First-principles calculations of spin-triplet andreev reflection spectra at half-metallic ferromagnet/superconductor interface

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Combining the first-principles noncollinear calculations of scattering matrices with Andreev approximation, we investigated the spin-triplet Andreev reflection (AR) spectra for the interface between half-metallic ferromagnet Co$_2$MnSi and s-wave BCS superconductor Al with and without interfacial roughness, where the orientations of magnetic moments near the interface are randomly distributed. The calculated results show that the AR spectra have peak structures near zero bias for the clean interface with relative weak magnetic disorder. With increasing the degree of interfacial roughness or magnetic disorder, these subgap peaks of conductance spectra will be washed out. The results also show that the value of subgap conductance spectrum can be raised significantly by the magnetic disorder. Finally, our calculations reveal that the long-range spin-triplet AR in Co$_2$MnSi/Al(001) interface can be enhanced by a small amount of interfacial roughness.

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I. INTRODUCTION

In normal-metal/s-wave BCS superconductor heterostructure (NM/S), below the superconducting energy gap a spin up or down electron comes into NM/S interface will be reflected as a hole in the opposite spin subband, which is called spin-singlet Andreev reflection (AR) process, and this phenomenon has been well studied by the first-principles approach very recently. If the NM is ferromagnet (F), the number of propagating states at the Fermi energy is different for majority and minority spin owing to the presence of exchange splitting. Therefore only a fraction of incident electron can be Andreev reflected and then the conductance of this spin-singlet AR is suppressed significantly. The most extreme case is the half-metallic ferromagnet (HF) with only one spin subband at Fermi level, thus the spin-singlet AR process will be totally prohibited in HF/S interface.

However, since the striking observation of Josephson current in S/HF/S junction has been reported, the spin-triplet AR has attracted a great deal of attention in recent years. It is because that the exotic phenomena such as 0–π transitions in S/HF/S Josephson junction may have some potentially applications on superconducting quantum computing circuits. At HF/S interface, the spin-triplet AR means that the incident electrons undergo a spin-flip scattering and penetrate into BCS superconductor as Cooper pair, leaving a phase coherence hole in the same spin which can be survived at HF side. So the incident electrons and phase-coherently reflected holes (or vice versa) within the same spin subband can induce the spin-triplet superconducting correlation in HF. Further, these spin-triplet pairing is immune to the exchange splitting in the ferromagnet, which results in the long-range proximity effect as well as the supercurrent across the S/HF/S heterostructure. In addition to all this, the spin-triplet AR induced by spin-flip scattering at HF/S also has been proposed to explain the experimental spin polarization (P) of HF deviated from its theoretical prediction P=100%.

Up to now, the theoretical calculations about spin-triplet AR generally deal with the interface properties by a simple delta-function potential barrier or scattering matrices coefficients with model parameters, where the complex Fermi surfaces and electronic band structure of real materials have not been considered yet. For realistic HF/S interface, it is argued that multichannel scattering theory of propagating states at Fermi surface should be taken into account for calculating AR conductance. Moreover, the symmetry of band states when matching wave-function between two bulk crystal has effect on transport properties of the interface, which can be large and should not be neglected. Although the first-principles method has been applied to AR of Fe/Al interface with spin-flip it is difficult to distinguish the spin-singlet and spin-triplet AR in normal F (with incomplete spin polarization), while in full HF there is only spin-triplet AR. Therefore, the main purpose of this paper is to perform the first-principles density functional theory calculation without introducing any arbitrary parameters to study the spin-triplet AR conductance spectra in HF/S.

On the other hand, the well-known Heusler alloy Co$_2$MnSi belongs to HF and can be grown by sputtering techniques. It also can be well matched in the (001) direction with s-wave BCS superconductor Al by rotating 45°, which makes the Co$_2$MnSi/Al(001) interface a good candidate for studying spin-triplet AR in details.

In this paper, we use the method which combined the scattering matrix approach with Andreev approximation in the frame of Bogoliubov-de Gennes equations to evaluate the spin-triplet AR conductance in Co$_2$MnSi/Al(001) interface with and without interfacial roughness. The spin-flip scattering process at HF/S required by spin-triplet AR is introduced by the disordered distribution of interfacial magnetic moments direction. Here the transmission and reflection matrices of magnetic noncollinear textured HF/S interface are calculated by first-principles scattering wave-function match-
II. METHOD

In order to investigate AR within scattering framework, following Beenakker’s model we consider a general F/S junction with inserting a fictitious region N between F and S, which is composed of S in its normal state. The F/S interface with ideal F as left lead and ideal N as right lead constitute a well-defined scattering problem. Meanwhile, there is only AR at N/S interface owing to no potential difference at this interface. The sketch of the whole scattering problem including AR is shown in Fig. 1(a).

At the F/N interface, the incoming and outgoing scattering state coefficients for electrons and holes quasiparticles at excitation energy \( \varepsilon \) can be written as:

\[
\begin{pmatrix}
C_e^{-}(1) \\
C_o^{-}(1) \\
C_o^{+}(1) \\
C_n^{-}(1) \\
C_n^{+}(1) \\
C_n^{+}(2) \\
C_o^{+}(2) \\
C_o^{-}(2)
\end{pmatrix} =
\begin{pmatrix}
r_{11}^{c}\varepsilon(\varepsilon) & t_{12}^{c}\varepsilon(\varepsilon) & 0 & 0 \\
0 & r_{21}^{c}\varepsilon(\varepsilon) & t_{22}^{c}\varepsilon(\varepsilon) & 0 \\
0 & 0 & r_{11}^{h}\varepsilon(\varepsilon) & t_{12}^{h}\varepsilon(\varepsilon) \\
0 & 0 & t_{21}^{h}\varepsilon(\varepsilon) & r_{22}^{h}\varepsilon(\varepsilon)
\end{pmatrix}
\begin{pmatrix}
C_e^{+}(1) \\
C_o^{+}(2) \\
C_o^{+}(1) \\
C_n^{+}(1) \\
C_n^{+}(2) \\
C_n^{-}(2) \\
C_o^{-}(2) \\
C_o^{-}(1)
\end{pmatrix}
\]

where subscript 1 refers to F, to N and + (−) denotes right or left going waves as shown in Fig. 1(a). The coefficient vectors \( C_{e(h)}^{+} (i) \) \((i = 1, 2)\) are amplitudes of propagating Bloch states of electron (hole) in the left lead F and right lead N. Here \( r_{ij}^{e(h)}(\varepsilon) \) and \( t_{ij}^{e(h)}(\varepsilon) \) \((i, j = 1, 2)\) is reflection and transmission matrix of electron (hole) for F/N interface which is 2×2 block matrix in spin space. In our calculation the spin-flip scattering process originating from noncollinear magnetic moments distribution at F/N interface is just described by the off-diagonal scattering matrix elements in this spin space.

Moreover, these normal-state reflection and transmission matrix can be calculated by a two-steps procedure. First the effective single-electron potential for collinear F/N interface that obtained by density functional theory will serve as input to the second step. Secondly, in the rigid potential approximation the noncollinear transport coefficients can be evaluated using wave-function matching method.

Considering the right and left going quasiparticles in N region is connected by AR at N/S interface, the reflection matrix for the whole F/N/S system can be obtained by solving the above scattering equation Eq. (1). According to Beenakker’s model we introduce Andreev approximation at N/S interface, i.e., \( C_o^{-}(1) = \alpha C_n^{+}(1)e^{i\phi} \) and \( C_n^{+}(1) = \alpha C_o^{-}(1)e^{-i\phi} \), where \( \phi \) is phase of superconductor. The factor \( \alpha = \exp[-i\arccos(\varepsilon/\Delta_0)] \) for \( |\varepsilon| < \Delta_0 \) and \( \alpha = |\varepsilon - \text{sgn}(\varepsilon)\sqrt{\varepsilon^2 - \Delta_0^2}/\Delta_0 | \) for \( |\varepsilon| > \Delta_0 \), where \( \Delta_0 \) is s-wave superconductor pair potential at zero temperature. Therefore substituting the formula of Andreev approximation into Eq. (1), we can obtain the relationship of the incoming and reflected states only at F region, which determines the reflection matrix of whole F/N/S system. Namely,

\[
R_{ee} = r_{11}^{c}\varepsilon(\varepsilon) + \alpha^2 r_{12}^{c}\varepsilon(\varepsilon)r_{22}^{h}\varepsilon(\varepsilon)\left[1 - \alpha^2 r_{22}^{c}\varepsilon(\varepsilon)r_{22}^{h}\varepsilon(\varepsilon)\right]^{-1}t_{21}^{c}\varepsilon(\varepsilon)
\]

\[
R_{he} = \alpha e^{-i\phi}r_{12}^{h}\varepsilon(\varepsilon)\left[1 - \alpha^2 r_{22}^{c}\varepsilon(\varepsilon)r_{22}^{h}\varepsilon(\varepsilon)\right]^{-1}t_{21}^{c}\varepsilon(\varepsilon)
\]

So in the linear-response regime, the total conductance of F/S is

\[
G_{FS}(\varepsilon) = \frac{e^2}{h} \text{Tr} \left( 1 - R_{ee}R_{ee}^\dagger + R_{he}R_{he}^\dagger \right)
\]

and we define the spin-triplet AR conductance is

\[
G_{\text{triplet}}(\varepsilon) \equiv \frac{e^2}{h} \text{Tr} (R_{he}^\dagger R_{he}^\dagger + R_{he}^\dagger R_{he}^\dagger)
\]

In order to study the character of subgap AR conductance spectrum, here we define the normalized conductance \( g(V) \equiv |G_{FS}(V) - G_{FN}(0)|/G_{FN}(0) \), where \( G_{FN}(0) \)
is conductance when the superconductor in its normal-state and $V$ is normalized bias defined as $V \equiv \varepsilon / \Delta_0$. Here the zero temperature pair potential is very small, e.g., $\Delta_0(\text{Al}) \approx 0.34$ meV.\textsuperscript{29} The difference of electronic structure within this tiny energy region can be ignored, so in our calculations the reflection and transmission matrices at excitation energy $\varepsilon$ in Eq. (2) and (3) is approximately equal to the conductance at Fermi level.

### III. COMPUTATIONAL DETAILS

Co$_2$MnSi is a representative compound of Heusler alloys, where the Co atoms form a simple cubic lattice while the Mn and Si atoms are situated the alternate body center positions. The experimental lattice constant of bulk Co$_2$MnSi is 10.68 [a.u.]\textsuperscript{30} and the $s$-wave superconductor Al has fcc crystal structure with lattice constant 7.653 [a.u.].\textsuperscript{29} So the fcc Al and Co$_2$MnSi lattice can be matched in (001) direction, by rotating 45° around (001) axis for fcc Al lattice. The mismatch of this heterostructure is about 1.3% and we keep the lattice constant of bulk Co$_2$MnSi unchanged and slightly compress the bulk Al lattice.

In our calculations, the atom sphere approximation (ASA)\textsuperscript{32} is taken into account for obtaining electronic structure and conductance. These radius of spheres are determined by the crystal Hartree potential\textsuperscript{32} with filling up all the space. The atom spheres of the Co$_2$MnSi are chosen as $r$(Co) = 2.576 [a.u.] and $r$(Mn) = $r$(Si) = 2.681 [a.u.]. Similarly, the radius of Al atom sphere is chosen as $r$(Al) = 2.951 [a.u.]. Due to the CoCo monolayer and MnSi monolayer stack alternately in the (001) direction, the Co$_2$MnSi(001) has two kinds of terminations (CoCo and MnSi) adjacent to Al lead. The distance of these two ideal CoCo/Al and MnSi/Al interfaces can be determined as follows. From Fig. 1(b), one can see that the interfacial cubic unit cell consists of 0.5 Co (0.25Mn, 0.25Si) and 0.5 Al, where these Co (MnSi) and Al should fill up the space of the cubic with volume $V = d x (a_{\text{Co}_2\text{MnSi}}/2)^2$. Since we have chosen the radius of Co (MnSi) and Al, the interface distance between CoCo (MnSi) and Al monolayer $d$ can be determined by the above formula. In addition, the other distance for Al-Al and Co-MnSi layers still maintain their bulk values.

The structural model for alloyed rough interface of CoCo/Al and MnSi/Al are constructed by a 5 × 5 lateral supercell which contains 50 interfacial Co atoms in CoCo/Al and 25 interfacial Mn atoms in MnSi/Al, where the lattice has no distortion but the interfacial atoms occupy the sites randomly and the possibility of occurrence is according to their concentrations $x$. As shown in Fig. 1(c), here we assume that Co (MnSi) and Al atoms diffuse into each other for one monolayer at interface, which can be denoted as $[\text{Co}_{1-x}\text{Al}_x]_2[\text{Co}_2\text{Al}_{1-x}]$ or $[(\text{MnSi})_{1-x}\text{Al}_x](\text{MnSi})_2\text{Al}_{1-x}$. For MnSi/Al interface we also assumed that the Mn and Si have the same chance to diffuse into Al monolayer. Furthermore, in our rough interface model the numbers of interfacial Co (MnSi) and Al atoms are equal to that of clean interface. Hence, for filling the interfacial space, we take the distance between CoCo (MnSi) and Al monolayer of rough interface as same as the values of clean interface.

In this paper, the effective single-electron potential of Co$_2$MnSi/Al(001) interface is obtained from tight-binding linearized muffin-tin-orbital (TB-LMTO) surface Green function method with ASA approximation.\textsuperscript{23–25} For rough interface, the coherent potential approximation (CPA)\textsuperscript{15,16,27} is used to deal with the alloyed interfacial roughness. After performing self-consistent electronic structure calculation, we put the atomic sphere potentials into the corresponding sites for evaluating the AR conductance. Here the exchange-correlation potential is taken within local spin density approximation (LSDA) using von Barth-Hedin parametrization.\textsuperscript{38} The electron is treated scalar-relativistically and the cutoff of orbital angular momentum of basis is $l_{\text{max}} = 2$, corresponding to $spd$-basis. The reciprocal lattice vectors of Brillouin zone (BZ) are each divided into NK=12 intervals for calculating the self-consistent electronic structure of interface. Meanwhile, the calculations of AR conductance are performed with a $k_{\parallel}$-mesh density equivalent to 2500 $k_{\parallel}$-mesh points in the 2D BZ of 1 × 1 lateral unit cell.

Here we assume that the spin-flip scattering, which is necessary for spin-triplet AR in full HF, only originates from the noncollinear random distribution of interfacial magnetic moments direction. Therefore the magnetic disorder in interfacial magnetic atoms is also simulated by 5 × 5 lateral supercell as the structural model for rough interface. For these interfacial magnetic atoms, the deviation angle between the local magnetic moment direction and the global quantum axis satisfy Gaussian random distribution, where the distribution width is $\Delta \theta$. In addition, the rigid potential approximation\textsuperscript{39} has been employed here and the spin-orbit coupling is neglected in our noncollinear calculations.

### IV. RESULTS AND DISCUSSION

In this section, we first examine the self-consistent calculated electronic structure of bulk Co$_2$MnSi as left lead and Co$_2$MnSi/Al(001) interface respectively. The resulting band structure along $\Gamma$–X direction and density of states (DOS) for bulk Co$_2$MnSi are shown in Fig. 2. The shape of DOS agrees well with the previous results reported in Ref.\textsuperscript{29}. The majority band is totally metallic and there is a semiconductor energy gap ($\sim 0.4$ eV in agreement with Ref.\textsuperscript{29}) in minority spin. The Fermi level is inside the minority spin gap near the conduction band minimum, which indicates that the bulk Co$_2$MnSi is a full half-metallic ferromagnet and the current injected into Co$_2$MnSi/Al(001) interface from left lead has 100% spin polarization. Further, Fig. 3 shows the layer-resolved magnetic moments for CoCo- and MnSi-
FIG. 2: The electronic structure of bulk Co$_2$MnSi. (a) and (c) The energy along Γ-X direction for majority and minority channel respectively. (b) The spin-resolved density of states (per formula unit).

FIG. 3: The layer-resolved magnetic moments for (a) CoCo- and (b) MnSi-terminated Co$_2$MnSi/Al(001) interfaces without interfacial roughness. The magnetic moments of Co or Mn atoms have been suppressed at the interface and recover to their bulk values away about 4 [ML] from interface.

Next we concentrate on the conductance spectra of Co$_2$MnSi/Al(001) interface without interfacial roughness. It can be seen that the interfacial magnetic moments are suppressed due to the charge transfer between Co (Mn) and Al atoms. However, the magnetic moments of Co and Mn atoms can recover to their bulk values just about 4 [ML] away from interface. In addition, the results for rough Co$_2$MnSi/Al(001) interface is similar to that of clean interface and have not been demonstrated here for simplicity.

Next we concentrate on the conductance spectra of Co$_2$MnSi/Al(001) interface for different interfacial magnetic disorder (Δθ). The interfacial alloy concentration of rough interface is 50%-50% (x = 0.5). For clean interface with Δθ < 60°, it is clear that the peak structure emerges near the zero bias. However, the larger amount of magnetic disorder (Δθ > 60°) or interfacial roughness can wash out this subgap conductance structure.

FIG. 4: The normalized conductance spectra for different degree of interfacial magnetic disorder (Δθ). The interfacial alloy concentration of rough interface is 50%-50% (x = 0.5). For clean interface with Δθ < 60°, it is clear that the peak structure emerges near the zero bias. However, the larger amount of magnetic disorder (Δθ > 60°) or interfacial roughness can wash out this subgap conductance structure.

conductance spectra of Co$_2$MnSi/Al(001) interface for different Δθ. In the case of Δθ = 0° the conductance below the gap is exactly zero because the left lead Co$_2$MnSi only has majority spin band at the Fermi energy and the AR process is prohibited without spin-flip. Once there is spin-flip scattering created by the noncollinear random distribution of interfacial magnetic moments direction, a conductance peak structure will emerge gradually at zero bias as shown in Fig. 4(a) and (c). This zero bias conductance peak (ZBCP) has the largest relative height when Δθ ≈ 30° and will convert into a small zero bias conductance dip (ZBCD) with increasing Δθ. The presence of ZBCP in our calculation can be attributed to the Fermi wave-vector mismatch between Co$_2$MnSi and Al combined with high spin polarization in ferromagnet, which has been predicted by the extend BTK theory in Ref. 41. For Δθ > 60° the above-mentioned ZBCP or ZBCD structure have both vanished and the subgap conductance becomes V shape. Moreover, Fig. 4(b) and (d) shows the conductance spectra of rough Co$_2$MnSi/Al(001) with 50%-50% interfacial alloy concentration (x = 0.5). One can observe that the interfacial alloy roughness can also totally wash out the ZBCP structure for all Δθ. Considering that these subgap conductance resonance peaks is sensitive to the phase of reflection coefficients, so the disappearance of peak structure can be attributed to the random phase shift gained at Co$_2$MnSi/Al(001) interface.

Now turning our attention to the value of normalized conductance below the gap, we can see that it increases with increasing Δθ as shown in Fig. 4. In general, the extend BTK theory is employed to fit this conduc-
It is lack of detailed information about the magnetic disorder in Co$_2$MnSi/Al(001), the distribution width $\Delta \theta$ in ferromagnet/nonmagnetic metal interface can be up to $40 \sim 70^\circ$ according to first-principles calculations.$^{25,53}$ Therefore, our results imply that the interfacial magnetic disorder in point contact AR experiments can explain the extracted $P$ deviated from theoretical value $P=100\%$.

Finally, in order to study the dependence of spin-triplet AR on interfacial alloy concentration, we have calculated the spin-triplet AR using Eq. (5) with concentration from $x = 0.1$ to 0.5. Fig. 5 shows the spin-triplet AR conductance spectra with $\Delta \theta = 30^\circ$ for different $x$. When the concentration $x < 0.2$ the AR spectrum is similar to that of clean interface ($x=0$) and once $x$ is greater than $0.2$ the conductance peak structure disappeared owing to the random of phase shift at Co$_2$MnSi/Al(001) interface.

Fig. 6 shows the zero bias spin-triplet AR conductance $G_{\text{triplet}}(0)$ and normal-state conductance $G_{\text{FN}}(0)$ at zero bias as function of $x$. In order to check the statistical error, the results given for different randomly generated disorder configuration are also shown in Fig. 6. One can see that the variation of sample to sample is larger for spin-triplet AR ($\sim \pm 20\%$) than for normal-state conductance ($\sim \pm 5\%$) due to the AR is more sensitive to the fluctuation of interfacial phase shift, It is noted that the expression of AR coefficient in Eq. (3) has the form like multiple beam interference, suggesting the AR conductance is more sensitive to the amplitude of normal-state scattering coefficients and phase coherence of the interface. As a consequence, the spread of spin-triplet AR is more pronounced than that of normal-state conductance.
Here as a coarse qualitative examination, next we only consider the average values of the calculated conductances. It can be seen that as the interfacial Al and Co (MnSi) atoms mixing with each other, the spin-triplet AR will be enhanced and reach maximum around $x \approx 0.2$. When the interfacial alloy concentration $x$ increases up to 0.5, the spin-triplet AR will go back to be around the value of clean interface. Further, in low concentration $x$ the interfacial alloyed atoms can be regarded as the impurity at interface, such as Al atoms in CoCo (MnSi) monolayer. Hence, our results of AR conductance in Co$_2$MnSi/Al(001) demonstrate that the impurity at interface can assist the spin-triplet AR process based on first-principles calculations, which agrees with the prediction in the recent theoretic investigation by model Hamiltonian.

From Eq. (3) and (5) one can see that the spin-triplet AR conductance is evaluated by normal-state transmission and reflection coefficients, so the dependence of spin-triplet AR on $x$ is mainly dominated by the transparency of interface. As shown in Fig. 6 the normal-state and spin-triplet AR conductance has the similar tendency on $x$, but the relative range of variation is larger for spin-triplet AR. With increasing $x$, the effect of interfacial alloy can smooth the potential step within the two layers adjacent to interface, hence the electronic structure mismatch between two leads is reduced, which results in lower normal-state reflection and resistance. For example, in Fe/Cr interface it has been found the interfacial roughness can raise the conductance by three times. On the other hand, with increasing the roughness the phase coherence of transport process will be destroyed, which generally suppress the conductance. So combining the two above-mentioned effects on AR conductance, it is reasonable that the spin-triplet AR will reach maximum at some interfacial alloy concentration $x$.

V. SUMMARY

In summary, in order to study the effect of interfacial magnetic disorder on spin-triplet AR spectra, we calculated the scattering matrices of Co$_2$MnSi/Al(001) interface in normal-state by first-principles noncollinear transport calculation. Next, combining scattering theory with Andreev approximation, we obtained the spin-triplet AR spectra for clean and rough interface with different degree of interfacial magnetic disorder.

The calculated results show that there is a conductance peak below the superconducting energy gap near zero bias for clean interface with interfacial magnetic disorder. Once the fluctuation of magnetic moments directions is sufficient strong or the interface has a small amount of roughness, this subgap conductance peak structure will be smeared out. In addition, our calculations show that the value of subgap conductance will increase with increasing the interfacial magnetic disorder. Therefore we argue that the reduction of experimental spin polarization from observation of nonzero subgap conductance in HF is owing to interfacial magnetic disorder effect. Meanwhile, the spin polarization of bulk HF is still 100%. Moreover, we also found that for rough interface the spin-triplet AR conductance at zero bias has the maximum value with interfacial alloy concentration around $x \approx 0.2$, which suggests that the impurity at interface can enhance the spin-triplet AR process.

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