Odd-parity superconductivity from phonon-mediated pairing

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Motivated by the proposed topological state in Cu$_2$Bi$_2$Se$_3$, we study the possibility of phonon-mediated odd-parity superconductivity in spin-orbit coupled systems with time-reversal and inversion symmetry. For such systems, we show that, in general, pure electron-phonon coupling can never lead to a triplet state with a higher critical temperature than the leading singlet state. The Coulomb pseudopotential, which is the repulsive part of the electron-electron interaction and is typically small in weakly correlated systems, is therefore critical to stabilizing the triplet state. We introduce a chirality quantum number, which identifies the electron-phonon vertex interactions that are most favorable to the triplet channel as those that conserve chirality. Applying these results to Cu$_2$Bi$_2$Se$_3$, we find that a phonon-mediated odd-parity state may be realized in the presence of weak electronic correlations if the chirality-preserving electron-phonon vertices are much stronger than the chirality-flipping vertices.

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Introduction.—The discovery that gapped electronic systems can be topologically nontrivial has sparked enormous interest [1, 2]. While there now exists several clear examples of topological insulators, such as Bi$_2$Se$_3$ [3] and SnTe [4], the unconventional gap structure of topological superconductors make these systems much rarer [5]. Intriguingly, a superconducting state appears upon doping superconductors, making these systems much rarer [5]. In particular, the conditions under which the electron-phonon interaction can stabilize a triplet state remain unknown.

In this letter we study the fundamental question of whether an electron-phonon interaction can stabilize a triplet state, and thus evaluate the conditions required for the proposed topological superconductivity in Cu$_2$Bi$_2$Se$_3$. We first prove a theorem, showing that the symmetries of the electron-phonon vertex functions ensure that, purely with electron-phonon coupling, the critical temperature of the leading triplet state never exceeds that of the leading singlet. Therefore, the stabilization of the triplet state must depend on the so-called Coulomb pseudopotential, which is typically small. We then define a generalized chirality operator, which allows us to identify electron-phonon coupling vertices that would stabilize a triplet gap. Materials where chirality preserving vertices dominate could be candidates for electron-phonon mediated triplet superconductivity. Finally, we apply these insights to a model of Cu$_2$Bi$_2$Se$_3$ [7], and identify the electron-phonon vertices that cause an attractive interaction in the triplet channel. If these terms dominate the electron-phonon interaction, the topological state could be realized in the presence of weak correlations.

Electron-phonon interaction and pairing.—We start by considering a system with inversion ($I$) and time-reversal ($T$) symmetries, so that every eigenstate is at least doubly degenerate. Assuming for simplicity that a single band crosses the Fermi energy, we can index the degenerate states by a pseudospin variable $s = \pm$, such that $I |k, s\rangle = | -k, s\rangle$ and $T |k, s\rangle = s | -k, -s\rangle$. In the presence of strong spin-orbit coupling the electron-phonon interaction may not conserve pseudospin (in contrast to Ref. [22]), and so we have the general form

$$H_{e-p} = \sum_{k,k',s,s'} \sum_{\eta} g_{\eta}^s(k', \eta)(b_{k-k',\eta}^\dagger + b_{k'-k,\eta}) c_{k',s'}^\dagger c_{k,\eta},$$

(1)

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(1)
where $b_{q,s}$ is the annihilation operator for a phonon in mode $\eta$ with momentum $q$, and $c_{k,s}$ is the annihilation operator for an electron in state $|k, s\rangle$. The inversion and time-reversal symmetries require that the vertex functions satisfy $g_{\nu,s}^r(k', k) = \pm \eta g_{\nu,s}^r(-k', -k)$ and $g_{\nu,s}^\prime(k', k) = s s' g_{\nu,-s'}^l(-k', -k)$, respectively, where the sign $\pm \eta$ under inversion depends on the phonon mode.

Within the BCS approximation, the electron-phonon coupling generates the pairing interaction

\[
V_{s_2,s_1,s_3,s_4}(\chi', \chi) = -\sum_{\eta} g_{s_1,s_3}^0(k', k) g_{s_2,s_4}^0(-k', -k) \frac{\omega_{k' - \eta}}{\omega_{k' - \eta}} \times \Theta(\omega_D - |\epsilon_k|) \Theta(\omega_D - |\epsilon_k'|),
\]

where $\omega_{q,\eta}$ is the dispersion of phonon mode $\eta$, $\epsilon_k$ is the electronic dispersion, and $\omega_D$ is a cutoff on the order of the Debye energy. The pairing interaction is the kernel of the linearized BCS equation for the matrix gap function $\Delta(k)$, which is formulated as an eigenvalue problem

\[
\lambda \Delta_{s_1,s_2}(k') = -\sum_{k,s_3,s_4} V_{s_2,s_1,s_3,s_4}(k', k) \Delta_{s_3,s_4}(k).
\]

Only solutions with positive eigenvalues have a finite critical temperature, and the solution with the largest eigenvalue is the leading instability. Inversion symmetry limits physical solutions to either even-parity pseudospin singlet or odd-parity pseudospin triplet states.

**Singlet vs. triplet pairing.**—In the conventional case, i.e. in the absence of spin-orbit coupling, electron-phonon coupling is expected to lead to the singlet channel being dominant. Such a singlet pairing state is described by a gap function $\Delta^{(s)}(k) = f^{(s)}(k)(\hat{\sigma}^y)$, where $f^{(s)}(k)$ gives the momentum dependence of the pairing function. For the general electron-phonon interaction, the symmetries of the electron-phonon vertices yield a gap equation in the singlet channel of the form

\[
\chi^{(s)} f^{(s)}(k') = \sum_{k,s,\eta} \left| g_{s,s'}^{\nu}(k', k) \right|^2 + \left| g_{s,s'}^{\nu}(k', k) \right|^2 f^{(s)}(k),
\]

where the momenta are restricted to the shell of thickness $\omega_D$ about the Fermi surface. The singlet gap function is therefore an eigenstate of a matrix with nonnegative entries. It follows from the Perron-Frobenius theorem that the gap function $f^{(s)}(k)$ of the dominant instability has no sign changes as a function of the wavevector $k$, as is characteristic of conventional singlet pairing.

We now consider the triplet pairing function with the largest critical temperature, $\Delta^{(t)}(k)$. To compare with the singlet channel, we apply a momentum dependent pseudospin-rotation transformation so that it is recast in the form $\Delta^{(t)}(k) = \chi_k f^{(t)}(k) \hat{\sigma}^x$, where $f^{(t)}(k)$ and $\chi_k$ are the magnitude and sign of the triplet gap, respectively. In other words, we have rotated the pseudospin at $k$ and $-k$ so that in the new pseudospin basis the triplet pair formed from these states has vanishing $z$-component of pseudospin. Note that this rotation does not affect the singlet pairing, nor does it alter the symmetry properties of the electron-phonon vertices. The gap magnitude $f^{(t)}(k)$ satisfies the eigenvalue equation

\[
\chi^{(t)} f^{(t)}(k') = \sum_{k,s,\eta} \chi_k \chi_k' \frac{|g_{s,s'}^{\nu}(k', k)|^2 - |g_{s,s'}^{\nu}(k', k)|^2}{\omega_{k' - \eta}} f^{(t)}(k).
\]

The magnitude of the matrix elements in Eq. (5) are bounded by the corresponding elements in the singlet gap equation. By a corollary to the Perron-Frobenius theorem, the maximal eigenvalue of Eq. (5) therefore cannot exceed the maximal singlet eigenvalue. Since the leading triplet gap satisfies Eq. (5), we have our first major result which can be stated as the following theorem: in a system with inversion and time-reversal symmetry, the critical temperature of the leading triplet gap never exceeds that of the leading singlet gap for a purely phonon-mediated pairing interaction.

Our analysis implies that electronic correlations are vital to stabilizing a triplet state. In particular, the spatial separation of the electrons in a triplet Cooper pair reduces the pair-breaking effect of the Coulomb pseudopotential compared to a s-wave singlet state. A sufficiently large Coulomb pseudopotential may therefore reduce the critical temperature of the leading singlet state below that of the triplet. Such a strong Coulomb pseudopotential is the necessary condition for the triplet superconductivity to emerge in the system.

**Degenerate singlet and triplet states.**—While the singlet pairing typically may be expected to dominate over triplet pairing, it was pointed out in previous work by Fu and Berg, that the singlet and triplet states would be degenerate if the Dirac-like Hamiltonian considered by them commuted with a chirality operator. Motivated by this observation, we introduce a generalized “chirality” index $\nu$, that can be defined for any Hamiltonian for electrons at a wavevector $k$ as $\nu = \chi_k s$. Replacing the pseudospin indices in the gap equations by chirality indices, we obtain

\[
\chi^{(a)} f^{(a)}(k') = \sum_{k,s,\eta} \frac{|g_{s,s'}^{\nu}(k', k)|^2 + |g_{s,s'}^{\nu}(k', k)|^2}{\omega_{k' - \eta}} f^{(a)}(k),
\]

where the plus (minus) sign in the summand holds for $\alpha = s$ ($t$). Comparing the transformed equations in the singlet and triplet channels, it is clear that the singlet and triplet eigenvalues $\lambda^{(t)} = \lambda^{(s)}$ are degenerate if the electron-phonon vertices do not flip the chirality index i.e. $g_{s,s'}^{\nu}(k', k) = 0$. The chirality index associated with the fermions can be used to define a chirality operator $O_{\chi_k} = \sum_\nu f(k, \nu) [k, \nu]$. When only electron-phonon interactions which commute with $O_{\chi_k}$ are present, every singlet solution $\Delta^{(s)}(k)$ is degenerate with a triplet solution $\Delta^{(t)}(k) = U(k) \Delta^{(s)}(k) U(k)^\dagger$, where $U(k) = \chi_k f^{(t)}(k) \hat{\sigma}^x$. 


exp(iπΩ_{ch}(k)/4). On the other hand, an electron-phonon interaction which does not commute with the chirality operator is triplet pair-breaking. Therefore, triplet gaps are favored by electron-phonon couplings with the property that they preserve an appropriately defined chirality index that is even under time-reversal but odd under inversion, opposite to the pseudospin. This is the second major result of our letter.

We emphasize that it is not necessary to solve the gap equations to define the chirality index, as this only depends upon the sign structure of the triplet gap. This is very convenient, as it is common to approximate the exact solution of the gap equations by a simple function consistent with the point group. Given such a time-reversal-invariant triplet state, we can hence define a chirality operator which relates it to a singlet state with non-negative gap. The effective coupling constants for these two states, obtained by taking the inner product of the gap functions with the pairing interaction Eq. (2), are then degenerate if only electron-phonon vertices which preserve the chirality are present.

Application to Cu$_2$B$_2$Se$_3$.—The proposed odd-parity pairing state of Cu$_2$B$_2$Se$_3$ provides an excellent illustration of the preceding discussion. We start by introducing an effective Hamiltonian valid near the Fermi surface, where the electronic states are primarily derived from the Se $p_z$-orbitals at the top and bottom of the quintuple-layer unit cell. Denoting these two distinct sites by $s^z = \pm 1$, the low-energy spectrum is described by the $k \cdot p$ model [7]

$$H_0 = \sum_k \psi^\dagger(k) \left[ -\mu \hat{s}^0 \otimes \hat{\sigma}^0 + m \hat{s}^x \otimes \hat{\sigma}^0 + v_z k_z \hat{s}^y \otimes \hat{\sigma}^0 
+ \nu \left( k_x \hat{s}^z \otimes \hat{\sigma}^y - k_y \hat{s}^z \otimes \hat{\sigma}^x \right) \right] \psi(k).$$

Here $\psi(k) = (c_{k,1,\uparrow}, c_{k,1,\downarrow}, c_{k,-1,\uparrow}, c_{k,-1,\downarrow})^T$, where $c_{k,n,\sigma}$ destroys an electron with momentum $k$ and spin $\sigma$ at site $n$. The Pauli matrices in site and spin space are denoted by $\hat{s}^i$ and $\hat{\sigma}^i$, respectively. We consider the physical case where the chemical potential lies in the conduction band, i.e. $\mu > m$. The Hamiltonian is symmetric under inversion ($\mathbf{I} = \hat{s}^x \otimes \hat{\sigma}^0$) and time-reversal ($\mathbf{T} = i\hat{s}^y \otimes \hat{\sigma}^y \mathbf{K}$), and so the eigenstates of Eq. (7) can be labeled by a pseudospin [14].

The site degree of freedom allows odd-parity superconducting states in a relative $s$-wave, such as the $A_{1u}$ state $\Delta_{A_{1u}} i\hat{s}^y \otimes \hat{\sigma}^x$ proposed in Ref. [7]. As it opens an full gap on the Fermi surface [13], and has surface bound states consistent with point-contact spectroscopy measurements [12,13], it is one of the most promising candidates for a topological state in Cu$_2$B$_2$Se$_3$. We have seen, however, that the phonon-mediated pairing interaction generally favors an even-parity state with a full gap.

The simplest example of this is the topologically-trivial $A_{1g}$ state $\Delta_{A_{1g}} i\hat{s}^y \otimes \hat{\sigma}^x + \Delta'_{A_{1g}} i\hat{s}^z \otimes \hat{\sigma}^y$ [7].

In the absence of the mass term in Eq. (7), the Bogoliubov Hamiltonian for the $A_{1g}$ state with $\Delta'_{A_{1g}} = 0$ can be mapped into that for the $A_{1u}$ state by the unitary transformation $U = \exp(i\pi \hat{s}^y \otimes \hat{\sigma}^x/4)$. This immediately identifies the chirality operator as $\Omega_{ch}(k) = \hat{s}^y \otimes \hat{\sigma}^z$.

Let us now introduce the electron-phonon interaction

$$H_{e-p} = \sum_{k,k',\eta} \sum_{\mu,\nu} f_{\mu,\nu}(k',k) \left( b_{k-k',\eta}^\dagger b_{k',k,\eta} + b_{k',k,\eta}^\dagger b_{k-k',\eta} \right) \times \psi^\dagger(k') \hat{s}^\mu \otimes \hat{\sigma}^\nu \psi(k).$$

If only vertex functions $f_{\mu,\nu}(k',k)$ for which $\hat{s}^\mu \otimes \hat{\sigma}^\nu$ commutes with the chirality operator $\hat{s}^y \otimes \hat{\sigma}^z$ are nonzero, it follows from the discussion above that the coupling constants for the $A_{1u}$ and $A_{1g}$ states are identical. Vertex functions for which $\hat{s}^\mu \otimes \hat{\sigma}^\nu$ anticommutes with $\hat{s}^y \otimes \hat{\sigma}^z$ are generally expected to be present, however, giving the $A_{1g}$ state the higher coupling constant.

In the general case of a finite mass gap, the Fu and Berg $A_{1g}$ and $A_{1u}$ Hamiltonians cannot be mapped into one another by a chirality transformation. In the vicinity of the Fermi surface, however, we can define a chirality operator that relates the two gaps [24]. This is sufficiently close to the chirality operator in the massless limit that the classification of the electron-phonon vertices obtained above remains valid to good approximation. Specifically, the chirality-preserving electron-phonon vertices for the massless case are now either still chirality-preserving, or contain chirality-flipping terms which are smaller by a factor of $m/\mu \approx 0.3$ than the chirality-preserving [8]. A similar analysis holds for the vertices which flip the chirality in the $m = 0$ limit.

We make our discussion more concrete by considering a toy model where the electrons couple to a dispersionless optical mode with frequency $\omega_0$. From Eq. (5) we include only the $(\mu,\nu) = (0,0)$ and $(x,0)$ terms, representing chirality-preserving and flipping vertices, respectively. We assume that the corresponding vertex functions $g_0$ and $g_z$ are constant. The Fu and Berg $A_{1g}$ and $A_{1u}$ states are then exact eigenstates of the phonon-mediated pairing interaction, with eigenvalues $\lambda_{A_{1g}} = (g_0^2 + g_z^2 + 2|g_0 g_z| m/\omega_0)$ and $\lambda_{A_{1u}} = (g_0^2 - g_z^2)(1 - (m/\mu)^2)/\omega_0$, respectively. The $A_{1g}$ state is the leading instability for nonzero $g_z$ or $m$, while the $A_{1u}$ state only has finite critical temperature for $|g_z| < |g_0|$. We also include the on-site repulsion $H_{e-e} = U/V \sum_q \sum_{\eta,\mu} \rho_{s,\mu}(q) \rho_{s,\mu}(q)$ where $\rho_{s,\sigma}(q) = \sum_{k,k',q,s,\sigma} c_{k+q,s,\sigma}^\dagger c_{k,s,\sigma}$ and $V$ is the volume. As the first $A_{1g}$ gap $\Delta_{A_{1g}}$ involves on-site pairing, a finite $U > 0$ will tend to lower its critical temperature. On the other hand, the intersite $A_{1u}$ state is unaffected by $H_{e-e}$.

We study the pairing in our model within the mean-field approximation. For simplicity, the conductor band is assumed to extend from $-\mu - m$ below the Fermi surface to $W + \mu$ above, with constant density of states $\rho_0$ and $W \gg m$. Deriving the gap equations, we find that the
Following the notation of Ref. 7, the gap equations
are
\[ \nu \]
while for the on-site repulsion the
\[ \chi \]
and (b) nonzero mass gap. In the absence of
gap region N the system remains normal down to zero temperature.
We set \( W = 10\mu, \omega_D = 0.1\mu, \) and \( g_0/\omega_0 = 0.1225/\eta_0. \)

Critical temperature of the \( A_{1g} \) state satisfies

\[
\text{det} \begin{vmatrix}
\left( \frac{\omega_0^2 + g_0^2}{\omega_0} - \frac{U}{2} \right) \chi_0 - 1 & -\frac{U}{2} \chi & \frac{2g_0}{\omega_0} \chi_0^1 \\
-\frac{U}{2} \chi & -\frac{U}{2} \chi - 1 & 0 \\
\frac{2g_0}{\omega_0} \chi_0^1 & 0 & \frac{\omega_0^2 + g_0^2}{\omega_0} \chi_1 - 1
\end{vmatrix} = 0,
\]

while for the \( A_{1u} \) state we have to solve \( \lambda_{A_{1u}} - 1 = 1. \)

Following the notation of Ref. 7 the gap equations are expressed in terms of
\( \chi_0 = \nu_0 \int_{-\mu}^{\omega_D} \frac{d\epsilon}{\epsilon} \tanh(\epsilon/2k_B T_c)/\epsilon, \)
\( \chi_0^1 = (m/\mu)\chi_0, \chi_1 = (m/\mu)^2 \chi_0, \) and \( \chi = \nu_0 \int_{-\mu}^{\omega_D} \frac{d\epsilon}{\epsilon} \tanh(\epsilon/2k_B T_c)/\epsilon. \)
The resulting phase diagram is shown in Fig. (1) for the cases of (a) vanishing and (b) nonzero mass gap. In the absence of
on-site repulsion the \( A_{1g} \) state has higher critical temperature than the \( A_{1u}, \) except for \( m = g_x = 0 \) where the two are degenerate. Sufficiently strong on-site repulsion nevertheless suppresses the critical temperature of the \( A_{1g} \) state below that for the \( A_{1u}. \) For small ratios \( g_x/g_0 \lesssim 0.5, \) this requires only a relatively weak repulsion \( U \approx 0.1W. \) If \( g_x/g_0 \) is close to unity, however, a repulsive potential on the order of the bandwidth is necessary, and the critical temperature will be very small.

Since \( \text{Cu}_2\text{Bi}_2\text{Se}_3 \) is likely weakly-correlated, we conclude that the \( A_{1u} \) state could be realized if the chirality-preserving electron-phonon vertices are much larger than the chirality-flipping, which is the final major result our work. It is not obvious that this should be the case, however, and this problem requires detailed microscopic modeling beyond the present discussion. Interestingly, Wan and Savrasov have recently proposed that a strong phonon-modulated spin-orbit coupling is generic to layered semiconductors [10], although a nodal \( A_{2u} \) state then has highest eigenvalue in the triplet channel.

Summary.—In this letter we have shown that the leading instability of a phonon-mediated pairing interaction can be a triplet state, but this must be degenerate with a singlet solution. Our analysis relies only on the symmetries of the electron-phonon vertex functions. We have additionally formulated a condition in terms of a chirality operator for when this degeneracy holds. We have hence identified the electron-phonon vertices that produce an attractive interaction for the topological state proposed for \( \text{Cu}_2\text{Bi}_2\text{Se}_3, \) and which are pair-breaking. If the former dominate the latter, we show that weak electronic correlations could stabilize the odd-parity state. Large-scale (and quantitatively accurate) first principles calculations can in principle determine whether specific systems (e.g. \( \text{Cu}_2\text{Bi}_2\text{Se}_3, \) \( \text{Sn}_{1-x}\text{In}_x \text{Te}, \) etc.) satisfy the necessary theoretical constraints derived in our work, providing a route to the realization of topological superconductivity in ordinary electronic materials.

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