Spin Caloritronics and the Thermomagnetoelectric System

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Work performed with
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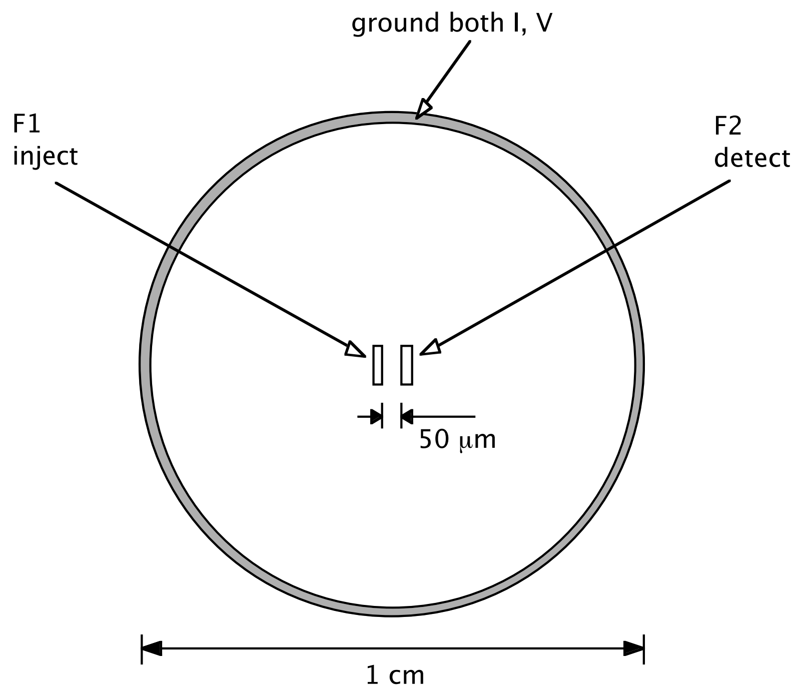
Outline

- Introduction - a little history
- Entropy production derivation
- Thermodynamic equations of motion
  * Bulk and interface
- Entropy currents
  * Spin relaxation in N, near F / N interface
  * In external field H, generate either heating or cooling
- Summary
- Appendix - spin transport across F / N interface

Some history: First attempts at spin injetion

- I started working in Silsbee's lab in summer 1980. He had worked out the theory of spin injection, accumulation and detection, and had sketched out the expected line shape for the Hanle effect.
- Experimentally, we had no photo-lithography. Silsbee (correctly) estimated the spin diffusion length in films as $\delta_S \sim 100$ nm, too short for our techniques. So we worked with bulk, foil samples ($\delta_S \sim 100$ μm at low T) and used shadow masks to fabricate the F injector and detector films.
- We used a variation of the Corbino disk geometry to minimize background magnetoresistance.
- After several years of attempts, we could not eliminate a quadratic background resistance, and never observed any reproducible signatures of spin injection.



One variation of Corbino disk geometry
Top View of foil sample

Junction Thermoelectric Effect

- Around 1983, Cornell EE Dept. developed a photo-lithography facility that was made available to other departments. I started developing recipes for lithographic processing on bulk foils.
- In late 1983, Silsbee was concerned about the lack of publications for this project. We had documented an unusual quadratic I-V characteristic in the Corbino disk samples and Silsbee developed a model for a "junction thermoelectric effect"

$$\begin{pmatrix} I_q \\ I_Q \end{pmatrix} = \begin{pmatrix} L_{11} & L_{12} \\ L_{21} & L_{22} \end{pmatrix} \begin{pmatrix} \Delta V \\ \Delta T \end{pmatrix}$$

- The idea: given a low transmission barrier between two metals, with a high enough resistance to support a temperature difference ΔT across the junction.
- If the transmission of the barrier is temperature dependent (even weakly), then there would be a thermoelectric effect across the barrier.
- He believed this to be a novel idea.

Foiled by Tinkham

PHYSICAL REVIEW B VOLUME 22, NUMBER 9 1 NOVEMBER 1980

New thermoelectric effect in tunnel junctions

A. D. Smith,* M. Tinkham, and W. J. Skocpol
Department of Physics and Division of Applied Sciences, Harvard University, Cambridge, Massachusetts 02138
(Received 23 June 1980)

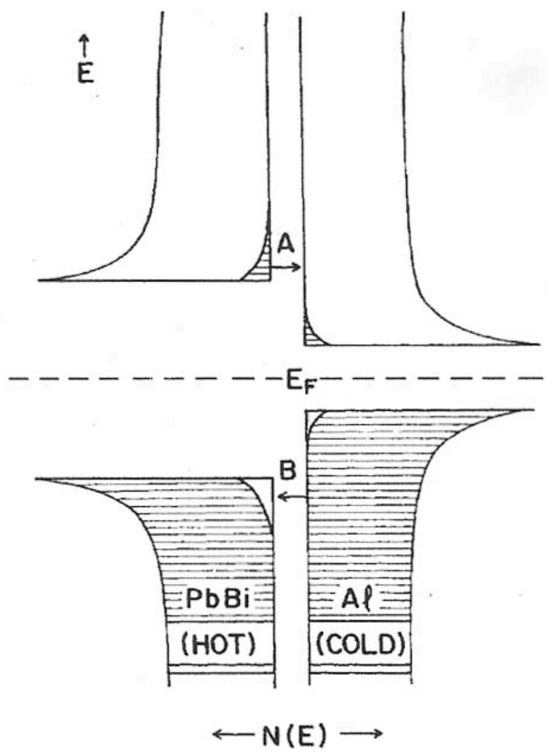


FIG. 1. Semiconductor model of tunneling, shown for an Al-PbBi junction illuminated on the PbBi side.

- I went to the library and did a literature search. In those pre-internet days, large hard copy citation volumes were published quarterly.
- I found a relevant article by Tinkham et al.
- Showing the article to Silsbee, he immediately recognized it as describing the same junction TE effect. Thus, it was no longer a novel (publishable) idea.

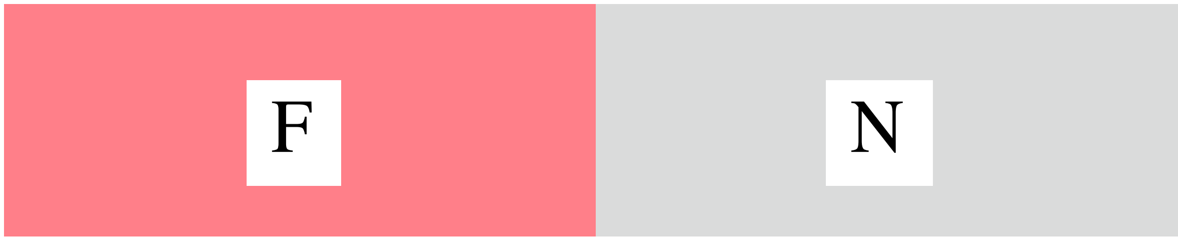
Genesis of the manuscript

- After a long silence, I cautiously ventured a remark:
- MJ: "What about the other terms?"
- RHS: "What do you mean?"
- MJ: "Aren't there terms for the spin injected current?"

$$\begin{pmatrix} I_q \\ I_Q \\ I_M \end{pmatrix} = \begin{pmatrix} L_{11} & L_{12} & L_{13} \\ L_{21} & L_{22} & L_{23} \\ L_{31} & L_{32} & L_{33} \end{pmatrix} \begin{pmatrix} \Delta V \\ \Delta T \\ \Delta - H^* \end{pmatrix}$$

- After another silence, Silsbee started sketching out a derivation. He required all his students to do some theory, so I read Callen and began working on the derivations he outlined. He quickly decided to include derivations for thermodynamic equations in bulk materials generalized to include spin.
- By mid 1984 I began to focus on experiments using foil samples and the linear nonlocal geometry, and the "Onsager" manuscript was put on a back burner.
- The Appendix was Silsbee's idea. It never occurred to me that there ever would be interest in the details of transport at the F/N interface.

Thermodynamic Approach: Deriving equations of motion for charge and spin



- **Approach:** develop thermodynamic equations of motion from "entropy production"
- Derive these equations in the bulk in F and N; and for the F-N interface
- Apply boundary conditions
  - * J_q is continuous at F-N interface
  - * J_M is continuous if there is NO interfacial spin-flip scattering
    - Interface may have spin asymmetry **(introduce η)**
- "A Thermodynamic Analysis of Interfacial Transport and of the Thermomagnetoelectric System," M. Johnson and R.H. Silsbee, PRB **35**,4959 (87)

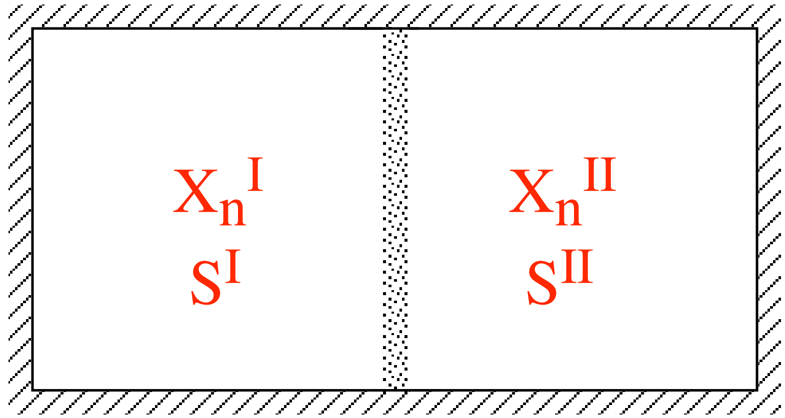
Entropy Production Calculation

Total entropy $S^0 = S^I + S^{II}$

Variable X_n = charge, energy

J_n = flux of X_n

Calculate entropy production associated with transport:



X_n flows until S^0 is maximized

$$\dot{S} = \frac{dS^0}{dt} = \sum_n \frac{\partial S^0}{\partial X_n} \frac{dX_n}{dt} = \sum_n F_n J_n$$

F_n is the *affinity*, or *generalized force*, associated with variable X_n

$$\dot{S} = \Delta\left(\frac{1}{T}\right) I_Q - \frac{1}{T} \Delta V \;\; I_q$$

… for the thermoelectric system.

Linear dynamic equations of motion

Expand J_n and keep 1st order terms (Linear Response):

$$J_n = \sum_m L_{mn} F_n$$

The L_{mn} are kinetic coefficients.

$$-I_q = \frac{L_{11}}{T} \Delta V + L_{12}\ \Delta\left(\frac{1}{T}\right)$$

$$I_Q = \frac{L_{21}}{T} \Delta V + L_{22}\ \Delta\left(\frac{1}{T}\right)$$

… for the thermoelectric system.

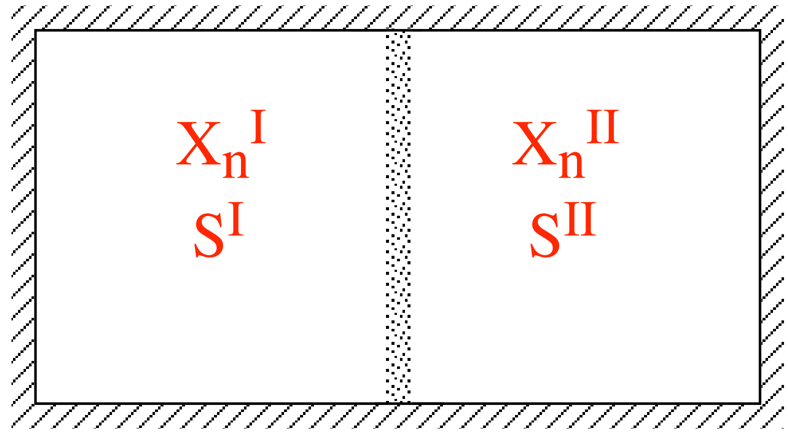
Entropy Production Calculation

Total entropy $S^0 = S^I + S^{II}$

X_n = charge, energy, moment

J_n = flux of X_n

Calculate entropy production associated with transport:



$$\dot{S} = \Delta\left(\frac{1}{T}\right) I_Q - \frac{1}{T}\Delta V \; I_q - \frac{1}{T}\Delta\left(-H^*\right) I_M$$

where $$\left(-H^*\right) \equiv \frac{\tilde{M}}{\chi} - H$$ is the generalized force associated with noneq. spin

… for the **thermomagneto-electric** system.

New linear dynamic equations of motion...

… for the thermomagneto-electric system.

$$-I_q = \frac{L_{11}}{T}\Delta V + L_{12}\,\Delta\left(\frac{1}{T}\right) + \frac{L_{13}}{T}\Delta\left(-H^*\right)$$

$$I_Q = \frac{L_{21}}{T}\Delta V + L_{22}\,\Delta\left(\frac{1}{T}\right) + \frac{L_{23}}{T}\Delta\left(-H^*\right)$$

$$-I_M = \frac{L_{31}}{T}\Delta V + L_{32}\,\Delta\left(\frac{1}{T}\right) + \frac{L_{33}}{T}\Delta\left(-H^*\right)$$

Johnson and Silsbee, PRB **35**, 4959 (87)

Thermodynamic equations of motion

Eq. (1)

$$\begin{pmatrix} I_q \\ I_Q \\ I_M \end{pmatrix} = -G \begin{pmatrix} 1 & \frac{k_B^2 T}{e\varepsilon} & \frac{\eta\beta}{e} \\ \frac{k_B^2 T^2}{e\varepsilon} & \frac{a k_B^2 T}{e^2} & \eta' \frac{\beta}{\varepsilon}\left[\frac{k_B T}{e}\right]^2 \\ \frac{\eta\beta}{e} & \eta' \frac{\beta T}{\varepsilon}\left[\frac{k_B}{e}\right]^2 & \xi \frac{\beta^2}{e^2} \end{pmatrix} \begin{pmatrix} \Delta V \\ \Delta T \\ \Delta(-H^*) \end{pmatrix}$$

At the interface

In the Bulk

$$\begin{pmatrix} J_q \\ J_Q \\ J_M \end{pmatrix} = -\sigma \begin{pmatrix} 1 & \frac{a'' k_B^2 T}{e E_F} & p\frac{\beta}{e} \\ \frac{a'' k_B^2 T^2}{e E_F} & \frac{a' k_B^2 T}{e^2} & p' \frac{\beta}{E_F}\left[\frac{k_B T}{e}\right]^2 \\ p\frac{\beta}{e} & p' \frac{\beta T}{E_F}\left[\frac{k_B}{e}\right]^2 & \zeta \frac{\beta^2}{e^2} \end{pmatrix} \begin{pmatrix} \nabla V \\ \nabla T \\ \nabla(-H^*) \end{pmatrix}$$

Eq. (2)

$\xi,\ \zeta = 1$

Spin Caloritronics

- The most important experiments in Spin Caloritronics have used ferromagnets [e.g. S. Maekawa et al., Nature **455**, 778 (2008)]. This ground-breaking work continues.

- Effects also can be expected in Ferromagnet / Nonmagnet (F / N) systems [refer to M. Johnson, Solid State Commun. **150**, 543 (2010)].
- Consider an F / N system with interface at x = 0. Both F and N have thickness larger than a spin diffusion length, $\delta_{S,F}$ in F and $\delta_{S,N}$ in N. A spin polarized current J_M accompanies a bias current J_e from F to N. Nonequilibrium magnetization $\tilde{M}_N$ ($\tilde{M}_F$) is created in N (F) within distance $\delta_{S,N}$ ($\delta_{S,F}$) from the interface.

Entropy current

The change of entropy associated with internal, irreversible change of magnetic moment $\tilde{M}_n$ resulting from spin relaxation in N is

$$dS = \frac{-1}{T}(-H^*)d\tilde{M} = \frac{-1}{T}\left(\frac{\tilde{M}_n}{\chi} - H\right)d\tilde{M}_n$$

and the associated entropy current is

$$J_s = \frac{-1}{T}\left(\frac{\tilde{M}_n}{\chi} - H\right)J_M$$

First term of two

The magnitude of spin accumulation in sample volume "Vol" is:

$$\tilde{M}_n = \frac{J_M T_2}{\mathrm{Vol}} = \frac{\eta \mu_B T_2}{\delta_s \; e} J_e \; e^{-x/\delta_s}$$

where the interface is given unit area and the third dimension of "Vol" is taken to be the spin depth.

The first term is independent of external field,

$$J_s(x) = -\frac{T_2}{T\chi\delta_s}\left(\frac{\eta\mu_B}{e}\right)^2 e^{-2x/\delta_s} \; J_e^2$$

First term - like Joule heating

Using the continuity equation for conservation of entropy production and flow,

$$(\partial s/\partial t) + (\partial J_s/\partial x) = \dot{s}$$

we have $(\partial s/\partial t) = 0$ and therefore $\dot{s} = (\partial J_s/\partial x)$. Integrate from 0 to ∞ to get the total entropy production in the N region and then find $\dot{Q} = T\dot{s}$:

$$\dot{Q} = \frac{T_2}{\chi\delta_s}\left(\frac{\eta\mu_B}{e}\right)^2 \; J_e^2$$

Note that $\dot{Q} \propto J_e^2$, similar to joule heating power.

Second term - linear with H

Consider next the second term, proportional to field H:

$$J_s = \frac{H}{T} J_M = \frac{H}{T} \frac{\eta \mu_B}{e} J_e \, e^{-x/\delta_s}$$

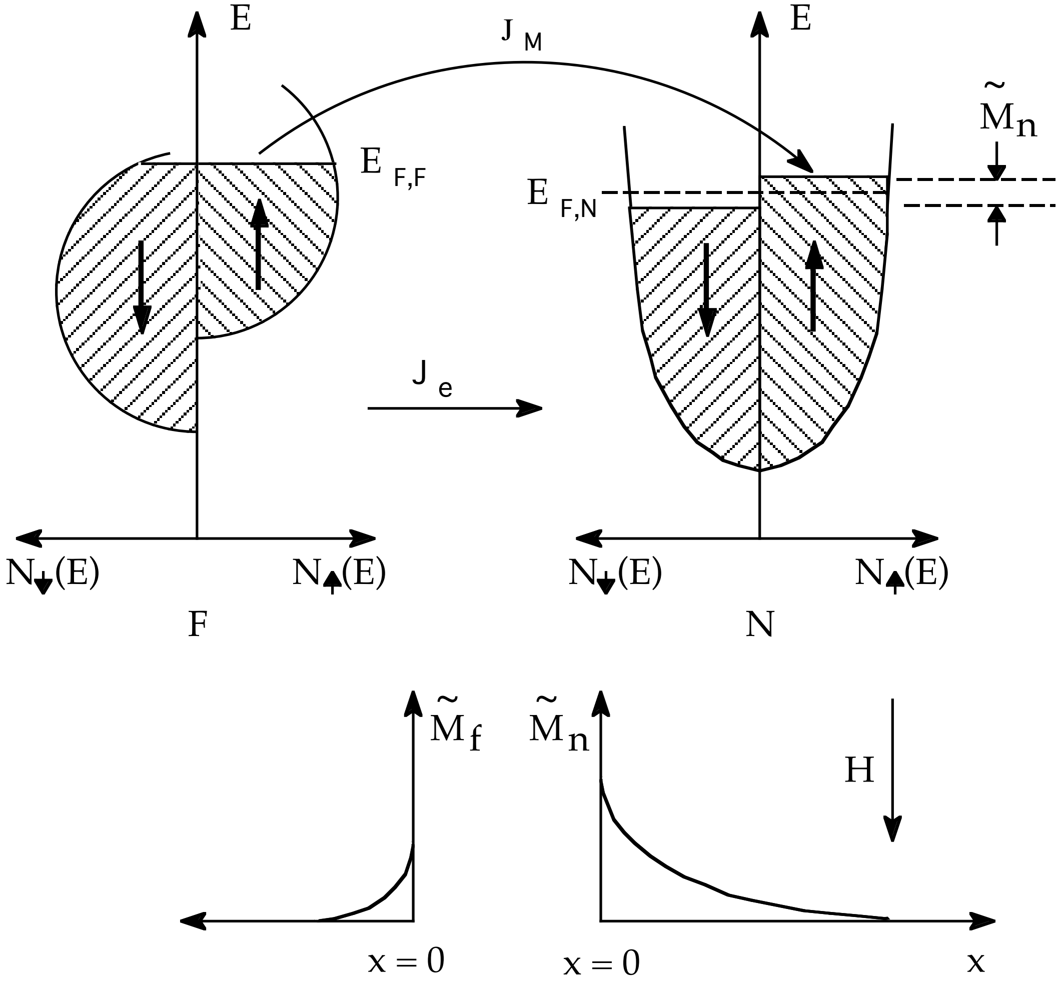
Solving for $\dot{s}$, integrating from 0 to ∞ in N, and finding $\dot{Q} = T\dot{s}$ gives

$$\dot{Q} = \frac{H \eta \mu_B}{e} J_e$$

Note that $\dot{Q}$ is independent of T and is proportional to H

Further note that η and H may be positive or negative. For a given choice of the sign of H, the heat production associated with spin relaxation could be positive or negative. Recall that polarization factor η is defined as the relative difference between the partial currents of the spin subbands.

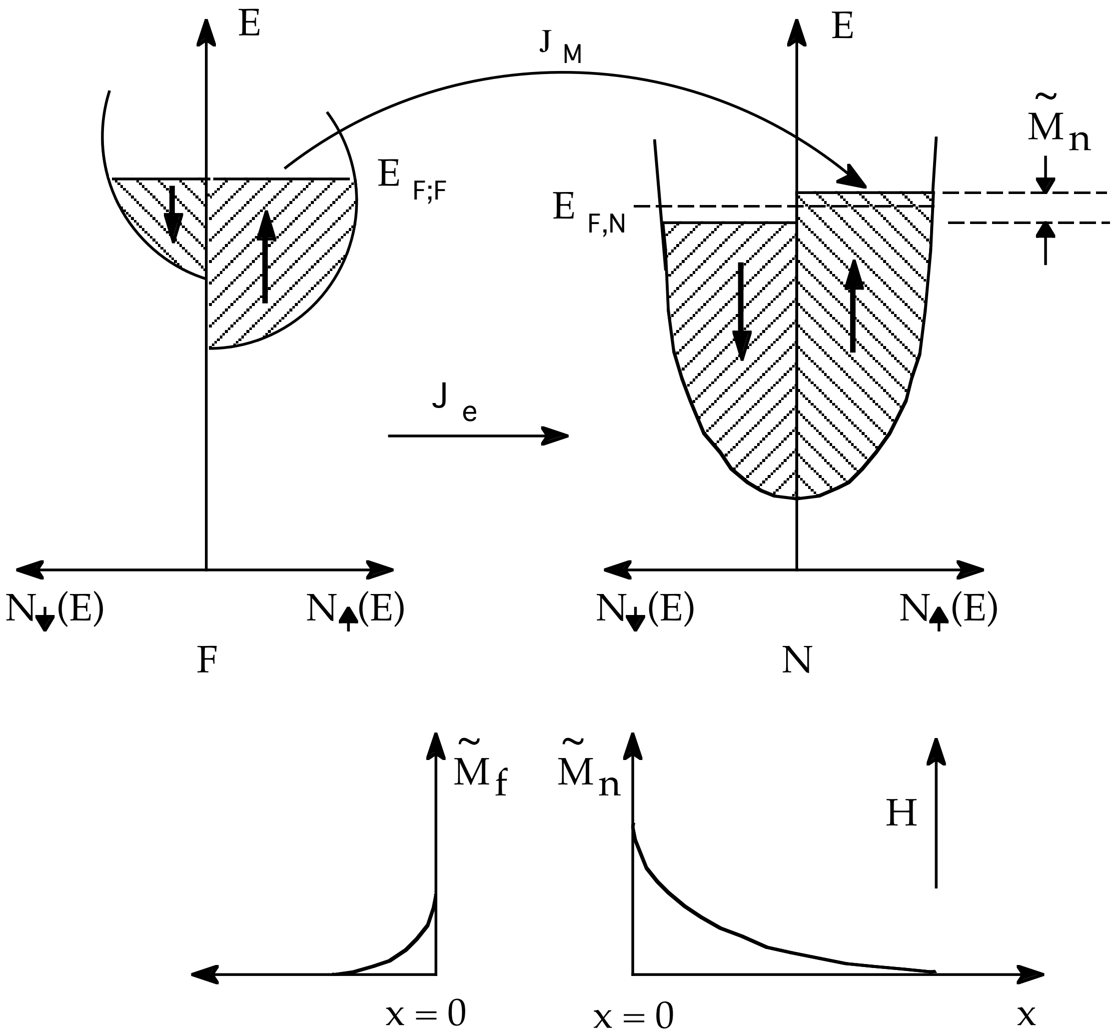
Heating by spin relaxation



(a)

- **The minority spin subband in F dominates**
- η is positive, spin accumulation is positive
- External field - |H| results in M_0 along downspin direction
- Field is antiparallel with noneq'm spins
- System has lower energy after relaxation to equilibrium
- Therefore: Spin relaxation results in heating

Cooling by spin relaxation



- **The majority spin subband in F dominates**
- η is negative, spin accumulation is positive
- External field + |H | results in M_0 along downspin direction
- Field is parallel with noneq'm spins
- System has lower energy after relaxation to equilibrium

Therefore: Spin relaxation results in cooling

For experimentalists:

- By choosing appropriate ferromagnetic spin injector, spin relaxation in presence of an external magnetic field may generate either heating or cooling
- Must worry about the other (Joule) term - second term must dominate
- Experimental: alternate layers in a multilayer sandwich to amplify effect
- NOTE: The converse effect also should exist. Heating (or cooling) should generate spin accumulation. Such effects may be relevant to spin torque switching [Hatami, Bauer, Zhang and Kelly, PRL **99**, 066603 (07)], which should be called Slonczewski - Berger switching.

Summary

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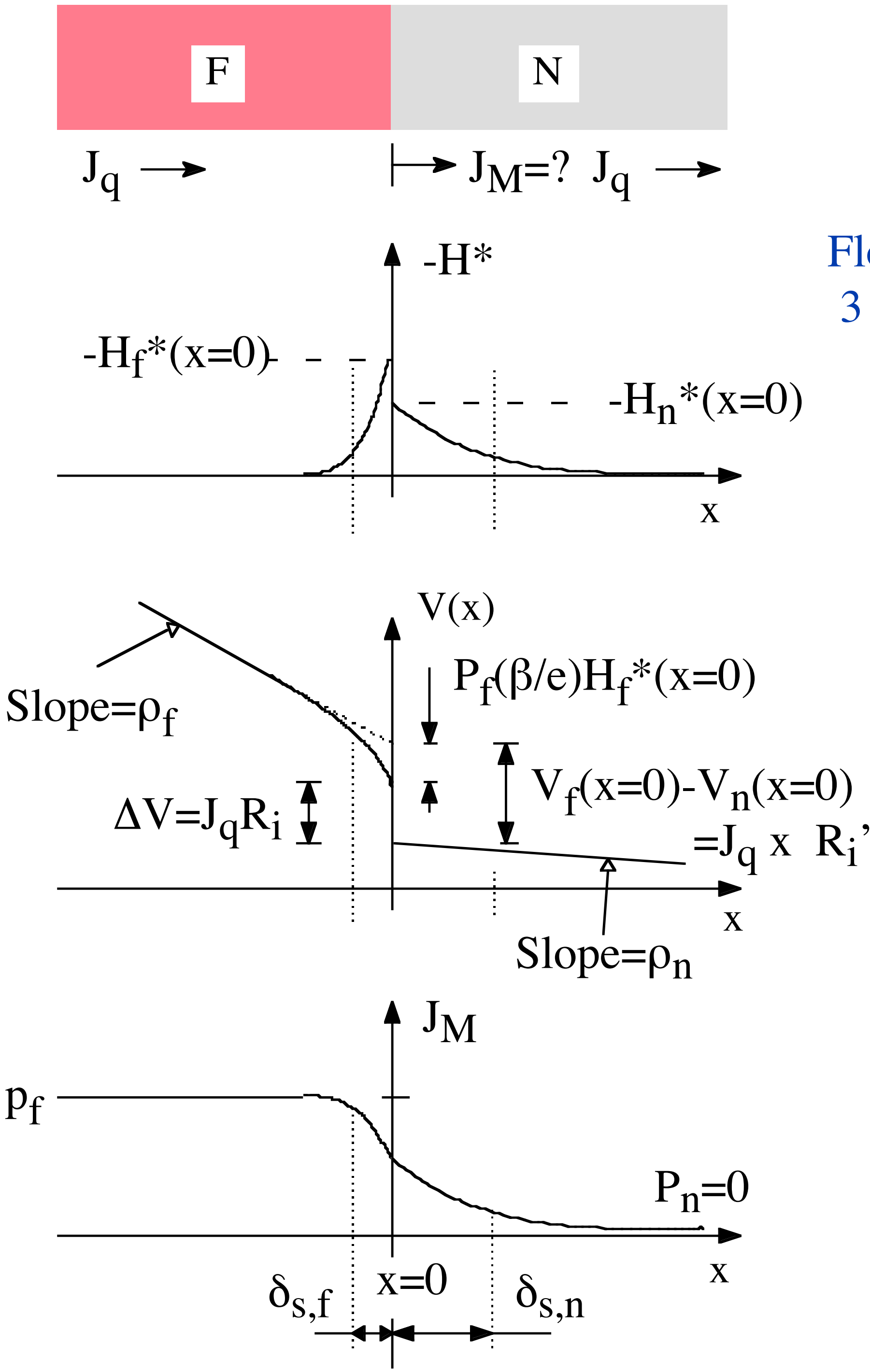


PRB (87) Appendix

Johnson & Silsbee, PRB **35**,4959 (87)

Flow of Spin and Charge across F/N Interface
3 - dim model (choose unit values for y, z ->
1 dim model) - **general case**

- Away from the interface -

p_f : polarization of J_M in F

$p_n = 0$: polarization of J_M in N

- Near the interface -

$-H_n^* = (M^{\sim}/\chi)_n - H$

$-H_f^* = (M^{\sim}/\chi)_f - H$

$\delta_{s,f}$ = spin diffusion length in F

$\delta_{s,n}$ = spin diffusion length in N

$\beta = \mu_B$ = Bohr magneton

R_i : intrinsic interface resistance, 1/G

$r_f = \rho_f\, \delta_f$ (= R of 1 spin depth of F)

$r_n = \rho_n\, \delta_n$ (= R of 1 spin depth of N)

Spin accumulation in N flows back across F-N interface

General Case: Solution

$$J_M = \left[\eta\left(\frac{\beta}{e}\right)J_q\right]\frac{1+\left(\frac{p_f}{\eta}\right)\left(\frac{r_f}{R_i}\right)\left(\frac{1-\eta^2}{1-p_f^2}\right)}{1+\left(1-\eta^2\right)\left[\left(\frac{r_n}{R_i}\right)+\left(\frac{r_f}{R_i}\right)\left(\frac{1}{1-p_f^2}\right)\right]} \qquad \text{Eq. (3)}$$

Some typical values (for metals):

- $R_i = R_c \sim 10^{-9}$ Ω-cm^2 ; K. Bussman et al., IEEE Trans, Mag. **34**, 924 (98) (N-N interface, lithographically processed samples)
- $r_f \sim 2\text{x}10^{-5}$ Ω-cm x $5\text{x}10^{-7}$ cm = 10^{-11} Ω-cm^2 (F film)
- $r_n \sim 2\text{x}10^{-6}$ Ω-cm x $1\text{x}10^{-4}$ cm = $2\text{x}10^{-10}$ Ω-cm^2 (N film)
- In general, all terms in Eq. (3) are important

E.I. Rashba, Phys. Rev. B **62**, 16,267 (2000)

$$J_M \equiv \eta^* \left(\frac{\beta}{e}\right) J_q = \left[\eta\left(\frac{\beta}{e}\right) J_q\right] \frac{1 + G\left(\frac{p_f}{\eta}\right)\left(\frac{\delta_f}{\sigma_f}\right)\left(\frac{\xi - \eta^2}{\zeta_f - p_f^2}\right)}{1 + G\left(\xi - \eta^2\right)\left[\left(\frac{\delta_n}{\sigma_n \zeta_n}\right) + \left(\frac{\delta_f}{\sigma_f}\right)\left(\frac{1}{\zeta_f - p_f^2}\right)\right]}$$

JS PRB 87 Eq. (A11)

$$\gamma = \frac{\left[r_F\left(\Delta\sigma / \sigma_F\right) + r_C\left(\Delta \sum / \sum\right)\right]}{r_F + r_N + r_C}$$

E.I.R. PRB 00 Eq. (18)

$$\eta^* \equiv \gamma$$

$$p = \frac{\Delta\sigma}{\sigma}$$

$$\eta = \frac{\Delta\sum}{\sum}$$

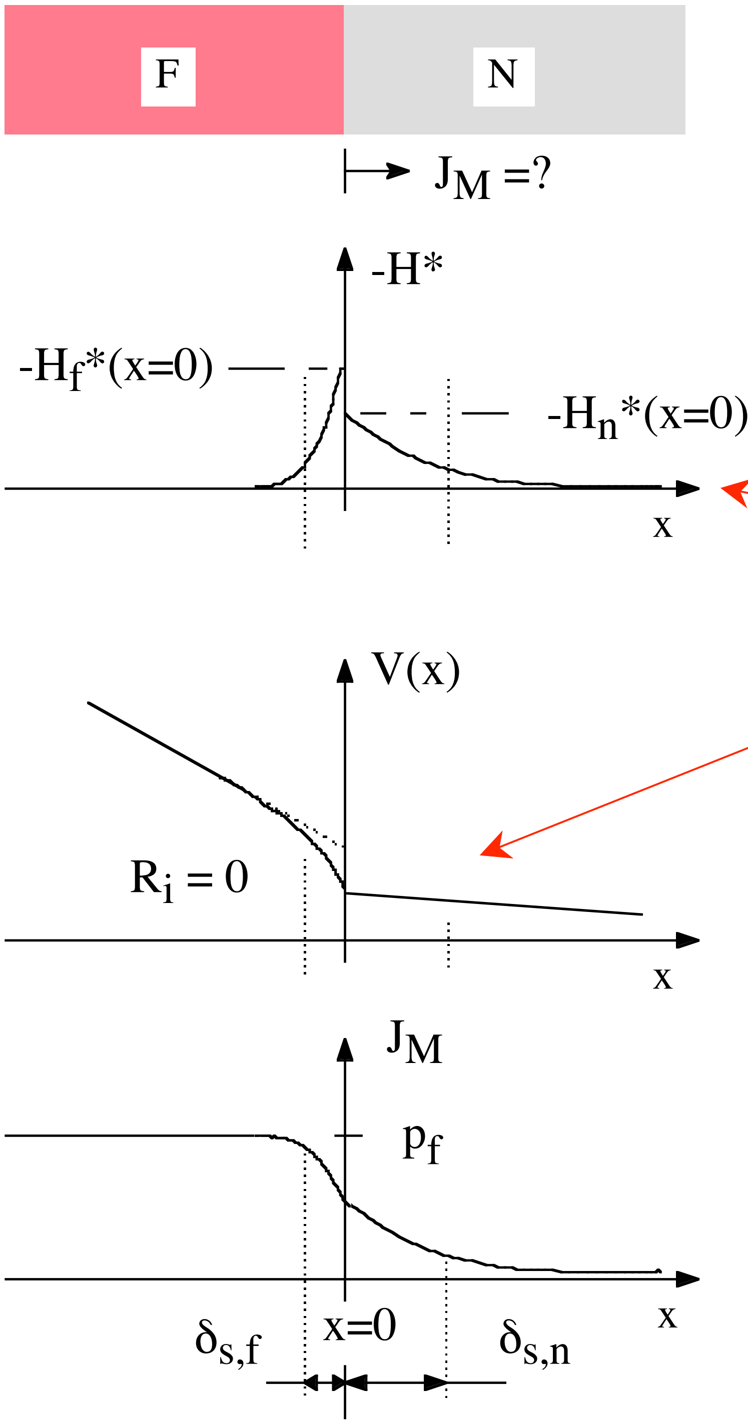
$$G\left(\xi - \eta^2\right) = 1 / r_C$$

$$\frac{\delta_n}{\sigma_n \zeta_n} = r_N$$

$$\frac{\delta_f}{\sigma_f\left(\zeta_f - p_f^2\right)} = r_F$$

Rashba result agrees with JS general result

Private communication from E.I. Rashba, 5/2/02



Special Case 1

- $R_i = 0$ (infinite G)
- Low diffusion and long relaxation in N
- Spin pileup (accumulation) in N; backflow into F
- "apparent" interface resistance (from F)
- J_M reduced by backflow
- Solution is ….(next page)

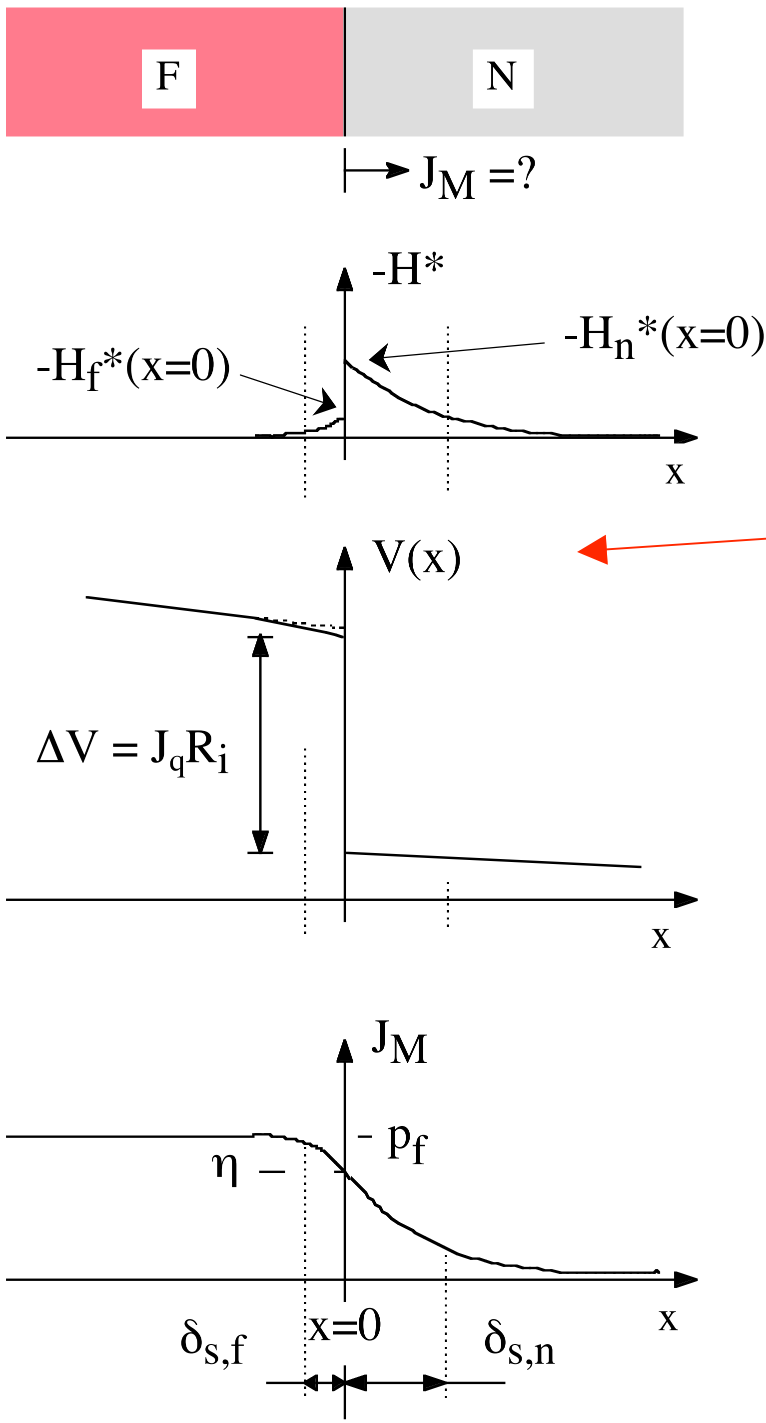
CASE 1: Solution

$$J_M = p_f \left(\frac{\beta}{e}\right) J_q \frac{1}{\left[1 + \left(\frac{r_n}{r_f}\right)\left(1 - p_f^2\right)\right]} \qquad \text{Eq. (5)}$$

$$P = \left(p_f\right) \frac{1}{\left[1 + \left(\frac{r_n}{r_f}\right)\left(1 - p_f^2\right)\right]} = p_f \left(\frac{1}{1 + M}\right)$$

"mismatch" factor M

- Fractional polarization efficiency is reduced from the polarization in F because of the **resistance mismatch** at the F/N interface; Physics - reduction comes from **backflow** of spins, back into F



Consider an interface resistance between F and N

CASE 2

- R_i large (G -> 0)
- R_i need not be a perfect tunnel barrier (but η depends on interface)
- V(x) is drawn on different scale (note that slopes look flatter)
- H_f^* is small because the barrier blocks back diffusion; weak coupling between F and N
- Interface dominates spin transport
- η involves spin asymmetry (spin-dependent transmission) of interface, $\eta = p_f$ but note $J_{M,f} = J_{M,n}$ (cont'ous)
- $J_M = \eta(\beta/e)J_q$; P = η Eq. (6)

Therefore ….

When $R_i > r_f , r_n$, interfacial spin transport is determined by η, and spin injection from a ferromagnetic metal electrode to a 2DEG is possible.

Schmidt et al. [PRB **62**, 4790 (2000)] noted that resistance mismatch can be a dominant effect for an interface between a ferromagnetic metal and a semiconductor. But they neglected the interface resistance.

Rashba [PRB **62**, 16,267 (2000)] noted that a tunnel barrier can mediate the mismatch effect.

However, it's not necessary that $R_i \rightarrow \infty$ nor to have a perfect tunnel barrier. The key requirement is $R_i > r_f , r_n$