Enhanced superconductivity due to forward scattering in FeSe thin films on SrTiO$_3$ substrates

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Abstract

We study the consequences of an electron–phonon (e–ph) interaction that is strongly peaked in the forward scattering ($q = 0$) direction in a two-dimensional superconductor using Migdal–Eliashberg theory. We find that strong forward scattering results in an enhanced $T_c$ that is linearly proportional to the strength of the dimensionless e–ph coupling constant $\lambda_m$ in the weak coupling limit. This interaction also produces distinct replica bands in the single-particle spectral function, similar to those observed in recent angle-resolved photoemission experiments on FeSe monolayers on SrTiO$_3$ and BaTiO$_3$ substrates. By comparing our model to photoemission experiments, we infer an e–ph coupling strength that can provide a significant portion of the observed high $T_c$ in these systems.

A flurry of scientific activities has been generated by the discovery of an enhanced superconductivity in FeSe monolayers grown on SrTiO$_3$ (STO) substrates [1–20]. On its own, bulk FeSe has a modest superconducting transition temperature $T_c \sim 9$ K [21]; however, when a monolayer is grown on an STO substrate, $T_c$ is increased dramatically [1]. Most reported $T_c$ values cluster within 55–75 K, close to the boiling point of liquid nitrogen (77 K). (A surprisingly high $T_c \sim 107$ K has also been reported in $\textit{in situ}$ transport measurements [9].) This discovery has opened a pathway to high-$T_c$ superconductivity through interface engineering, which has already produced high-$T_c$ systems such as FeSe on BaTiO$_3$ (BTO) [8] and FeTe$_{1-x}$Se$_x$ on STO [22].

Determining the origin of the $T_c$ enhancement in these interface systems is critical. At the moment, proposals include charge transfer between the substrate and FeSe [2–4, 20], electric field [6] and strain effects due to the substrate [5, 8], and lattice related effects such as enhanced electron–phonon (e–ph) coupling in the FeSe layer [1, 13, 16] or across the interface [7, 19]. Strong evidence for the latter has been provided by a recent angle-resolved photoemission spectroscopy (ARPES) study [7], which observed replica bands in the single-particle spectral function of the FeSe monolayer. These replicas are interpreted as being produced by coupling between the FeSe 3$d$ electrons and an optical oxygen phonon branch in the STO substrate. Moreover, the replica bands are complete copies of the corresponding main bands, which implies that the responsible e–ph interaction is strongly peaked in the forward scattering direction (small momentum transfers). Such momentum dependence is notable because it can enhance superconductivity in most pairing channels [23–31]. As such, this cross-interface coupling provides at the same time a suitable mechanism for the $T_c$ enhancement in the FeSe/STO and FeSe/BTO systems [7, 8].

We explore this possibility here by examining the consequences of strong forward scattering in the e–ph interaction for superconductivity and the spectral properties of a two-dimensional system. By solving the momentum dependent Eliashberg equations, we show that a pronounced forward scattering results in a $T_c$ that
scales linearly with the dimensionless $e$–$ph$ coupling constant $\lambda_{\text{m}}$ (see below) in the weak coupling limit. This is in stark contrast to the usual exponential dependence predicted by BCS theory. Furthermore, this coupling produces distinct replica structures in the spectral function similar to those observed experimentally. By comparing our model to experiments [7], we infer a significant $e$–$ph$ contribution to the total $T_c$ observed in the FeSe/STO system with a modest value of $\lambda_{\text{m}}$.

1. Formalism

To model the FeSe monolayer we consider a single-band model for the FeSe electron pockets, which includes coupling to an oxygen phonon branch in the STO substrate. The Hamiltonian is given by

$$ H = \sum_{k,\sigma} \xi_k \epsilon_{k,\sigma} c_{k,\sigma}^\dagger c_{k,\sigma} + \sum_{q} \Omega_q b_q^\dagger b_q + \frac{1}{\sqrt{N}} \sum_{k,\sigma} g(k, q) \epsilon_{k+q,\sigma} c_{k,\sigma} (b_{-q}^\dagger + b_q), $$

where $\epsilon_{k,\sigma}$ ($c_{k,\sigma}$) and $b_q^\dagger$ ($b_q$) are electron and phonon creation (annihilation) operators, respectively, $\xi_k$ is the band dispersion, $\Omega_q$ is the phonon dispersion, and $g(k, q)$ is the momentum-dependent $e$–$ph$ coupling constant.

We calculate the single-particle self-energy due to the $e$–$ph$ interaction using Migdal–Eliashberg theory. Using the Nambu notation with fermionic Matsubara frequencies $\omega_n = (2n + 1)\pi / \beta$, where $\beta = 1 / T$ is the inverse temperature, the self-energy is $\Sigma(k, i\omega_n) = i\omega_n [1 - Z(k, i\omega_n)] \tilde{\gamma} + \chi(k, i\omega_n) \tilde{\gamma} + \phi(k, i\omega_n) \tilde{\gamma}$, where $\tilde{\gamma}$ are the Pauli matrices, $Z(k, i\omega_n)$ and $\chi(k, i\omega_n)$ renormalize the single-particle mass and band dispersion, respectively, and $\phi(k, i\omega_n)$ is the anomalous self-energy, which is zero in the normal state. The self-energy is then computed by self-consistently evaluating the one-loop diagram and is given by

$$ \Sigma(k, i\omega_n) = -\frac{1}{N\beta} \sum_{q,m} g(k, q) |D(0)(q, i\omega_n - i\omega_m)|^2 \tilde{G}(k + q, i\omega_m) \tilde{\gamma}, $$

where $D(0)(q, i\omega_n) = -\frac{4\pi^2}{\omega_q + \omega_n^2}$ is the bare phonon propagator, and $\tilde{G}^{-1}(k, i\omega_n) = i\omega_n \tilde{\gamma} - \xi_k \tilde{\gamma} - \Sigma(k, i\omega_n)$ is the dressed electron propagator.

Migdal’s theorem states that the vertex corrections to considerations given above are on the order of $\lambda_{\text{m}}^2$ for a momentum-independent interaction. One might therefore wonder whether the use of Migdal–Eliashberg theory is justified in FeSe/STO, since the ratio $\Omega / E_F$ is of order unity. This is bolstered by the fact that Migdal’s theorem also breaks down in the limit that the small $q$ scattering processes dominate. In this limit, however, one can show that the vertex corrections are still proportional to the dimensionless coupling $\lambda_{\text{m}}$ [32]. As such, any corrections to the self-energy due to the diagrams neglected in Migdal–Eliashberg theory are of order $\lambda_{\text{m}}^2$, and can be treated perturbatively if the coupling is sufficiently small. As we will show, the value $\lambda_{\text{m}}$ needed to reproduce the ARPES data is small ($\lambda_{\text{m}} \sim 0.15–0.25$), indicating that we are indeed in the perturbative regime. (This is further justified by observing that the contributions to the self-energy from the second order and higher order rainbow diagrams, which are of the same order as the crossing diagrams, are small.) In this context, the work of Pietronero and coworkers is relevant [33, 34] as they have studied the contributions of the vertex corrections in the perturbative regime and found that their inclusion serves to increase the superconducting $T_c$ when the $e$–$ph$ coupling is dominated by small $q$ processes. Based on these considerations, we proceed using Migdal–Eliashberg theory assuming that corrections beyond this approach can be treated at a perturbative level and will likely increase $T_c$ further.

In what follows we parameterize the electronic dispersion as $\xi_k = -2t \left[ \cos(k_x a) + \cos(k_y a) \right] - \mu$ with $t = 75$ meV and $\mu = -235$ meV. This choice in parameters produces at $\Gamma$ an electron-like Fermi pocket with $k_y = 0.97 / a$ and a Fermi velocity $v_F = 0.12$ eV·Å/$\hbar$ along the $k_y = 0$ line, where $a$ is the in-plane lattice constant. This closely resembles the electron pocket at $M$ point measured by ARPES experiment. Since first principles calculations indicate that the relevant oxygen phonon branch in STO is relatively dispersionless near the $\Gamma$-point [14, 35], we approximate the phonon with a flat Einstein mode $\Omega_q = \Omega = 100$ meV ($\hbar = 1$), which is consistent with the observed energy separation of the replica bands [7]. Furthermore, as we are interested in the case of forward scattering, we neglect any potential fermion momentum dependence in the $e$–$ph$ interaction and set $g(q) = g_0 \exp(-|q|/q_0)$, as microscopically derived before [7, 19]. Here, $q_0$ sets the range of the coupling in momentum space. For different values of $q_0$, we adjust $g_0$ to obtain the desired value of the dimensionless $e$–$ph$ coupling constant $\lambda_{\text{m}}$, which is computed from the Fermi surface averaged mass enhancement in the normal state $\lambda_{\text{m}} = \left( -\frac{\partial E}{\partial \omega} \right)_{\omega=0}$, $\left( \frac{\partial^2 E}{\partial \omega^2} \right)_{\omega=0}$. (We are using $\lambda_{\text{m}}$ to distinguish this definition from the standard one involving a double Fermi surface average of the coupling constant $|g(k, q)|^2$. See the supplementary material [6] for further details.) Throughout we assume an $s$-wave symmetry for the gap function, consistent with the observations of a

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6 For further details, please see the online supplementary material available at stacks.iop.org/npj/18/022001/mmedia.
fully gapped state on the Fermi level [1, 7, 10, 18]. Finally, we neglect the Coulomb pseudopotential $\mu^*$ in what follows. One should therefore regard our $T_c$ values as upper bounds for the e–ph contribution to the FeSe/STO system. (Some considerations for $\mu^*$ are provided in the supplementary materials\(^6\). We find that its inclusion reduces $T_c$ by $\sim 20\%$ for typical values of $\mu^*$.)

2. Analytical results

Before proceeding to full numerical solutions, we can gain some insight by first considering the case of perfect forward scattering, where the e–ph matrix element is a delta function $g(q) = g^2 \delta_{|q|} N$ with $g^2 = \lambda_m \Omega^2$ (see supplementary material\(^5\)). In the weak coupling limit, we further set $Z(k, \omega_n) = 1$, $\chi(k, \omega_n) = 0$, and therefore $\phi(k, \omega_n) = \Delta(k, \omega_n)$. With these approximations, the gap function on the Fermi surface is given by

$$\Delta(\omega_n) = \frac{\lambda_m \Omega^2}{\beta_c} \sum_m \frac{2\Omega}{\Delta^2(\omega_m) + (\omega_n - \omega_m)^2} \omega_m^2.$$ 

To determine $T_c$, we take the ansatz $\Delta(\omega_n) = \Delta_0/[1 + (\omega_n/\Omega)^2]$ and follow the usual steps [37]: the gap equation is linearized by setting $\Delta^2(\omega_m) = 0$ for $T \sim T_c$. We then get $\omega_m = 0$ and set $\omega_n = 0$. This results in the condition for $T_c$,

$$1 = \frac{\lambda_m \Omega^2}{\beta_c} \sum_m \omega_m^2 (1 + \omega_m^2/\Omega^2)(\Omega^2 + \omega_m^2).$$

The Matsubara sum can be performed exactly, yielding our final expression

$$1 = \frac{\lambda_m \beta_c}{2} \frac{2\Omega + \Omega \cosh(\Omega \beta_c) - (3/\beta_c) \sinh(\Omega \beta_c)}{1 + \cosh(\Omega \beta_c)}.$$ 

For the case of FeSe, $T_c < \Omega$, and the hyperbolic functions dominate. To the leading order, the critical temperature is quasi-linear in the coupling strength in the weak coupling limit, $T_c = \frac{\lambda_m}{\beta_c + \beta_m} \Omega$. (A similar result was obtained in [23] in the context of the cuprates using square-well models.) For $\lambda_m = 0.16$ and $\Omega = 100$ meV one obtains $T_c = 75$ K, which is a remarkably high temperature for such a modest value of $\lambda_m$.

The increased $T_c$ should be compared to the standard BCS value obtained for a momentum-independent coupling. In this case, the linearized gap equation simplifies to [36, 37]

$$1 = \frac{\pi T_c \lambda_m}{2} \sum_{|\omega| < \Omega_0} \frac{1}{|\omega|} = \lambda_m \left[ \ln \left( \frac{\Omega_0}{2\pi T_c} \right) - \psi \left( \frac{1}{2} \right) \right],$$

where we have expanded at large $\Omega_0/T_c$ and $\psi(z)$ is the digamma function (see supplementary material\(^5\)). This form produces the usual exponential behavior for the critical temperature, $T_c = 1.13\Omega_0 \exp(-1/\lambda_m)$, which predicts a $T_c = 2.5$ K for $\lambda_m = 0.16$ and $\Omega_0 = 100$ meV.

Comparing these two results, one sees that the origin of the enhanced $T_c$ lies in the momentum decoupling [24] that occurs in the Eliashberg equations when the interaction is strongly peaked at $q = 0$. In the BCS case, the integration over the Fermi surface is equally weighted at all momenta, leading to a $\omega_m^2$ term in the BCS gap equation and subsequently a leading logarithmic behavior. In the forward scattering case, there is no integration over momentum so the $\omega_m^2$ term remains, resulting in a leading behavior that scales like $1/T_c$ (see supplementary material\(^5\)). Thus, strong forward scattering serves as an ideal mechanism for producing high-$T_c$ superconductivity [30]. Furthermore, a strong forward scattering peak in the coupling constant means that this interaction will contribute in most pairing channels [7, 23–31]. It can therefore act in conjunction with other active unconventional channels, providing another means to increase $T_c$ further.

3. Numerical results

In real materials the e–ph interaction is expected to have a finite range $q_0$ in momentum space [7]. Therefore we now consider an interaction with a finite width by numerically solving the full Eliashberg equations for an e–ph coupling constant $g(q) = g_0 \exp(-|q|/q_0)$. Figure 1 shows the superconducting gap at the lowest Matsubara frequency $\Delta(k_0, \text{i} \pi T / \beta)$ as a function of temperature for several values of $\lambda_m$ and $q_0 = 0.1\mu$. We find that the superconducting $T_c$ is already large for a modest value of $\lambda_m$ and increases approximately linearly with $\lambda_m$ in the weak coupling limit; however, the finite range of the coupling in momentum space reduces the total $T_c$ slightly with respect to the perfect forward scattering limit (see the inset of figure 1). The linear dependence of $T_c$ with respect to $\lambda_m$ may account for the wide variation of reported $T_c$ values in the literature, as differences in sample preparation or doping are likely to result in differences in the screening of the e–ph coupling and subsequently $T_c$. 

3
3.1. Replica bands

The above results show that, in principle, a modest coupling to a phonon with a forward scattering peak is capable of accounting for the large \( T_c \) enhancement observed in FeSe on STO and BTO. The natural question is then how much of the experimental \( T_c \) is accounted for by this coupling? The observed shape and intensity of the replica bands \[7, 8\] provide us with a direct means to estimate this by comparing our model to experiment. To do so, we calculate the single particle spectral function \( \sigma_{pp} = - \text{Im} G_{11}(k, \omega)/\pi \), which requires the analytic continuation of the self-energy to the real frequency axis using the method of \[41\] (see also supplementary material\[6\]). Figure 2 plots the temperature evolution of the spectral function obtained from a full numerical solution to our model for several values of \( \lambda_m \), as indicated on the left, and \( \lambda_m = 0.10 \). In all cases clear replica bands are produced by the coupling, offset in energy from the main band by a fixed energy, which is \( \Omega \) for small values of \( \lambda_m \). The separation, however, grows for increasing \( \lambda_m \). This is due to \( \omega(k, \omega) \), which shifts the main band upward in energy. This is most clearly seen in the \( \lambda_m = 0.33 \) results, where the value of \( k_F \) has visibly shrunk in the main band. In addition, for stronger values of \( \lambda_m \) we begin to see the formation of a second replica band located at \( \sim \Omega \) below the main band. Thus the observation of only a single replica band in the bandstructure of FeSe/STO is consistent with a small \( \lambda_m \). An intuitive picture for the intensity and energy splitting of the replica band can again be obtained in the limit of perfect forward scattering. On the real axis, the zero-temperature self-consistent equation for the self-

Figure 1. The superconducting gap at the smallest Matsubara frequency \( \Delta(\pi/\beta) \) as a function of temperature for various values of the e-ph coupling strength \( \lambda_m \), as indicated. The e-ph coupling constant \( g(q) \) is strongly peaked in the forward scattering direction with \( q_0 = 0.1/a \). The inset shows \( T_c \) as a function of \( \lambda_m \), which is extracted from the data in the main panel. The thin dashed line is the result in the limit of perfect forward scattering (see text). The shaded area represents the values of \( \lambda_m \) that are relevant for FeSe/STO (see supplementary material\[6\]).

Figure 2. The temperature dependence of the spectral function for several values of the e–ph coupling \( \lambda_m \).
energy in the normal state can be written as $\Sigma(\omega) = g_0^2 G(\omega + \Omega)$. For $\xi_k \rightarrow 0^-$, the lowest-order solution is $\Sigma(\omega) = \frac{g_0^2}{\omega + \Omega}$ (note that the $\xi_k \rightarrow 0$ solution can be obtained by shifting the self-energy $\Sigma(\omega) = \Sigma(\omega - \xi_k)$). The poles of the Green’s function are at $\omega = \Sigma(\omega)$, which has the solution $\omega_{\pm} = -\frac{\Omega}{2} \pm \frac{1}{2} \sqrt{\Omega^2 + 4g_0^2}$. The spectral weight of each pole is given by $Z_{\pm} = \left[1 - \frac{\partial \Sigma}{\partial \omega} \big|_{\omega = \omega_{\pm}} \right]^{-1} = \left[1 + \frac{g_0^2}{\omega_{\pm} + \Omega} \right]^{-1}$. For small $\lambda_m = g_0^2/\Omega^2$, we find that the average energy separation between the poles is $\Delta \omega = \Omega[1 + 2\lambda_m + O(\lambda_m^2)]$ and the ratio of the spectral weight is $\frac{Z_+}{Z_-} = \lambda_m + O(\lambda_m^2)$, thus providing a direct measure of $\lambda_m$.

The spectral weight ratio and energy splitting between the main and replica bands can be extracted from our numerical simulations for finite values of $q_0$. Figure 3 shows $A(k, \omega)$ for $k = (k/a, 0)$ as a function of $\lambda_m$ with $q_0 = 0.1/a$. The behavior matches our expectations gained from the perfect forward scattering limit: both the distance between the bands and the relative spectral weight grow with increasing $\lambda_m$, though the rate of increase is slower than for the case of perfect forward scattering. ARPES experiments on the FeSe/STO system [7] observe a spectral weight ratio of $\sim 0.15 - 0.2$ (see supplementary material8). Comparing to our model calculations, we extract a value of $\lambda_m \sim 0.15 - 0.2$. This corresponds to a $T_c \sim 60 - 70$ K and a gap magnitude of $\Delta \sim 10 - 15$ meV, which are consistent with measurements [1, 7, 10, 18].

In Figure 4 we present the evolution of the spectral function for increasing values of $q_0$ where $\lambda_m$ is fixed to give the same value of $Z_+/Z_-$. As expected, the replica bands are observed to smear both in energy and momentum as the value of $q_0$ is increased. This shows that a weakly momentum-dependent coupling (large $q_0$) to an optical mode does not reproduce the observation of a perfect replica band, with the same effective mass and
termination points in the Brillouin zone. Consequently, strong forward scattering is a necessary ingredient to understand the experimental observations [7].

4. Discussion

We have examined the consequences of an e–ph coupling that is strongly peaked in the forward scattering direction on the spectral properties and superconducting transition of a two-dimensional electronic system. We demonstrated that such a coupling produces distinct replica bands in the electronic bandstructure consistent with recent ARPES measurements on FeSe/STO and FeSe/BTO interface systems. In order to reproduce the experimentally observed spectral function, we find that relatively modest values of the e–ph coupling are needed with \( \lambda_{\text{int}} \sim 0.15 \sim 0.2 \). Strong forward scattering results in a momentum decoupling of the Eliashberg equations, which subsequently produces a larger superconducting \( T_c \) in comparison to the predictions of conventional BCS theory. As a result, the inferred values of \( \lambda_{\text{int}} \) predict \( T_c \) values on the order of 60–70 K due to e–ph coupling alone.

We stress that our results do not exclude the presence of another unconventional pairing channel such as spin fluctuations. The predicted values of \( T_c \) and \( \Delta \) will be reduced somewhat by the inclusion of the Coulomb pseudopotential \( \mu^\ast \). (For example, we examined the influence of \( \mu^\ast \) and found that \( T_c \) is lowered by \( \sim 20\% \) for \( \mu^\ast \sim 0.2 \) (see supplementary material). This is much smaller than the factor of 2–5 commonly obtained for conventional phonon-mediated pairing [42, 43]. This robustness is linked to the same momentum decoupling responsible for the linear dependence of \( T_c \) on \( \lambda_{\text{int}} \).) This reduction, however, can be overcome by the combination of the e–ph and unconventional interactions, since forward scattering will contribute to Cooper pairing in most channels [7]. An obvious way to distinguish between these possible scenarios is to measure the oxygen isotope effect. If a purely phononic mechanism is present then \( T_c \) should have an isotope coefficient \( \alpha = -\partial \log (T_c)/\partial \log (M) = 1/2 \), while the energy separation between the replica bands should decrease by \( \sim 0.5(18 \sim 16)/16 \sim 6\% \) for \(^{18}\text{O} \) rich substrates. Alternatively, in a multi-channel scenario, the isotope coefficient \( \alpha \) will be reduced from 1/2 when the unconventional channel is significant in comparison to the e–ph interaction [38–40]. This provides a clear means to distinguish between these scenarios.

Finally, we note that e–ph coupling with a pronounced forward scattering peak has been studied in several contexts related to of unconventional superconductivity in the cuprates [24–30] and pnictides [31]. Moreover, it is also now being addressed in the context of nematic fluctuations [44, 45]. This suggests forward scattering has a broader applicability in enhancing superconducting beyond interface systems.

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