are only moderate changes to the results, consistent with a small reduction in effective λ.

FIG. 8. (Color online) Graphene spectral function at the K point. ∆ = t and λ is varied. The spectral gap (seen at approximately E = 5t) is robust. The spectral functions broaden with increased λ, and the polaron band rapidly drops in energy leading to an enhanced transport gap.

To demonstrate that the effects are not artifacts of the local Holstein interaction, Fig. 3 shows the spectral functions computed for interactions that include nearest neighbor forces, the leading correction to the Fröhlich interaction (N.B. Sums associated with the Fourier transforms of the force functions are truncated so that they are not prohibitively computationally expensive. A truncated near neighbor interaction contains sufficient physics to obtain good agreement with the full interaction [26,27]). The plots are qualitatively similar to those in Fig. 2. The main difference is that the longer range forces slightly reduce the effects of interactions at a particular λ: for the Fröhlich interaction, there is slightly less drop in the ground state energy of the polaron band, and the flat features appear at slightly higher energies relative to the bottom of the band. The slight increase in the width of the polaron band occurs because the distortions associated with the Fröhlich polaron are already preformed before the electron hops between sites, so the effective mass is smaller. The differences between Holstein and Fröhlich interactions are more pronounced for larger interaction strengths, as can be seen by comparing the λ = 2.8 results.

To clarify the effects of interaction and bare gap on the spectral function, Fig. 4 shows the same data, but with both spectral function types superimposed on the same plot. For clarity, spectral weight below a cutoff of less than 0.1 is not shown (variations on this order of magnitude are not distinguishable below this scale, and the resulting curve appears as a series of straight horizontal lines obscuring the plots). From top to bottom ∆ = 0, ∆ = t/2 and ∆ = t and λ = 0.7 in all panels. The
FIG. 9. Effects of dispersion, $\Delta \Omega$, and phonon frequency, $\Omega$ on the spectral function. $\lambda = 2.8$ throughout. Broadening the phonon dispersion has no major effects on the spectral function. On the other hand, decreasing the phonon frequency leads to a broadening of all features corresponding to a sharp decrease in quasi-particle lifetime, and indicating a significant increase in scattering.

gap opening can clearly be seen. The effects of interaction, which increases the width of the spectral function at large energies (where real phonons can be created) is much clearer in these plots.

Figs. 5 and 6 show how the graphene spectral function changes across the Brillouin zone when the electron-phonon coupling is varied and $\Delta = t$ (from top to bottom, $\lambda = 0.7, 1.4, 2.1$ and $2.8$). Spectral functions for the A sub-lattice can be seen on the left of Fig. 5 (panels (a), (c), (e), (g)) and spectral functions for the B sub-lattice are shown on the right hand side of the plot (panels (b), (d), (f) and (h)). Self-interactions lower the polaron energy, and it can be seen that spectral weight at the $\Gamma$ point moves to lower energies relative to the non-interacting bands as $\lambda$ increases. At large $\lambda$, a sharply defined polaron band separates from the main band, and can be seen as a low energy band that is almost flat and has a high quasi-particle lifetime (again, this is the singular band found in conventional Migdal–Eliashberg theories). The remnant of the non-interacting band is visible as a side-band above the polaron band. The quasi-particle lifetime decreases dramatically with increased $\lambda$, seen as a broadening of the spectral function. A shadow band can be seen in the spectral functions for B sites, although it is relatively weak, and decreases in weight as $\lambda$ is increased. Examination of Fig. 6 (which shows spectral functions for both sub-lattices superimposed onto the same plot), shows that the spectral gap is robust against increase in $\lambda$, although a number of additional excitations appear. Again, for completeness, the effects of longer range interactions are shown in Fig. 7. For the larger bare band gaps, the effects of interaction range are extremely small for all values of $\lambda$.

Finally, Fig. 8 shows how the spectral function at the K point evolves as the electron-phonon coupling is increased. Three main peaks are visible: (1) The lowest corresponds to the polaron band, (2) electrons on the A sub-lattice have at least 1 excited state, and (3) B type electrons can be seen at the highest energies. In the strongly doped system, the gap between the A excited state and the B electron energy is essentially unchanged by an increase in the electron-phonon coupling, with the gap remaining robust. The polaron band can be seen splitting off from the non-interacting band, with the polaron energy dropping rapidly at small $\lambda$, followed by a sustained decrease. This is related to the flattening of the polaron band seen in Fig. 5. As the electron-phonon coupling is increased, the quasi-particle lifetime drops, seen
as a broadening of the peak. There is an increase in the energy difference between the bottom of the A band and the bottom of the B band at the Dirac point, indicating an increase in the potential barrier formed by the higher energy A sites. This indicates an increased transport gap at the K point due to an increase in the energy difference between sub-lattices. This provides additional evidence that strongly polarizable substrates and superstrates could be used to enhance transport gaps opened by a substrate. Further work is underway to examine spectral functions and gaps close to half filling.

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