Spin Dependence of Interfacial Reflection Phase Shift at Cu/Co Interface

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(Dated: February 2, 2008)

The spin dependent reflection at the interface is the key element to understand the spin transport. By completely solving the scattering problem based on first principles method, we obtained the spin resolved reflectivity spectra. The comparison of our theoretical results with experiment is good in a large energy scale from Fermi level to energy above vacuum level. It is found that interfacial distortion is crucial for understanding the spin dependence of the phase gain at the Cu/Co interface. Near the Fermi level, image state plays an important role to the phase accumulation in the copper film.

PACS numbers: 75.70.Cn, 75.47.-m, 75.47.Jn

I. INTRODUCTION

The spin dependent transparency of the nonmagnetic metal/ferromagnet (NM/FM) interface plays a crucial role in the magnetoelectronics. The spin dependent transmission and reflection result in the spin dependent interface resistance, which dominates the giant magnetoresistance effect. More interestingly, the spin dependent reflection at the NM/FM interface also yields the spin torques at the interface. Spin torque across NM/FM can be formulated in terms of the mixing conductance

\[ G_{\text{mix}} = \frac{1}{2} (M - \sum_{nm} |r_{nm}\rangle |r_{nm}\rangle \langle \exp(\ii \phi_{nm} - \phi_{nm}')| , \]

where \( M \) is the total number of channels (eigenstates), and \( |r_{nm}\rangle \exp(\ii \phi_{nm}')\rangle \) denotes the reflection from channel \( m \) to channel \( n \). The reflection phase gain difference (PGD) defined as \( \phi_{nm}' - \phi_{nm} \) plays an important role to understand the spin torque.

However, measuring spin dependent transport through FM/NM is far from trivial, since the electrons with different spin orientation flow together in most of the electric transport process. It is possible to probe the spin dependent transmission by making use of the two current model. The Andreev reflection (AR) spectroscopy at ferromagnet-superconductor interfaces in ballistic point contacts (PCAR), as an alternative way to measure the spin polarization, contains all the information of the scattering matrix at the interface but experimental data are very difficult to be resolved. It is deserved to have one method which can measure the transmission or reflection through an interface for different spins separately.

Recently, Wu et al. performed the measurement of the spin-dependent electron reflection on the Vacuum/Cu(001) system is obtained from first principles calculation and compared with experiment in a large energy scale. The interfacial distortion is found to be important for understanding the spin dependence of phase gain at Cu/Co interface. Effects due to the distortion on the spin transport are also discussed. Besides, we also estimate the possible phase gains from image states at the surface of Cu film. Such extra phase gain could be obvious near Fermi level.

This paper is organized as following: In Sec.II, we give a brief introduction of our theoretical method. The modelling of interfacial relaxation and the optimized structure are specified. The experiment details will also be given in this section. In Sec.III, we compare the theoretical results with experimental data, where interfacial alloy and image potential are taken into account. We also discuss the effect of structure relaxation on the spin transport at interface. Our results will be summarized in Sec.IV.

II. THEORETICAL METHOD AND EXPERIMENT

Several works tried to calculate the phase gain at the NM/FM interface, but the definition of the phase gain at a single interface for a given energy is not unified due to the arbitrary phase when solving the Schrodinger equation. Wei and Chou obtained the phase gain at Ag/Fe interface by comparing quantum well (QW) states positions of free standing Ag film and Ag film on top of Fe. While An et al. defined the phase gain at Cu/Co interface by a special fitting wave function. In fact, what can be measured in experiment is not the phase gain at single interface, but the sum of the phase gains at two opposite interfaces or the phase gain difference of two spin channels at the same interface.
Based on a recently developed first principles method\cite{12}, we obtained the phase gains by calculating the scattering matrix at interfaces. The arbitrary phase can be removed by noticing that the incoming wave of one interface is the outgoing wave of another interface for electrons scattering between two opposite interfaces. The phase accumulation obtained by the present method can be used to reproduce the positions of QW states from the electronic structure.

In this method, the bulk and the interface potentials are determined in the framework of the tight binding (TB) linearized muffin tin orbital (MTO) with surface green function (SGF) based on density functional theory (DFT) and local density approximation (LDA). Disordered systems are treated using the layered coherent potential approximation (CPA). As SGF is incorporated into TB-LMTO, the electronic structure of interfaces and other layer system can be treated in our framework. The potentials obtained in this way were used as input to a TB-MTO wave-function-matching (WFM) calculation of the transmission and reflection coefficients between Bloch states on either side of the interface. The details of WFM can be found in Ref.\cite{12}.

The interfacial distortion was calculated based on a recently developed first principles simulation package (VASP)\cite{15} with ultrasoft pseudopotentials\cite{16} and generalized gradient approximation\cite{17} for exchange correlation energy. Common lateral lattice constant used was the average value of the lattice constant of both metals obtained in VASP, viz. \( a_{\text{Cu}} = a_{\text{Co}} = 3.595 \) Å. The distortion was limited along [001] direction. For Cu/Co interface, the modelling is based on the supercell structure which consists of 6ML \( fcc \) Co(001) and 6ML Cu(001) with 12Å vacuum inserted between them. The three layers closed to vacuum at Co side and the three layers closed to vacuum at Cu side are fixed. The in-plane lattice constant is fixed to the common lattice constant. Energy cut-off is set to 400 eV and \( k \) grid is 19x19x1. For a given supercell the structure is relaxed to reach the minimum energy. The cell size is then varied, and a curve of structure vs energy can be obtained. The optimized structure with the lowest total energy is deduced from this curve. For the surface Vac/Cu, the supercell structure consists of 6ML Cu(001) and 12Å vacuum. Only the three Cu layers closed to vacuum are allowed to move. The optimized structure of Cu surface is obtained when the supercell reaches its minimum energy after relaxation.

Due to work function mismatch, the distance between Cu and Co layers is stretched to 1.86Å. At Cu side the distance between the neighboring layers is stretched by 3%. At Co side, the distance between the topmost two layers is contracted by 5% and the distance between the second and third layers is contracted by 1%. At Cu surface, the distance between the topmost two surface layers is contracted by 1%. With the atomic structure obtained from VASP as the input into the calculations of electronic structure and transport. The volume of atomic sphere at interface is a little different from that in bulk and selected to satisfy the principle of full space filling. The unrelaxed part of the two leads of interface are concatenated with bulk, where electronic structure of bulk has been obtained independently and the self-consistent potential of interface are treated as the "embedding potential" of the bulk system with aid of SGF\cite{12}. We found the relaxation of Co side is crucial to understand the spin dependent electron reflection correctly.

The spin dependent normal reflection with normal incidence in Vac/Cu/Co(001) system is calculated. As there is only one channel of \( \Delta \) symmetry in copper at the \( \Gamma \) point in the 2-dimensional Brillouin zone (2D BZ), the total reflectivity back to the vacuum for each spin is

\[
R = \frac{r_{B}^{2} + e^{-4d/\lambda}r_{C}^{2} - 2e^{-2d/\lambda}r_{BR}r_{BC}\cos(2k_{F}d - \phi_{B} - \phi_{C})}{1 + e^{-4d/\lambda}r_{B}^{2}r_{C}^{2} - 2e^{-2d/\lambda}r_{BR}r_{BC}\cos(2k_{F}d - \phi_{B} - \phi_{C})}
\]

where \( k_{F} = k_{BZ} - k \), \( k_{BZ} \) is the BZ wave vector, \( k \) is the electron momentum vector, \( r_{BZ}\exp(i\phi_{B}) \) and \( r_{BC}\exp(i\phi_{C}) \) are the reflection coefficients from Cu side at the Cu/Vac and Cu/Co, and \( d \) is the thickness of Cu film. Due to the inelastic scattering and the impurity scattering in Cu film, a mean free path (MFP) \( \lambda \) has to be incorporated, where MFP in the energy scale is assumed to be uniform to minimize our adjustable parameter in spite of possible energy dependence. By this way, only elastic reflection are taken into account and no inelastic reflection will be considered. The reflectivity asymmetry is defined as \( (R_{p} - R_{ap})/(R_{p} + R_{ap}) \) with \( R_{p}(R_{ap}) \) denoting total reflectivity with incident spin parallel to majority (minority) spin in Co.

To justify the theoretical calculation, we made the comparison between our calculation and the experimental data quantitatively. The experimental electron reflection spectrum for the electron energy higher than the vacuum level was obtained by the spin-polarized low-energy electron microscopy (SPLEEM) measurement. The experiment was performed at Lawrence Berkeley national laboratory. The detailed experimental description can be found in Ref.\cite{9}. In the SPLEEM, a spin polarized electron beam is directed at the sample surface at normal incidence and the spin-dependent electron reflectivity can be measured simultaneously during the Cu growth on 5ML Co/Cu(001). As shown in Eq.\cite{18}, the maximum electron reflectivity takes place at the interference condition of \( 2k_{F}d - \phi_{B} - \phi_{C} = 2\pi n \) where \( n \) is integer. So by measuring the thickness dependent electron reflectivity at certain electron energy, the \( k_{F} \) and the total phase gain \( \phi_{B} + \phi_{C} \) can be derived experimentally\cite{18}. However, the phase gain calculated using this method has larger error bar, which is larger than the PGD. To obtain the PGD precisely, we can determine the peak position difference \( \Delta \phi \) in the reflection spectrum of spin-up and spin-down electron, then the PGD can be calculated by the formula \( \Delta \phi = 2k_{F}\Delta d \).

The QW states for minority spin below the Fermi surface were measured by the angular resolved Photoemission at beamline 7.0.1.2 of the Advanced Light Source at
ical results. Due to the introduction of MFP in the Cu interface, the mean free path used in the calculation is 7ML. As the measurement of reflectivity is during the growth periods will be determined by the $\lambda = 7ML$. The mean free path used in the calculation is 7ML. The amplitude of theoretical reflectivity is not very sensitive to the interfacial distortion. The amplitude of theoretical reflectivity is not very sensitive to the interfacial distortion, since only the electron reflection at Cu interface is spin dependent. By quantitatively comparing our ab initio results and experimental data as shown in Fig. 1 (c), we can draw a conclusion that the interfacial distortion is very crucial to obtain the correct spin-dependent electron reflection spectrum. Here, we found that $\lambda = 7ML \pm 1ML$ in Cu from 8eV to 16eV gives the best reproduction of experimental data.

The effect due to the distortion of interface is more obvious for the phase gain at interface. In (a) and (b) of Fig. 2, the summation of the reflection phases of the two interfaces are shown for unrelaxed structure and relaxed structure respectively, where the shape of the spectra for two spins are similar to each other and the odd points of reflection phases appear at the band edge for each spin. The PGD spectra for the unrelaxed and relaxed Cu/Co interfaces are shown in Fig. 2 (c) with experimental data. Sign of PGD have been reversed from positive for the unrelaxed interface to negative for the relaxed interface, which is consistent with experimental results. Note that LDA fails to produce the image-like potential out of Cu surface. With referring to the empirical model, we have estimated the effect due to the image states on the reflection phase at Vac/Cu by shifting the potential at vacuum side to reproduce the surface potential. For energy far higher (> 8eV) than Fermi level, the effects due to the image states are small.

![FIG. 1: (color online) The calculated spin dependent electron reflectivity spectra with (a) unrelaxed interface, (b) relaxed interface and (c) the experimental spectra for the system of Vac/Cu(7ML)/Co(001). (d) The calculated reflection asymmetry for the spectra shown in (a)-(c). The experimental spectra have been multiplied by a factor 3 for better comparison. The mean free path used in the calculation is 7ML.](image1)

![FIG. 2: (color online) (a) Total reflection phase $\phi_B + \phi_C$ of the unrelaxed interfaces of Cu/Co and Cu/Vac. (b) Total reflection phase $\phi_B + \phi_C$ of the relaxed interfaces of Cu/Co and Cu/Vac. (c) Comparison of PGD of unrelaxed, relaxed interface with experimental data. (d) PGD of the two artificial interfaces discussed in the text.](image2)

III. NUMERICAL RESULTS AND DISCUSSION

To identify the effect of the distorted lattice, we calculated the energy dependence of reflectivity and reflectivity asymmetry for Vac/Cu(7ML)/Co(001) system with unrelaxed and relaxed interfaces shown in Fig. 1 where the unrelaxed interface is assumed to be of ideal fcc lattice. The amplitude of theoretical reflectivity is not very sensitive to the interfacial distortion. The amplitude obtained in experiment is almost 3 factor less than theoretical results. Due to the introduction of MFP in the Cu film, the amplitude is dominated by the Vac/Cu surface. As the measurement of reflectivity is during the growth of sample, the discrepancy mainly results from the impurities and inelastic scattering at Cu surface. However, the asymmetry spectra is more sensitive to the interfacial distortion, since only the electron reflection at Cu/Co interface is spin dependent.
and (ii). In (i), the structure at Co side of the relaxed Cu/Co interface was replaced by the ideal \textit{fcc} lattice. In (ii), the structure at Cu side of the relaxed Cu|Co interface was replaced by the idea \textit{fcc} lattice. As shown in of Fig. 2 (d), PGD of (i) is very similar to the spectrum of the completely relaxed interface, which indicates that Co side of the relaxed structure is the decisive factor for the PGD of the relaxed Cu|Co interface. The penetration of wave into the Co side will induce the spin dependent phase accumulation and the distortion of this part could affect greatly the PGD. The main difference of PGD between experimental data and our theoretical results is the general shape of the energy dependence. Two odd points exist in the theoretical results, however, there is no such clear point in experimental data at corresponding energy. Such difference can not be attributed to the uncertainty of interfacial structure model, as the odd points are due to the band edges of the two spin species(see Fig. 2 (a)&(b)) and the interfacial structure couldn’t smear the mismatch of band structures of Cu and Co. The possible contribution to the difference may come from the electron-phonon (ep) coupling at interface which is not incorporated in our calculation yet. The ep coupling has been found could be important for the phase shift at metallic interfaces\textsuperscript{22}. With the including of phonon, the energy dependence of reflection phase is dominated not only by the electron band structure but also by the phonon spectrum. It is expected that the difference of the energy dependence of PGD may be compensated by the mixing of electron band structure and phonon spectrum.

In experiment, one monolayer alloy could be located at interface. It was found that variation of the PGD due to 1ML substitutional alloy Cu\textsubscript{x}Co\textsubscript{1-x}(0 < x < 1) inserted between Cu and Co is small(< 2 DEG), which is consistent with above discussion as 1 ML alloy only slightly changes the structure at Co side. However, interfacial alloy might induce some extra phase gain when electrons are reflected from the interface, which could change both energy and Cu film thickness dependent spectra of reflectivity.

Up to now, the phase discussed is above Fermi level. The phase near Fermi level may be more meaningful as most electronic transport experiments are carried out around this energy. Experimentally, it is very hard to separate the contributions of majority and minority spin at Fermi level. However, it is well known that only the electrons with minority spin can form QW states at Fermi level in Cu/Co(001) system. So the phase gain of minority spin could be obtained experimentally. Fig. 3 shows our theoretical calculation\textsuperscript{28} and the experimental data. As we discussed, the image states may have the effect on the phase gain for the electron lower than Fermi level. The including of image states shifts down the total phase up to 50 DEG in this energy scale as shown by the uncertainty bar. Such large correction is important when comparing with experimental data. Our theoretical results give the good reproduction of the slope of total phase. The small discrepancy in magnitude can be roughly compensated by replacing the layer of Cu at interface with 1ML Cu\textsubscript{0.5}Co\textsubscript{0.5} alloy, which could further shift down the phase spectra as shown by the solid line in Fig. 3. The total phase of majority spin is also present, which is far from the experimentally measured one.

For $\Gamma$ point in 2D BZ, we have shown that the relaxation of atomic structure could greatly affect the spin transport. But, this is not enough to understand the spin transport. To discuss the effect of interfacial distortion on the quantities such as interface resistance or mixing conductance, we should extend our calculation to the whole 2D BZ at Fermi level. The results are shown in Tab. I. For Cu|Co interface, interface resistance of majority spin is less affected, however, minority spin is well affected by structure relaxation. For majority spin, $d$ bands in Cu and Co are fully filled and those states at Fermi level are from $s$ bands which are essentially free electron like states. The interface could be equivalent to a very low potential step for those states which have the same symmetry in both materials. Interfacial distortion

![FIG. 3: (color online) The calculated reflection phase $\phi_D + \phi_C$ of the majority spin(solid circle) and minority spin(open triangle) with the experimentally measured phase(open square). The solid line indicates the lower limit (position of error bar) of minority spin after taking interfacial alloy into account as discussed in text.](image)

**TABLE I: Interface resistance $SR$ in units of $f\Omega m^2$, the mixing conductance $G_{mix}$ in units of $10^{15} \Omega^{-1} m^{-2}$.**

| system                                    | $R_1$ | $R_1$ | $Re(G_{mix})$ | $Im(G_{mix})$ |
|-------------------------------------------|-------|-------|---------------|---------------|
| unrelaxed CuCo (001)                      | 0.303 | 1.91  | 0.556         | -0.021        |
| relaxed CuCo (001)                        | 0.304 | 2.42  | 0.547         | -0.021        |
| relaxed Cu$_{50}$Co$_{50}$ (001)          | 0.306 | 1.53  | 0.531         | -0.026        |
| unrelaxed AgFe (001)                      | 1.00  | 6.79  | 0.445         | -0.003        |
| relaxed AgFe (001)                        | 1.04  | 6.10  | 0.444         | -0.0006       |
| relaxed Ag$_{50}$Fe$_{50}$ (001)          | 1.02  | 2.59  | 0.441         | 0.0002        |

\textsuperscript{a}The slight difference of $SR$ between present results and those in Ref. 29 comes from different atomic potentials. Therein, $spd$ basis is used.
is just like the variation of the potential profile, which is expected to have little effect on the transport. For minority spin, $s$ band still dominates in Cu, but $d$ bands in Co are only partly filled, which lead to the multiple sheets of Fermi surface in Co. Interface resistance should be attributed to the mismatch of band structure in Cu and Co. e.g., mismatch of Fermi velocity and mismatch of symmetry of wavefunction. Interfacial distortion could play important roles in those issues. Furthermore, our results show that one layer of interfacial alloy could enhance such effect. This can be understood as more states of minority spin at Co side could be diffusively coupled to those at Cu side with aid of the alloy.

However, $G_{mix}$ is not sensitive to interfacial distortion. According to the definition of $G_{mix}$, there exists a term $\left| r^{nm}_i \right| \left| v^{nm} \right| \exp(i(\phi^{nm}_i - \phi^{nm}_o))$ with the summation of the 2D BZ. For the single point in 2D BZ this term is sensitive to the interfacial distortion, however, the summation could lead to the cancellation of this term among different points in 2D BZ due to the phase factor $\phi^{nm}_i - \phi^{nm}_o$. The real part of $G_{mix}$ obtained in our calculation is very close to the value of $\frac{2}{\pi} M$, which means strong cancellation happened. As a result, the sensitive issue of interface reflection phase could be trivial one.

To confirm above argument, we also studied the AgFe(001) interface. The modelling of the interfacial relaxation is just like that for Cu/Co interface, but note that in this case we need to rotate the bcc Fe(001) plane by 45 DEG to match the fcc Ag(001) plane. The lattice constant $a_{Fe} = \sqrt{2} a_{Ag} = 2.855\text{Å}$. After relaxation the distance between Fe and Ag layers is about 1.39 times that of neighboring layers’ distance in bulk Fe. The contraction of the top two layers at Fe side is by 1% and that at Ag side is less than 1%. Similar results were also obtained in this system. The interface resistance of majority spin and the real part of $G_{mix}$ are both less sensitive to the interfacial distortion shown in Tab. I. The magnitude of the imaginary part of $G_{mix}$ in this case greatly decreases for the relaxed interface, but the imaginary part is too small compared with the real part to contribute to spin transfer torque.

\section{IV. CONCLUSION}

The phase information obtained from experiments can be well understood by our \textit{ab initio} calculation in a large energy scale, it was found that structural distortion at interface is important for theoretical model to interpret experiment. The PGD of two spins obtained in our calculation is comparable with experimental results. At Fermi level, extra phase gain due to image states becomes obvious. After integration of 2D BZ, we also found that the interface resistance of minority spin can be well affected by the interfacial distortion, however, the interface resistance of majority spin and the PGD related quantity mixing conductance are not sensitive to the interface structure.

We thank Professor Z. Q. Qiu for the helpful discussions. This work is financially supported by NSF (Grant No. 10634070, 10604015, 10621063) and MOST (Grant No. 2006CB933000, 2006CB921300, 2006AA03Z402) of China.
Here, we only use the sphere potential to mimic the empirical model of image state. The results are only used as an estimation of uncertainty in our calculation.

Comparing with other theoretical and experimental results[24,25,26], our calculated $sp$ band which dominates QW stats here is lower by around 0.30 eV. The DFT-LDA band error[11,27] should respond to such discrepancy. The error is corrected by a rigid shift of band.

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