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Spin-Dependent Tunnelling from Transition-Metal Ferromagnets

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Spin-dependent tunnelling from ferromagnetic Ni, Co, and Fe, is theoretically examined and it is shown that the tunnelling current is predominantly due to *s* electrons, despite the much greater density of *d* states in these metals. *s*-*d* hybridization leads to a positive (parallel to the majority *d* electrons) polarization within ordinary Stoner (Hartree-Fock) theory, and it was found that it was insufficient to explain the experiment. However, if the self-energy due to spin-wave emission is included, good agreement with experiment for reasonable parameters was obtained. Hence, it is concluded that the band-theoretical description of the magnetism of these metals is compatible with the recent experiments of Tedrow and Meservey, provided that the dominant many-body effects are properly taken into account.

I. TUNNELLING FROM *d*-BAND METALS

In a recent series of experiments, Tedrow and Meservey measured the spin polarization of electrons tunnelling from ferromagnetic Fe, Co, Ni, and Gd into superconducting Al through a barrier layer of Al_2O_3.[1] In each case the polarization was parallel to the majority spins in the metal. In at least two of the cases (Ni, Co), this finding contradicted the most naive expectation, namely, that the polarization of the tunnelling current should be proportional to the polarization of the Stoner-Wohlfarth band-theoretical density of states at the Fermi energy. In both these metals, band-theoretical calculations show the majority-spin *d* band totally filled below the Fermi energy. Therefore (ignoring the *s* electrons, whose density of states is much lower than that of the *d* electrons), one would expect a total negative polarization of the tunnelling current, instead of the positive values observed.

However, within the WKB approximation Harrison showed that the tunnelling current in a band is not proportional to the density of states; the density-of-states factor is canceled by a corresponding change in the tunnelling matrix element.[2] On these grounds we can reject the picture presented in the preceding paragraph. In the following we show that the tunnelling current due to *d* electrons is further reduced.

Let us first consider electrons in the transition-metal electrode in terms of Bloch states.[3] The *s* electrons are almost free, with small band gaps at the Brillouin-zone edges. In a pseudopotential picture, they are made up of just a few plane waves, so their wave functions have nearly uniform modulus. On the other hand, *d* states, because of their localized nature as well as their more complex $l=2$ structure within one unit cell, are made up mostly of many plane waves of large wave vector (larger than the wave vectors of the plane waves which are used in making up the *s* states). Now, the tunnelling matrix element in WKB approximation is given by

$$D(E) \propto \exp\left(-\int_a^b \{2m[V(x)-(E-k_{\parallel}^2/2m)]\}^{1/2}dx\right), \quad (1)$$

where $k_{\parallel}$ is the wave vector in the plane of the interface, which is assumed conserved. We can see from (1) that tunnelling is much more difficult for the plane-wave components with large $|\vec{k}|$ in the *d* wave function (since their $k_{\parallel}$ are, typically, also large). Furthermore, these components describe a charge distribution localized within about 0.5 Å of the ionic sites, and add up incoherently to nearly zero outside this region. In picturing the neighborhood of the metal-insulator interface, we should place the "boundary" between metal and oxide (beyond which the wave function decays exponentially) something like halfway between the last metal ion and the first atom in the insulating layer. That is, it should be about one Wigner-Seitz radius away from the last metal ion, in a region where the large $|\vec{k}|$ components are interfering destructively. Hence, such components cannot contribute significantly to the tunnelling matrix element.

In addition, for the present case, even if these large $|\vec{k}|$ components penetrate through the insulating layer, their rapid variation in the plane of the oxide-Al interface does not match with the *s*-like wave functions of the Al. As far as these components are concerned, the Al might almost as well be another piece of insulator.

Hence, we conclude that the only components in the *d* wave function which contribute to the tunnelling are those of $|k| \lesssim \pi/l$. (l is the lattice constant.) If we write the renormalized-atomic d wave function approximately as

$$P_d(r) \approx (1/\sqrt{v})[R + \phi(r)],$$

the magnitude of such components is equal to R. Here, v is the volume of the Wigner-Seitz sphere and R is a constant representing the tail of the wave function. $\phi(r)$ is the part which describes the charge density near the core and vanishes near the boundary of the Wigner-Seitz sphere. On the basis of the calculation of Hodges, Watson, and Ehrenreich[4] we estimate $R = 0.3$ for the $3d$ wave functions. Hence, the tunnelling current due to d electrons is reduced by a factor of $(0.3)^2$ relative to what we would find if we approximated the d wave function by a plane wave. In essence, what we are saying is that in the Harrison theory the wave function for the d electrons, being approximated by a plane wave, should be normalized to R^2 instead of unity in the transition-metal electrode, in order to remove from consideration the charge localized near the ions, which cannot tunnel. We note that this rough estimate of the relative importance of d and s parts of the tunnelling current is consistent with the detailed calculations of Politzer and Cutler for the closely related field-emission current.[5]

Now, we consider the same problem by using the tunnelling theory of Caroli, Combescot, Noziéres, and Saint-James[6] (CCNS). They have formulated their theory in terms of both Wannier and Bloch states. For d electrons the former version is more convenient, although, then we must also treat s electrons in Wannier basis. For a single band, the expression for the current takes the form

$$\langle J \rangle = (2\pi)^2 e \int_{E_F}^{E_F+eV} \frac{d\omega}{2\pi} \left| G_{ba}(\omega) \right|^2 T^2 \rho_\alpha(\omega) T'^2 \rho_\beta(\omega) \;. \tag{2}$$

Here, α is the last site before the insulator in the left electrode, β is the first site in the right electrode, ρ_α and ρ_β are the local densities of states at these sites, and $G_{ba}(\omega)$ is the Green's function for propagation between site a, the first site in the insulator and site b, the last one. $T \equiv T_{a\alpha}$ and $T' \equiv T_{b\beta}$ are hopping matrix elements between those sites. It is not unreasonable to assume that T is *proportional* to $T_{i,i+1}$, the hopping matrix element in the bulk of the left electrode [the meaning of this statement becomes clearer in Eq. (5)], and that ρ_α is essentially the bulk electrode density of states. Then for a tight-binding band where the *average* density of state $\bar{\rho}_\alpha$ is proportional to the reciprocal of the hopping matrix element, we get a current

$$\langle J \rangle \propto T^2 \bar{\rho}_\alpha \approx T \approx 1/\bar{\rho}_\alpha \;. \tag{3}$$

If we apply this result uncritically to the s band as well as d bands, using their known densities of states, we predict that about 90% of the current should come from the s electrons, in agreement with our previous argument. This agreement is not completely accidental. $T_{i,i+1}$ is closely related to the overlap of tails of two neighboring Wannier states, and recalling our definition of R we can argue that $T^d/T^s \sim R^2$. Here $T^d(T^s)$ is $T_{a\alpha}$ for $d(s)$ electrons. In the following we proceed with the CCNS model because it is easily generalized to take s-d mixing into account. We will ignore the multiplicity of the d bands, assuming only one of the d states mixes with the s state at a given value of $\vec{k}$. Equation (2) can be extended to the two-band case in a straightforward manner, and the result can be expressed by simply replacing $T^2\rho_\alpha$ in Eq. (2) by

$$\hat{T}^\dagger \hat{\rho} \hat{T} = (T^s,\ T^d) \begin{pmatrix} \rho_{ss} & \rho_{sd} \\ \rho_{ds} & \rho_{dd} \end{pmatrix} \begin{pmatrix} T^s \\ T^d \end{pmatrix} .$$

Here, $\rho_{nn'} = \pi^{-1} \operatorname{Im} g_{\alpha\alpha}^{nn'}(\omega)$; $n, n' = s, d$, and $g_{\alpha\alpha}^{nn'}$ is the time Fourier transform of the Green's function for the bulk electrode:

$$g_{\alpha\alpha'}^{nn'}(t) = -i \langle T(C_\alpha^{(n)}(t) C_{\alpha'}^{(n')\dagger}(0)) \rangle ,$$

where $C_\alpha^{(n)\dagger}$ and $C_\alpha^{(n)}$ are creation and annihilation operators for the Wannier state in band n at site α. It then follows that the total current is given by

$$J \propto \left| T^s \right|^2 \rho_{ss} + \left| T^d \right|^2 \rho_{dd} + 2 T^s \rho_{sd} T^d \;. \tag{4}$$

Note the presence of the cross term, which comes from processes where an s electron propagates across the insulator in one direction and a d hole goes in the other direction.

II. s-d HYBRIDIZATION

Now given estimates of the ratio

$$T^s/T^d \approx T^s_{i,i+1}/T^d_{i,i+1} , \tag{5}$$

knowing the densities of s and d states (as well as the mixed density of states ρ_{ds}) in the transition-metal electrode, we can estimate the total current. What we need to know is what part of the states on the Fermi surface is s like and what part is d like. With a magnetized d band, this degree of hybridization will differ for up and down spins, giving rise to a polarized current. This is easy to calculate. In the spirit of the Mueller-Hodges-Ehrenreich-Lang parametrization scheme,[7] we consider an s-d mixing Hamiltonian

$$H_{\text{mix}} = \sum_{k\sigma} V_k C_{kd\sigma}^\dagger C_{ks\sigma} + \text{H. c.} \tag{6}$$

We will pretend that V_k is relatively constant over the Fermi surface, and we can estimate its size by looking at the splitting $2|V_k|$ it creates at points in the Brillouin zone where s and d electrons would otherwise have been degenerate. On the basis of the published Hodges-Ehrenreich-Lang (HEL) band structure,[7] $|V_k|$ is between 1.0 and 1.5 eV.

In the absence of hybridization, the s and d electrons have propagators

$$G^0_{nn,\sigma}(k,\,E)=1/(E-\epsilon_{kn\sigma}) \tag{7}$$

where n is a band index, s or d. The hybridization problem is exactly soluble; the renormalized propagators are

$$\begin{aligned} G_{ss,\sigma}(k,\,E)&=\frac{1}{E-\epsilon_{ks\sigma}-|V_k|^2/(E-\epsilon_{kd\sigma})}\,,\\ G_{dd,\sigma}(k,\,E)&=\frac{1}{E-\epsilon_{kd\sigma}-|V_k|^2/(E-\epsilon_{ks\sigma})}\,,\\ G_{ds,\sigma}(k,\,E)&=G^0_{ss,\sigma}V_kG_{dd,\sigma}=G_{ss,\sigma}V_kG^0_{dd,\sigma}\\ &=\frac{V_k}{(E-\epsilon_{ks\sigma})(E-\epsilon_{kd\sigma})-|V_k|^2}\,. \end{aligned} \tag{8}$$

They all have poles at the exact eigenenergies

$$E_k^{\pm}=\tfrac{1}{2}\{\epsilon_{kd\sigma}+\epsilon_{ks\sigma}\pm[(\epsilon_{ks\sigma}-\epsilon_{kd\sigma})^2+4|V_k|^2]^{1/2}\}, \tag{9}$$

but different residues at these poles. The residue of G_{ss} (G_{dd}) at $E^{\pm}$ tells how much of this exact eigenstate is made up of the bare-s (bare-d) state. If $Z_{nn',\sigma}(E)$ is the average value of this residue over all k's with $E^{\pm}_{k\sigma}=E$, we have

$$\rho_{nn',\sigma}(E)=Z_{nn',\sigma}(E)\,\rho_\sigma(E)\,, \tag{10}$$

where $\rho_\sigma(E)$ is the density of (exact) eigenstates at E. From (8) the Z's are

$$\begin{aligned} Z_{dd\sigma}(E)&=\left\langle\left(1+\frac{|V_k|^2}{(E-\epsilon_{ks\sigma})^2}\right)^{-1}\right\rangle_{\rm av}\,,\\ Z_{ss\sigma}(E)&=\left\langle\left(1+\frac{|V_k|^2}{(E-\epsilon_{kd\sigma})^2}\right)^{-1}\right\rangle_{\rm av}\,,\\ Z_{sd\sigma}(E)&=\left\langle\frac{V_k}{[(\epsilon_{ks\sigma}-\epsilon_{kd\sigma})^2+4|V_k|^2]^{1/2}}\right\rangle_{\rm av}\,, \end{aligned} \tag{11}$$

where the average is in the sense described above. We can then use (10) and (11) in (4) to calculate the tunnelling current.

III. NUMERICAL ESTIMATES

We apply this argument first to nickel. Consider as typical the [100] direction in the HEL band structure.[7] The d band which runs from Γ_{12} to X_5, which gives most of the magnetization of the system, does not mix with the s-band states. To the extent that this is true in all directions, the part of the current from this band is just

$$J_\sigma(\Gamma_{12}-X_5)=|T^d|^2\rho_{\Gamma_{12}\text{-}X_5,\sigma}(E_F)\,. \tag{12}$$

There is no cross term between this band and any of the others [as the last term in (4)] because of the absence of mixing. The important hybridization is between a Γ_{12}-X_1 d band and the s band. By comparing the $\epsilon(k)$ curves with those we would have in the absence of mixing, we conclude that the extent of the hybridization is maximal ($Z=\frac{1}{2}$) just inside the Fermi surface. The states on the so-called s-like part of the Fermi surface still contain a lot of d character. Because the minority-spin d electrons lie higher in energy, they will be mixed in more strongly (larger Z_{dd}) than the majority d electrons. To this extent it will be easier for majority spin electrons to tunnel.

If we adopt the estimates of the band splitting used by HEL or by Herring,[8] $\Delta\lesssim 0.4$ V, and if we infer

$$Z_{ss\downarrow}\approx 0.6=1-Z_{dd\downarrow} \tag{13}$$

from the HEL band-structure curves for the minority spins, we find

$$Z_{sd\downarrow}=0.49,\quad Z_{ss\uparrow}=0.7=1-Z_{dd\uparrow}\,,\quad Z_{sd\uparrow}=0.46\,. \tag{14}$$

If we assume further that the tunnelling matrix elements differ by a factor of 10, and that 90% of the total density of states comes from the Γ_{12}-X_5 band, we find a net polarization of

$$P=(J_\uparrow-J_\downarrow)/(J_\uparrow+J_\downarrow)\approx+1\%\,. \tag{15}$$

We have assumed here that the Γ_{12}-X_5 d band is completely polarized in the minority direction. The resulting P is quite sensitive to this, and to the ratio T^s/T^d, so Eq. (15) is not very precise. It does show how the s-electron polarization, although rather small, can begin to counteract the large d-electron polarization because of the larger tunnelling matrix element, but P is still an order of magnitude too small.

However, this estimate assumed a constant Stoner band splitting Δ everywhere in the d bands. We believe that this is naive, particularly in view of the fact that the derivation of the effective interaction strength $U_{\rm eff}$ (the Kanamori t matrix[9]) is only valid for small hole energies. This is not unreasonable near E_F. But an examination of the energy dependence of the t matrix[10] shows that at high hole energies (deep in the band) the interaction may even be enhanced by the same correlations which reduce it near E_F. The onset of ferromagnetic order weakens the Kanamori correlation effects somewhat, but the possibility of virtual-spin-wave emission now also shifts the single-particle energies.[11] We take the Hertz-Edwards theory as a guide to these effects in the ferromagnetic state.[12] Here the minority spin d electrons have the same band structure that they would in Stoner theory (because the majority band is full), but the majority states are significantly altered. Because of the possibility of spin-wave emission, the majority electrons have an energy shift (in addition to the Hartree or Stoner shift) which is positive near the top of the band and negative near the bottom. (These shifts have the same signs as those produced by the energy-dependent Kanamori t matrix

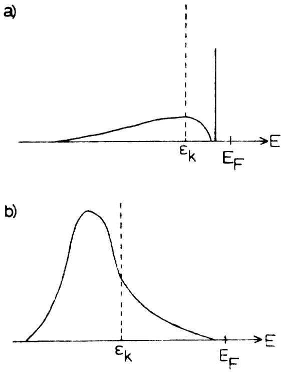

FIG. 1. Spectral weight functions for majority-spin *d* electrons (schematic) (a) near the top of the *d* band; (b) in the middle of the *d* band. The dotted line shows the position of the Hartree energy level ϵ_k.

mentioned above). This decreases the effective band splitting from its bare value near the Fermi level, and increases it deep in the band. For a given electron momentum, the majority electron spectral weight function will no longer have a single δ function of unit weight at $\epsilon_{k\downarrow} - \Delta$. It may have one δ function plus a continuous part (nonvanishing in the region where real-spin-wave emission is energetically allowed), or even two δ functions, one above and one below the continuum region. (To make contact with the spectral weight functions plotted in Ref. 12, remember that the "electrons" of that paper correspond to the holes in a real *d* band, so the spin axis and the sense of the energy should be reversed.)

For majority *d* electrons near E_F, most of the spectral weight should be concentrated in a δ function above the continuum, or in a fairly narrow resonance near the top of the continuum. This is shown in Fig. 1(a). This spike or resonant peak will lie at an energy above the position of the single spike in the Hartree spectral weight function—the effective band splitting is less than the bare value, as in the Kanamori-Herring picture. In our view, part of this reduction comes from the Kanamori effects and the rest of it comes from the spin-wave emission self-energy shift.

But this picture is only valid near the top of the band. The *d* states with which the *s* states at the Fermi surface mix lie fairly deep in the band—about halfway down or so. Here, the single-particle states are heavily damped, but their net self-energy shift from Kanamori and spin-wave emission processes is small, or perhaps even negative. Their spectral weight is, therefore, likely to be mostly in a resonance in the continuum part of the spectral weight function, centered near or slightly below the position of the Hartree spike, as shown in Fig. 1(b).

Let us now go back and recompute the averaged residues (11) for the case of an *s*-electron mixing not with a simple free *d* electron whose propagator is given by (7), but with the highly correlated *d* electron whose propagator is derived from a spectral weight function with all this extra structure. However, for the rough estimates we can make, we may as well pretend that all the majority *d* electron spectral weight is concentrated in a single spike at an energy $\epsilon_k - \Delta_{\text{eff}}(E_k)$, where E_k is the position of the resonance and Δ_{eff} includes the Hartree, Kanamori, and magnon emission terms. For the *d* states in question here, as we noted above, the last two terms are small and $\Delta_{\text{eff}}(E_k)$ is roughly the *bare* (Hartree) band splitting or a little larger, between 1.0 and 1.5 eV.

The fact that this shift is of the order of the hybridization matrix element V_k means that the majority states on the "*s*-like" part of the Fermi surface are much less *d*-like than their minority counterparts. If we continue to take $Z_{s\downarrow} = 0.6$ for the minority spin, we get

$$Z_{ss\uparrow} \approx 0.84 = 1 - Z_{dd\uparrow}, \quad Z_{sd\uparrow} = 0.37 \tag{16}$$

(using $\Delta = 1.3$ eV). From this we get a current polarized by

$$P = +8\%, \tag{17}$$

which is not far from the experimental value of +11%. Again, we could get values of P anywhere from 5 to 15% by making plausible revisions of our estimates of the tunnelling matrix elements. But, the order of magnitude (and sign) are always right, a result we could not obtain using the conventional small estimate for Δ.

Next, consider cobalt, whose band structure has been studied by Wakoh and Yamashita[13] and Wong *et al.*[14] The qualitative picture of the mixing between *s* and *d* states is similar to that in nickel. Here, the hybridization matrix element seems to be somewhat larger, probably over 2 eV. Partly as a consequence of this, the minority states on what we have been calling the *s*-like part of the Fermi surface are actually more *d* like than *s* like. This is not true of the majority states, since their *d* energies are low. Reference 13 used a Stoner band splitting of 1.7 eV, while Ref. 14 took $\Delta = 1.35$ eV. Again, we feel it is a better idea to use a larger Δ_{eff} in calculating the fractional parentage coefficients [Eq. (11)]. If we assume the same bare interaction U that we had in nickel and use the relation

$$\Delta = U\sigma, \tag{18}$$

where σ is the magnetization $n_\uparrow - n_\downarrow$ (= 1.6 for Co), we find a bare Δ of about 2.5 eV. The Δ_{eff} for the states which mix with the *s* states to form the states at E_F should be a little larger, maybe 3 or 4 eV.

If we use $Z_{ss\downarrow} = \frac{1}{3}$ for the minority spins, we find $Z_{ss\uparrow} \approx 0.84$ because of the larger Δ_{eff}, and the predicted polarization is

$$P = +26\%. \tag{19}$$

Again, the agreement with experiment is reasonable (the experimental P is 34%). The larger Δ_{eff} produces a larger P.

Finally, consider iron. Wakoh and Yamashita have also studied its band structure.[15] It differs from Co and Ni in that the d bands are not completely polarized (this is often called weak ferromagnetism), and the total magnetization is larger ($\sigma = 2.2$). To estimate P, we will use $|V_k| = 2.25$ eV, as in Co, for the hybridization, and $Z_{ss\downarrow} = \frac{1}{3}$. These values seem reasonable guesses to make from the band structure of Ref. 15. We need a Δ_{eff} larger than before, because of the larger magnetization, $\Delta_{eff} = 4.8$ eV. Then $Z_{ss\uparrow} = 0.89$. If we assume a + 30% polarization of the purely d electrons,[16] we find

$$P = +37\% ,$$

which is to be compared with the experimental $P = 44\%$. The theoretical value of P is not very sensitive to the assumed value of the pure d-electron polarization.

Throughout this discussion we have ignored *s-d exchange*. This would merely shift the up- and down-spin s bands relative to each other, and since their densities of states vary slowly with energy, it would not lead to any significant polarization of the current.

Related to the present are the spin-dependent photoemission experiment, in which it was found that photoelectrons from the neighborhood of E_F in ferromagnetic Ni, Co, and Fe are also polarized in the majority-spin direction.[16] However, the physics of the photoemission process is very different from that of the tunnelling we have examined here. Most significantly, photoemission spectra always show a large d-band contribution.[17] Furthermore, the energy resolution of the photoelectron spectra (about 0.5 eV) is much larger than that in a tunnelling experiment (a few meV). Anderson has discussed the possible influence of spin-wave emission self-energy shifts (of the sort described by Hertz and Edwards[12]) on photoelectron spectra.[11] It appears very difficult to calculate accurately the spin polarization of the photoemission spectra of these metals, but their qualitative features, notably the positive peak near threshold, are consistent with the Hertz-Edwards theoretical picture.[12,18]

In conclusion, we find that the Tedrow-Meservey tunnelling experiments are compatible with the band theory of ferromagnetism, providing s-d mixing is properly included and the energy dependence of the effective spin splitting is recognized. As a further test, it seems to us that the difference between majority and minority spin states should also show up in other Fermi-surface-sensitive experiments. Not only are there two Fermi surfaces, one for each spin, but the effective mass of the minority-spin electrons should be larger too, and this should be detectable.

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