Topological superconductivity facilitated by exchange interaction on the surface of FeTe_{0.5}Se_{0.5}

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Abstract
FeTe_{1-x}Se_x is a family of iron-based superconductors with a well-known critical temperature (T_c) of 14.5 K for x = 0.45. Also well-established is the presence of topological surface states of FeTe_{0.5}Se_{0.5}, in which topological superconductivity sets in. By using density functional calculations and the Bardeen–Cooper–Schrieffer (BCS) theory for traditional superconductors we calculated the $T_c$ of the surface layers of FeTe_{0.5}Se_{0.5} with the exchange interaction as the source of attractive force to form the Cooper pairs. The estimated $T_c$ is in reasonable agreement with the experimental value and suggests that the exchange interaction is behind the topological superconductivity on the surface of FeTe_{0.5}Se_{0.5}.

1. Introduction

Research on FeTe_{1-x}Se_x [1–16] with x around 0.5 has made considerable progress in the field of topological superconductivity and Majorana zero modes [17–19], the latter being associated with possible application in quantum computing. Experiment [1] using angle-resolved photoelectron spectroscopy (ARPES) with spin resolution has indicated that on the (001) surface of FeTe_{0.5}Se_{0.5} a Dirac cone is formed within the band gap of the projected bulk states and the direction of electron spin associated with each surface state is locked to its crystal momentum k. Calculations [1, 13] have also shown that the bulk electronic structure of FeTe_{0.5}Se_{0.5}, whose smaller primitive cell is more convenient for theoretical analysis, possesses a $Z_2$ invariant equal to one and is topologically nontrivial. Furthermore, the experiment is also able to detect an s-wave superconducting gap from the surface states below a $T_c$ of 14.5 K, providing clear evidence for the existence of topological superconductivity. However, the topological superconductivity exists only within the surface layers. Bulk electrons are not spin-polarized and their superconducting pairing is topologically trivial [1].

Spin fluctuation has been proposed as a possible mechanism behind the superconductivity associated with iron-based superconductors [14]. FeTe_{0.5}Se_{0.5} in particular has an isotropic nodeless superconducting gap according to the measurements of ARPES and its electron-pairing is strongly linked to antiferromagnetic exchange interaction [15]. A theoretical proof of the superconducting mechanism on the surface of FeTe_{0.5}Se_{0.5} would demand the ab initio values of the exchange interaction between electrons and other relevant parameters necessary for the construction of Bardeen–Cooper–Schrieffer (BCS) model to generate reasonable superconducting gap and critical temperature.

2. Calculation of the bulk and surface states of FeTe_{0.5}Se_{0.5}

In our derivation of the bulk electronic structure of FeTe_{0.5}Se_{0.5} we used lattice constants and chalcogen positions from the experimental data [16]. Density functional calculation based on the VASP code [20, 21] was then performed with general gradient approximation (GGA) [22] chosen as exchange–correlation functional. A larger unit cell consisting of four Fe atoms and two atoms for each of the Te and Se elements, as shown in figure 1(a), was adopted to better represent the mingled configuration of Se and Te atoms. Energy bands along major symmetry directions are plotted in figures 1(b) and (c) to separate contributions
Figure 1. (a) Unit cell consisting of four Fe, two Te, and two Se atoms in the calculation of band structure of FeTe$_{0.5}$Se$_{0.5}$. (b) Energy bands of FeTe$_{0.5}$Se$_{0.5}$ along symmetry directions, with the zoomed-in view of the (blue) rectangle showing an energy gap of 8.90 meV. Red and empty circles represent relative proportions of Te-p and Se-p orbitals respectively. (c) The same energy bands as in (b) showing relative proportions of Fe-d and Fe-p orbitals.

from different orbitals for the same band structure. A small gap of 8.90 meV can be found along the Γ–Z or (001) direction as is shown in the zoomed-in part of the bands within the blue rectangle in figure 1(b). The calculated energy gap is opened by spin–orbit interaction of the electrons and its size is close to the value of previous calculations [13]. It is clear that the contribution from Te p orbitals (red circles) is much greater than that of the Se p orbitals (empty circles), and $p_z$ is the dominant partial wave from either Te or Se. But d orbitals (consisting mostly of $d_{xy}$) from Fe are also present conspicuously in major parts of the valence and conduction bands, as indicated by red circles in figure 1(c). A band inversion, however, occurs around the band gap. The calculated band structure as a whole is very consistent with previous works [1, 13].

We went on to calculate the (001) surface states of FeTe$_{0.5}$Se$_{0.5}$ by using the maximally localized Wannier functions [23] with tight-binding parameters extracted from the bulk calculation. A total of five d orbitals from Fe and three p orbitals from each of Se and Te was included for each spin in the construction of the
Figure 2. Topological surface states calculated for 67 layers of FeTe$_{0.5}$Se$_{0.5}$ are separated from the projection of bulk states.

Figure 3. Spin texture of seven layers of FeTe$_{0.5}$Se$_{0.5}$ is presented in the energy–momentum topography within an energy window to the right. Each arrow represents the direction of electron spin associated with a $k$ point in the $k_x$–$k_y$ plane.

Hamiltonian. Surface states corresponding to a semi-infinite structure was simulated by including numerous stacked layers of FeTe$_{0.5}$Se$_{0.5}$ in the calculation. Figure 2 displays the result for a structure consisting of 67 layers of FeTe$_{0.5}$Se$_{0.5}$, in which a Dirac cone located in the gap of bulk energy bands (painted gray partially) is readily identified. The two crossing bands, each carrying a spin opposite to the spin of another band with opposite crystal momentum (not shown), are clearly separated from all projections of bulk states onto the (001) surface as required of topological surface states. A film of seven layers, however, is adequate for the accommodation of surface effects and overall convergence into the bulk properties. Analysis of its spin texture is illustrated in figure 3 by contours of equi-energy curves of $k$ points surrounding the $\Gamma$ point in the $k_x$–$k_y$ plane. It indicates that the spin carried by the electron at $k$ is opposite in direction to the spin of the electron at $-k$, making backscattering of electrons impossible without the flip of spin. This helical-spin texture further supports the topological nature of the surface states associated with the thin film.

3. BCS model and exchange interaction

For topological superconductivity to occur an attractive interaction must exist between pairs of electrons in the layers close to the surface or in a thin film. As is normally practiced in the BCS theory the Hamiltonian $H$ contains two major components given by

$$ H = \sum_{k,m,s} E_k a_{k,m,s}^+ a_{k,m,s} - V \sum_{k,k'} a_{k,s}^+ d_{-k,k'} d_{k,s}^+ a_{k',s}, $$

(1)
where $a^+_{k,m}$ ($a_{k,m}$) is the creation (annihilation) operator of a quasi-particle (electron) with crystal momentum $k$ and spin $m$ (↑ for up and ↓ for down) and $\varepsilon_k$ is the associated one-particle energy, which can be approximated by the eigenvalues derived from density functional calculation. The second term in the case of FeTe$_{0.5}$Se$_{0.5}$, however, includes the (screened) exchange interaction between two electrons forming a Cooper pair. The negative and uniform coupling of $-V$ is adopted to represent the attractive interaction and the s-wave nature of superconductivity.

Exchange energy between two electrons is the difference of the energies of their singlet and triplet states as expressed in the equation:

$$E_s - E_t = 2\int\int_{\varepsilon\text{GGA}}^{} e^2 |\mathbf{r} - \mathbf{r}'| \psi_k(\mathbf{r})\psi_k(\mathbf{r})\psi_k^\dagger(\mathbf{r}')\psi_k^\dagger(\mathbf{r}')d^3\mathbf{r}d^3\mathbf{r}' = -2V_{kk'}$$ \hspace{1cm} (2)

where $E_s$ ($E_t$) is the total energy of the two electrons under singlet (triplet) state and $\psi_k(\mathbf{r})$ is the Bloch wave function for an electron in one single-particle state associated with crystal momentum $k$, $e$ being the absolute value of the electron charge. In actual calculation, the Coulomb interaction $e^2/|\mathbf{r} - \mathbf{r}'|$ is also subject to many-electron screening and the GGA exchange–correlation functional is used.

To derive the average coupling $-V$ in equation (1), total energies of a unit cell having antiferromagnetic and ferromagnetic Fe atoms are calculated separately, which requires the integration of $V_{kk'}$ in equation (2) over the first Brillouin zone. The difference between the two total energies divided by the number of contributing pairs of electrons is the uniform $-V$. As a result, interaction between electrons separated by nearest neighbors of Fe is calculated to be 11.8 meV by using the primitive cell. Electrons separated by the distance between second-nearest neighbors of Fe has a slightly higher exchange interaction of 11.9 meV, which is extracted from the calculation of a larger unit cell containing twice the number of atoms in the primitive cell. For further separation of electrons the exchange interaction is greatly reduced. $V$ for distance between two third-nearest neighbors, for example, is only 1.33 meV due to the presence of another Fe atom halfway between, which strongly shields the Coulomb interaction.

The attractive exchange interaction is mediated by phonons which carry momenta equal to the difference of electron momenta $k$ and $k'$ in equation (1). By calculating the phonon dispersion relations of FeTe$_{0.5}$Se$_{0.5}$ one is able to obtain parameters accounting for electron–phonon interaction in the BSC model. Phonon dispersion curves calculated by VASP for FeTe$_{0.5}$Se$_{0.5}$ are shown along major symmetry directions in figure 4 along with the total density of states (DOS) and local density of states (LDOS) for each of the Fe, Te, and Se atoms, showing important contribution from Te. There are three acoustic and 21 optical phonon bands corresponding to a unit cell consisting of eight atoms, four for Fe and two for Te and Se each. The three acoustic phonon bands along various directions provide an average phonon speed of 2089 m s$^{-1}$, which is then used in the construction of Debye model. The Debye wave vector $k_D$ has the magnitude of 1.40 Å$^{-1}$ derived from the formula $k_D = (6\pi^2 n)^{1/3}$, where $n$ is the number of atoms per unit volume. Accordingly, the Debye frequency $\omega_D$ and the associated phonon energy are $2.92 \times 10^{13}$ s$^{-1}$ and 19.3 meV respectively.

Figure 4. Phonon dispersion curves of FeTe$_{0.5}$Se$_{0.5}$ along high-symmetry directions and the corresponding total DOS and LDOS from each of the constituents. The bands are derived from a unit cell of eight atoms.
4. Superconducting gap and critical temperature

In equation (1) an approximation is made so that the interaction is limited to electrons within the energy range between $\mu - \hbar \omega_D$ and $\mu + \hbar \omega_D$, where $\mu$ is the chemical potential. The BCS energy gap at 0 K denoted by $\Delta(0)$ is given by $2\hbar \omega_D e^{-1/\lambda}$, where $\lambda$ is the strength of electron–phonon coupling and is the product of $V$ multiplied by $N(\epsilon_F)$, the electronic density of states per unit volume for a single spin at the Fermi level. Using the seven-layer film of FeTe$_{0.5}$Se$_{0.5}$, which contains the effect associated with topological surface states, $N(\epsilon_F)$ can be calculated from the electronic bands. It turns out to be 30.44 per unit cell for a single spin, as is shown in figure 5. By taking either the exchange interaction for the nearest neighbors or interaction for second nearest neighbors, the value of $\lambda$ is found to be around 0.36 and is generally not considered within the range of weak coupling for superconductivity [12, 15]. $\Delta(0)$ is then calculated and the result is close to 2.4 meV, a value compared favorably with the experiments [1, 12].

Critical temperature $T_c$ for FeTe$_{0.5}$Se$_{0.5}$ is calculated directly from the formula $k_B T_c = 1.13 \hbar \omega_D e^{-1/\lambda}$, where $k_B$ is the Boltzmann’s constant. Using the higher value for $V$ (11.9 meV) leads also to a higher $T_c$ of 16.0 K, as compared to the 15.6 K for the slightly lower interaction of 11.8 meV. In either case the calculated $T_c$ is reasonably close to that of FeTe$_{0.55}$Se$_{0.45}$. It is also evident that the exponential dependence of the parameter $\lambda$ makes $T_c$ highly sensitive to even a small variation of the exchange interaction and/or density of states.

5. Conclusions

Our investigation indicates that antiferromagnetic exchange interaction is behind the topological superconductivity on the surface of FeTe$_{0.5}$Se$_{0.5}$. By using both density functional calculations and maximally localized Wannier functions topological surface states and strength of the interaction can be derived, which are subsequently employed in the s-wave BCS model to produce reasonable superconducting gap and critical temperature. The results also point to a possibility that exchange interaction is a major factor in forming Cooper pairs of electrons in many iron-based superconductors and beyond.

Acknowledgments

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