How to Define and Calculate the Degree of Spin Polarization in Ferromagnets

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Different ways to define and calculate the degree of spin polarization in a ferromagnet are discussed, particularly with respect to spin-polarized tunneling and Andreev reflection at the boundary between superconductor and ferromagnet. As an example, the degree of spin polarization for different experiments in Fe and Ni is calculated in the framework of the local spin density approximation and used to illustrate the differences between various definitions of spin polarization.

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Although solid state physicists use the notion of a degree of spin polarization (DSP) of a ferromagnet (FM) rather often, it is not well defined. While the total magnetization is uniquely defined as the difference between the number of spin up and spin down electrons, it tells us little about how much different spins do contribute to transport properties. In view of the growing number of experiments probing spin polarization [1], it becomes increasingly more important to be able to calculate the DSP in the framework of the conventional band theory (and eventually beyond it). Importantly, the DSP can be defined in several different ways. In order to compare the calculations with the experimental data it is crucial to make sure that a proper definition of the DSP is used. In particular, spin-polarized tunneling in various forms [1], including Andreev reflection [2], provides valuable information about the spin dependence of the electronic structure, but this information may be obscure and not very useful unless the measurements are backed by the calculation *appropriate for the experiment in question*.

Let us consider an extreme example, the so-called half-metallic magnets. Such systems do not have any electrons at the Fermi level in one of the two spin channels; they have 100% spin polarization according to any sensible definition. On the other hand, for a regular magnetic metal, which has Fermi surfaces in both spin channels, it is not obvious *a priori* how to define the degree of spin polarization.

The most natural, and probably the most popular definition is $P = (N_\uparrow - N_\downarrow)/(N_\uparrow + N_\downarrow)$, where $N_{\uparrow(\downarrow)}$ is the density of electronic states (DOS) at the Fermi level, defined as ($\hbar \equiv 1$)

$$N_i = \frac{1}{(2\pi)^3}\sum_\alpha \int \delta(E_{\mathbf{k}\alpha i})d^3k = \frac{1}{(2\pi)^3}\sum_\alpha \int \frac{dS_F}{v_{\mathbf{k}\alpha i}}, \tag{1}$$

and $E(v)_{\mathbf{k}\alpha i}$ is the energy (velocity) of an electron in the band α with spin i ($\uparrow$ or $\downarrow$) and the wave vector $\mathbf{k}$. A typical experiment that can probe P_N is spin-polarized photoemission. This definition of the DSP may be called "N"-definition, P_N, and its usefulness is limited by the fact that the transport phenomena usually are *not* defined by the DOS alone. This is particularly true for materials which have both heavy d-electrons and light s-electrons at the Fermi level (e.g., Ni). While the DOS is mostly defined by the former, the electric transport is primarily due to the fast s electrons (cf. a semiempirical recipe of defining DSP via partial s-DOS in transition metals [3]).

Classical Bloch-Boltzmann transport theory [4] lets us separate the currents of the spin-up electrons and the spin-down electrons, and to define DSP via the current densities $J_{\uparrow(\downarrow)}$ as $(J_\uparrow - J_\downarrow)/(J_\uparrow + J_\downarrow)$, $J_{\uparrow(\downarrow)} \propto \langle Nv^2\rangle_{\uparrow(\downarrow)}\tau_{\uparrow(\downarrow)}$. Assuming the same relaxation time τ for both spins, this definition leads to the "Nv^2" DSP (here and below we set $E_F = 0$),

$$P_{Nv^2} = (\langle Nv^2\rangle_\uparrow - \langle Nv^2\rangle_\downarrow)/(\langle Nv^2\rangle_\uparrow + \langle Nv^2\rangle_\downarrow), \tag{2}$$

where $\langle Nv^2\rangle_{\uparrow(\downarrow)}$ is defined as

$$\begin{aligned}\langle Nv^2\rangle_i &= (2\pi)^{-3}\sum_\alpha \int v^2_{\mathbf{k}\alpha i}\delta(E_{\mathbf{k}\alpha i})\,d^3k \\ &= (2\pi)^{-3}\sum_\alpha \int v_{\mathbf{k}\alpha i}\,dS_F\,. \end{aligned}\tag{3}$$

This quantity is sometimes denoted as $(\frac{n}{m})_{\mathrm{eff}}$ and is proportional to the contribution of the corresponding electrons to the plasma frequency (see, e.g., Ref. [4]). If spin-dependent or spin-flip scattering is present, the total current in each spin channel depends on the characteristics of both spin subsystems, and the expression for the DSP becomes very complicated.

Unfortunately, it is hardly possible to measure $J_\uparrow$ and $J_\downarrow$ separately. A typical experiment involves spin-polarized tunneling between a FM and another material. In particular, one can measure tunneling currents separately for both spin polarizations for a ferromagnet/superconductor contact. The question arises whether the DSP measured in such a way is P_N or P_{Nv^2}. To answer this, we start from the simplest case, a ballistic contact with no barrier, and neglect mismatch of the Fermi velocities at the contact. We repeat the original Sharvin [5] derivation, but allow for arbitrary Fermi surface geometry. Following Sharvin, we assume that an electron going through the contact experiences the acceleration by the electric field so that its energy increases by eU. If the field changes the electron's quasimomentum from $\hbar\mathbf{k}$ to $\hbar\mathbf{k}'$, we find that the phase space for this process is defined at $T = 0$ by the factor

$$\theta(E_{\mathbf{k}'})\theta(-E_{\mathbf{k}}) = \theta(E_{\mathbf{k}} + eU)\theta(-E_{\mathbf{k}}) = eU\delta(E_{\mathbf{k}}). \tag{4}$$

The fraction of electrons with a given $\mathbf{k}$ that can reach the contact in a unit time is $v_x A$ (the contact plane is perpendicular to x and A is the area of the contact). The total current is

$$\begin{aligned} I &= \frac{e^2UA}{(2\pi)^3}\sum_\alpha \int_{v_x>0} v_x\delta(E_{\mathbf{k}\alpha})\,d\mathbf{k} = e^2UA\langle Nv_x\rangle \\ &= \frac{e^2UA}{(2\pi)^3}\sum_\alpha \int_{v_x>0} v_x\,\frac{dS_F}{v} = e^2UAS_x, \end{aligned} \tag{5}$$

where S_x is the area of the projection of the Fermi surface onto the interface plane. For a Fermi sphere this reduces to the Sharvin result. Correspondingly, we arrive at the third, ballistic definition of spin polarization, P_{Nv}.

The next simplest model is that of a specular (δ-function) barrier with a ferromagnet/superconductor Fermi velocity mismatch. Here we need to take into account, in addition to the (v_x/v) factor, a finite barrier transparency. It depends on the Fermi velocities [6],

$$D = \frac{v_{xf}v_{xs}}{(v_{xf} + v_{xs})^2/4 + W_i^2}, \tag{6}$$

where $v_{s(f)}$ is the Fermi velocity in the superconductor (ferromagnet), and W is the strength of the barrier, $V(x) = W\delta(x)$ (for a one-band isotropic material W is related to the parameter Z of Ref. [7] as $Z = W/v_F$). Tunneling current is thus proportional to

$$\int_{v_{xf}>0} Dv_{xf}\,\frac{dS_F}{v_f} \propto \int_{v_{xf}>0} \frac{v_{xf}^2}{(v_{xf} + v_{xs})^2/4 + W^2}\,\frac{dS_F}{v_f}. \tag{7}$$

In the large W limit this reduces to $\langle Nv^2\rangle$ and the measured DSP is P_{Nv^2}. However, in the high transparency limit one cannot give a simple answer.

It was recently suggested [8] that Andreev reflection at the interface between a FM and a superconductor (SC) can be used for direct probing of DSP. The idea is simple: the Andreev reflection can be visualized as two currents of electrons with the opposite spins flowing inside the normal metal towards its interface with a SC. At the interface (more precisely, within the coherence length from the interface) the two currents recombine creating the current of Cooper pairs. In a paramagnet (or antiferromagnet with time reversal symmetry) both currents are the same, so one observes in the superconducting state the 100% increase in the net current over the normal state. de Jong and Beenakker [8] suggested that in a FM the total Andreev current is defined by that spin channel where the normal-state current is smaller, because the excess electrons in the other channel will not find partners to form pairs with. This was quantified in Ref. [8] via the number of spin-up and spin-down conductance channels, which they denoted as $N_{\uparrow(\downarrow)}$, thus arriving at an expression for the ratio of the current in the superconducting and the normal state as

$$I_s/I_n = 4\min(N_\uparrow, N_\downarrow)/(N_\uparrow + N_\downarrow). \tag{8}$$

The number of conductance channels cannot be directly evaluated. Besides, this expression can mislead the reader into a belief that the DSP measured through Andreev reflection is P_N (i.e., defined by the DOS).

Andreev reflection at a FM/SC contact has been recently attracting substantial theoretical interest [9]. This interest so far concentrated on such aspects as the symmetry of the order parameter, Fermi velocity mismatch, and generalization of the so-called BTK formula [7] onto spin-polarized case [2,9]. In terms of electronic structure, however, all the work was limited to the parabolic bands and/or spherical Fermi surface model. While revealing important fundamental physics, such an approach is of limited practical importance, because in real materials this approximation is unacceptable. In this Letter we, on the other hand, focus on the band structure effects in spin polarization, and in this context we need a better definition for the "number of conductance channels."

Comparing Eq. (8) with Eqs. (5) and (7), we observe that the DSP for Andreev reflection should be defined as either P_{Nv^2}, for a large barrier and/or diffusive current, or P_{Nv}, for low resistance ballistic contacts. This is, however, only the first approximation, while full expressions should include Fermi surface averages of more complicated functions of v_F. For one particular case, a fully ballistic (Sharvin) Andreev reflection, an explicit formula, reflecting the physics suggested by de Jong and Beenakker, can be derived, which is both suitable for band structure calculations and also quite illustrative.

de Jong and Beenakker treated incoming electrons and reflected holes as two separate currents. In purely ballistic regime, however, one has to take into account energy and momentum conservation (parallel to interface) for each reflected hole, so that P_{Nv}, with its independent averaging over each spin channel, does not necessarily correctly describe observable polarization. This may be

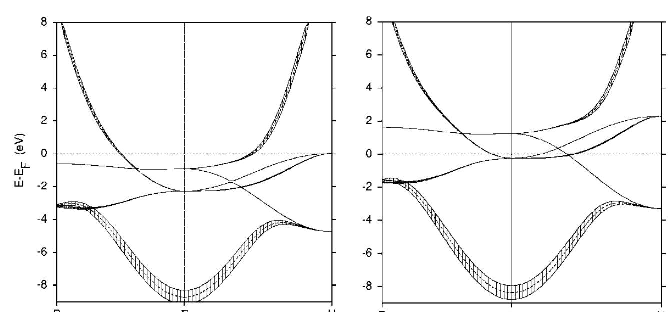

FIG. 1. Majority (left) and minority (right) spin band structure of Fe. The linewidth is proportional to the partial s character in each state ($E_F = 0$).

quantified as follows: Consider an incoming electron with a momentum $\mathbf{k} = (\mathbf{k}_\parallel, k_x)$, which is reflected as a hole with the momentum $\mathbf{q} = (\mathbf{q}_\parallel, q_x)$ in the other spin subband; $\mathbf{q}_\parallel = \mathbf{k}_\parallel$, $E_{\mathbf{k}\uparrow} = E_{\mathbf{q}\downarrow}$. This fixes for each $\mathbf{k}$ a countable number of wave vectors $\mathbf{q}$ satisfying this condition. For simplicity we will assume now that there is only one such $\mathbf{q}$ and will denote it $\tilde{\mathbf{q}}$. This adds an additional constraint to Eq. (5), so that instead we have, omitting summation over the band indices α,

$$I = \frac{e^2 UA}{(2\pi)^6} \int_{v_x > 0} \delta(\mathbf{q}_\parallel - \mathbf{k}_\parallel) \times \delta(q_x - \tilde{q}_x) v_x \delta(E_\mathbf{k})\, d^3k\, d^3q\,.$$

This can be rewritten in a symmetric way like

$$I = (2\pi)^{-6} e^2 UA \int_{v_x, u_x > 0} d^3k\, d^3q \times \int d^2\mathbf{a} [\delta(\mathbf{q}_\parallel - \mathbf{a}) \delta(\mathbf{k}_\parallel - \mathbf{a})] \times [u_x \delta(E_\mathbf{q})][v_x \delta(E_\mathbf{k})], \quad (9)$$

where we introduced a 2D vector $\mathbf{a}$, transformed the δ function of q into a δ function of $E_\mathbf{q}$, and introduced $\mathbf{u} = \partial E_\mathbf{q} / \partial \mathbf{q}$. We can further rewrite Eq. (9) as

$$I = e^2 UA \int d^2\mathbf{a}\, F_\uparrow(\mathbf{a}) F_\downarrow(\mathbf{a}) = (2\pi)^{-3} e^2 UAS\,, \quad (10)$$

where $F(\mathbf{a}) = (2\pi)^{-3} \int_{v_x > 0} \delta(\mathbf{k}_\parallel - \mathbf{a}) v_x \delta(E_\mathbf{k})$ is half (because $v_x > 0$) the number of crossings of the line $\mathbf{k}_\parallel = \mathbf{a}$ with the Fermi surface for a given spin. Using again projections of the Fermi surface for either spin onto the interface plane, one can define S as the overlap area of the spin-up and spin-down projections. This current can be expressed in terms of a spin polarization as in Eq. (8), thus giving yet another, "ballistic Andreev" definition of spin polarization, similar to, but not the same as, the "Nv" definition, $P_{\text{b.A.}} \neq P_{Nv}$ (they are equal only if the Fermi surface projection for one spin is entirely contained in that for the other spin).

So we observe that an experiment would probe different DSP's depending on the length scale of the problem, which is defined by the size of the contact and the length at which the voltage drops, and how it compares with the mean free path. The transparency of the barrier can also influence the measured DSP. In the pure ballistic limit the DSP is related to the average Fermi velocity, while in the purely diffusive regime it is defined by the average squared Fermi velocity [10]. One may ask why DOS is so often used as a measure of spin polarization, even though such definition is irrelevant for transport properties. The answer is that the most common way to perform tunneling or similar experiments is to follow the details of the contact conductance as a function of voltage. Probably the most spectacular and fruitful application of this technique is the tunneling spectroscopy of superconductors. In such a case the characteristic scale for the voltage change is the superconducting gap. The normal state electronic structure does not change over such a small energy range, so the only important factor is the variation of the superconducting DOS with energy. The normal state DOS and velocity can be assumed constant and factored out. Of course, it is not the case when two different sheets of the Fermi surface, as in Ref. [6], or two different spin channels, are compared (cf. Figs. 1–3).

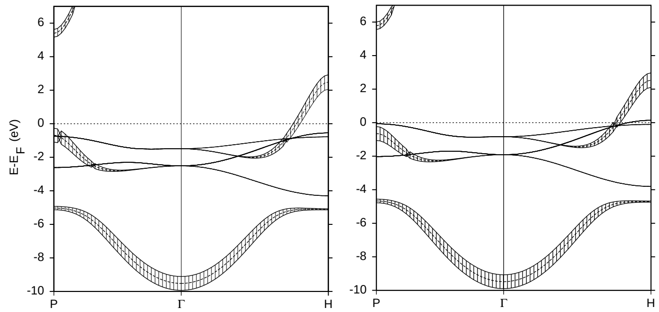

FIG. 2. The same as Fig. 1, for Ni.

Importantly, P_{Nv^2}, P_{Nv}, and P_N are entirely different in real materials (Figs. 4 and 5). The reason is (and the "s-DOS recipe" works for the same reason) that in transition metals one can often distinguish the pieces of the Fermi surface that are predominantly d in character and the pieces that are mostly s. The former have low velocity and are responsible for most of the DOS [Eq. (1)]. The latter have high velocity and provide the main contribution to Eq. (3). The larger the anisotropy of the Fermi velocity (angular anisotropy, in principle, works in the same way as interband one), the larger is the difference between P_{Nv^2}, P_{Nv}, and P_N.

To illustrate this we present here linear muffin-tin orbitals local spin density approximations (LSDA) calculations of the corresponding quantities in Fe and Ni. It is instructive to start from the band structure itself (Figs. 1–3). The "fat" bands in these figures correspond to the states with substantial sp character. One immediately

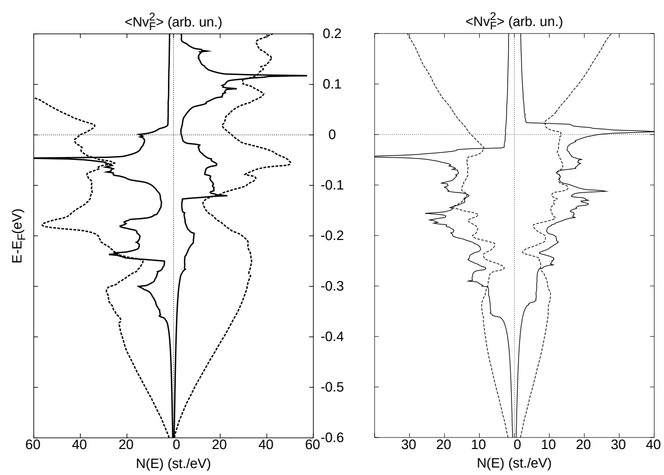

FIG. 3. Density of states, $N(E)$ (solid line) and $\left(\frac{n}{m}\right)_{\text{eff}}(E) = \langle N v_F^2 \rangle (E)$ (dashed line), for Fe (left panel), and Ni (right panel).

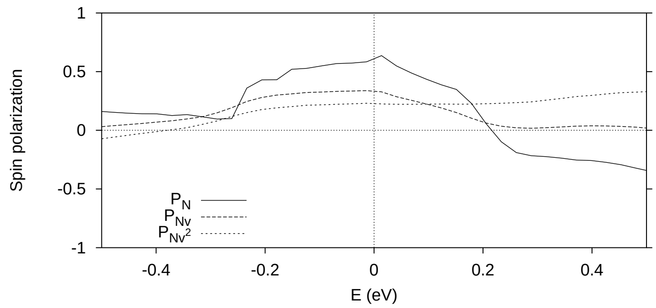

FIG. 4. Degree of spin polarization for Fe, calculated as P_N, P_{Nv}, and P_{Nv^2}.

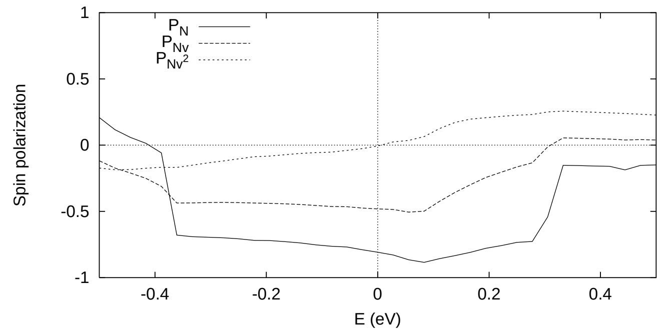

FIG. 5. The same as Fig. 4, for Ni. Note the kinks in P_N at $E - E_F \simeq \pm 0.35$ eV, which correspond to the top of the d bands in the two spin channels (cf. Fig. 3).

notices a qualitative difference between Fe and Ni: In the former all bands in both spin channels are heavily hybridized at the Fermi level, and one cannot convincingly classify bands as "predominantly sp" and "predominantly d." In the latter the d band is so deep that one can single out an sp-like pocket in the spin-up channel (in Fig. 2, the band crossing Fermi level between Γ-H), and d-like pockets (in Fig. 2, near H). The Fermi surface in the spin-down channel is entirely sp-like. This is similar to paramagnetic Pd [11], which is the only 4d metal where transport properties can be described by the $s - d$ scattering model.

In general a large P_N may be due to any of the two reasons: either the *areas* of the up- and down-Fermi surfaces are different, while velocities may be similar, or the areas are not too different, but the Fermi *velocity* for one spin channel is much smaller than for the other. In the former case the additional factors of v_F or v_F^2 may change the DSP somewhat, but qualitative changes, or, in an extreme case, the sign change, are unlikely. If, however, "light" and "heavy" electrons are present, P_N is dominated by the heavy pockets, and P_{Nv^2} by the light ones, so the two DSP's are likely to be very different and possibly have opposite signs.

The first situation is realized in Fe. One indeed can see that $N(E)$ and $\langle N v_F^2 \rangle (E)$ behave similarly (Fig. 3). Correspondingly, the difference between different DSP's is only moderate (Fig. 4). On the other hand, in Ni the DSP essentially drops to zero when the factor v_F^2 is included. Interestingly, most experimental results [1] indicate that the DSP observed in tunneling is *positive* and not too small ($>$20); in other words, the effect described above appears to be even stronger in reality than in band structure calculations. This is also to be expected: LSDA has a tendency to underlocalize d electrons. For instance, in Cu the fully occupied d band appears in the calculations about 0.4 eV higher than in experiments, and is also too wide. Similarly, LDA underlocalization of the d electrons in Ni leads to an overestimation of the d bandwidth and of the exchange splitting (by approximately a factor of 2). As a result, the separation of carriers into sp-like and d-like in Ni should be even more pronounced compared to LSDA calculations, and thus the effect of Fermi velocity on DSP even stronger. This leads, in turn, to the DSP sign reversal, observed in tunneling experiments. In the calculation, a similar situation occurs at $E - E_F \gtrsim 0.1$ eV.

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