Boundary conditions for the nonequilibrium magnetization of conduction electrons at tunnel junctions

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(Submitted 10 May 1983)
Zh. Eksp. Teor. Fiz. **86**, 187–192 (January 1984)

Boundary conditions for the nonequilibrium magnetization of conduction electrons at the junction of two metals are derived from a model tunnel Hamiltonian. The magnetization fluxes across the interface depend on the electronic properties of metals in the same way as the tunnel current. The spectral characteristics of conduction-electron spin resonance in a bimetallic plate are calculated. It is indicated that the method of conduction-electron spin resonance might be used to study superconductors and metals having a short spin-relaxation time.

1. INTRODUCTION

Experiments have recently been reported on the conduction-electron spin resonance in bimetallic systems.[1–5] When electrons tunnel across an interface they mix the nonequilibrium magnetizations of the metals; the mixing is seen experimentally as changes in the shape, width, and g-factor of the resonance line. These effects are customarily described by introducing phenomenological boundary conditions[1,6]

$$-D_i\mathbf{n}_i\nabla m_i^+(\mathbf{r},\omega)\big|_{\mathbf{r}=\mathbf{r}_s}=\sum_{j=1}^{2} b_{ij}m_j^+(\mathbf{r}_s,\omega), \qquad (1)$$

which relate the nonequilibrium magnetization flux m_i^+ ($i=1,2$) across the interface $\mathbf{r}=\mathbf{r}_s$ to the values of m_i^+ at the interface through certain coefficients b_{ij}. In (1), D_i is the diffusion coefficient of the conduction electrons, and $\mathbf{n}_i$ is the normal to the interface directed away from the i-th metal. By solving Maxwell's equations and the Bloch equations for the metals along with (1), one can calculate the spectrum of conduction-electron spin resonance and then compare the results with experiment to determine the values of b_{ij} for various pairs of metals: Li-Cu (Refs. 1–3), Cu-Ni and Cu-Fe (Ref. 4), and Cu-Nb (Ref. 5). Graham and Silsbee[5] have observed that the coefficients b_{ij} decrease sharply after the niobium becomes superconducting. At present we lack a suitable theoretical interpretation of a temperature dependence of this sort.

A question which arises in connection with these experiments is that of the microscopic "structure" of the coefficients b_{ij}. In other words, what information can we obtain from measurements of b_{ij} for different junctions? It is clear that the coefficients b_{ij} must depend not only on the properties of the interface itself but also, and strongly, on the electronic spectrum of the metals themselves.

In this paper we offer a microscopic derivation of boundary conditions (1). It turns out that the b_{ij} depend directly on the electronic structure of each of the metals, in much the same way as the tunnel current across the junction does. We show that this circumstance adds substantially to the list of materials (superconductors and metals with a short spin-relaxation time) whose physical properties might be studied by the method of conduction-electron spin resonance across a tunnel junction, through the use of one of the metals, in which the resonance is clearly observable, as a probe.

2. BOUNDARY CONDITIONS

We describe the bimetallic system by the model Hamiltonian

$$\mathscr{H}=\mathscr{H}_1+\mathscr{H}_2+\mathscr{H}_T; \qquad (2)$$

Here $\mathscr{H}_1$ and $\mathscr{H}_2$ are the Hamiltonians of the individual metals, and

$$\mathscr{H}_T=\sum_{p,q,\sigma} T_{pq}a_{p\sigma}^+b_{q\sigma}+\text{c.c.} \qquad (3)$$

is the tunnel Hamiltonian,[7] which describes transitions of electrons from one metal to the other through the barrier at the junction. The electron creation and annihilation operators $a_{p\sigma}^+$, $a_{p\sigma}$ and $b_{q\sigma}^+$, $b_{q\sigma}$ in (3) correspond to the first and second metals, respectively (σ is the spin index), and the matrix element T_{pq} is proportional to the overlap integral of the wave functions of the electrons from the two metals.

To calculate the magnetization flux across the junction we use the method of a nonequilibrium statistical operator.[8] We are interested in the behavior of the system over long times and over long distances: t, $(r/v_f)\gg\tau_{tr}$, where τ_{tr} is the transport relaxation time and v_f is the Fermi velocity. The magnetizations of the two metals can then be described by the Bloch hydrodynamic equations. Under these conditions the relationship between the quasi-integrals of motion $m_i^+(\mathbf{r},t)$ and the conjugate thermodynamic parameters $F_i^\pm(\mathbf{r},t)$ is a local relationship:

$$F_i^\pm(\mathbf{r},t)=\frac{\beta}{2\chi_i}\langle m_i^\pm(\mathbf{r},t)\rangle. \qquad (4)$$

Here β is the reciprocal temperature, $\chi_i\equiv\chi_i^{+-}(\omega=0)\approx\langle m_i^z\rangle_0/H_0$ are the transverse static susceptibilities of the conduction electrons, and $\langle m_i^z\rangle_0$ is the equilibrium expectation value of the longitudinal magnetization in the external magnetic field H_0. The magnetization flux density across the junction, $j_i^+(\mathbf{r})$, is determined by the time evolution of the magnetic moment of the metal due to electron tunneling:

 0038-5646/84/010106-03$04.00

$$j_i^{\pm}(\mathbf{r}=\mathbf{r}_s)=\frac{i}{S}[M_i^{\pm},\mathscr{H}_T],\quad M_i^{\pm}=\int d^3r m_i^{\pm}(\mathbf{r});\qquad(5)$$

where S is the surface area of the junction, and the junction is assumed to be homogeneous.

Using the nonequilibrium statistical operator to average Eq. (5) by the standard method,[8]

$$\rho(t)=\frac{1}{Q}\exp\Big\{-\beta\mathscr{H}-\sum_{i=1}^{2}\int d^3r[F_i^-(\mathbf{r},t)m_i^+(\mathbf{r})+F_i^+(\mathbf{r},t)m_i^-(\mathbf{r})$$
$$-\int_{-\infty}^{0}dt'e^{\varepsilon t'}\frac{\partial}{\partial t'}[F_i^-(\mathbf{r},t+t')m_i^+(\mathbf{r},t')+F_i^+(\mathbf{r},t+t')m_i^-(\mathbf{r},t')]]\Big\},\qquad(6)$$

$Q=\mathrm{Sp}\exp\{\ldots\};\quad m(t)=\exp(i\mathscr{H}t)m\exp(-i\mathscr{H}t);\quad \varepsilon\to+0$

we easily find the following expression for the magnetization flux across the junction:

$$j_i^+(\mathbf{r}_s,\omega)=\sum_{j=1}^{2}b_{ij}(\omega)m_j^+(\mathbf{r}_s,\omega),\qquad(7)$$

$$b_{ij}(\omega)=\frac{1}{2S\chi_j}\int_0^\beta d\tau\int_{-\infty}^0 dte^{-i(\omega+i\varepsilon)t}\langle\dot M_j^-(t-i\tau)\dot M_i^+\rangle,\qquad(8)$$

where

$$\dot M_j^{\pm}=[\mathscr{H}_T,M_j^{\pm}].$$

In (7), $j^+(\omega)$ and $m^+(\omega)$ are Fourier components of the expectation values $\langle j^+(t)\rangle$ and $\langle m^+(t)\rangle$, respectively. In deriving (7) we also assumed that the overlap region of the electron wave functions (the "thickness" of the junction) is no greater than $v_f\tau_{tr}$. For this reason, the magnetizations which appear on the right side of (7) are the magnetizations at the junction itself.

From Eq. (7) we can immediately find boundary conditions of the type in (1) for the magnetization, since we can use $j_i^+(\mathbf{r},\omega)=-D_i\cdot\mathbf{n}_i\nabla m_i^+(\mathbf{r},\omega)$ in a hydrodynamic description. The kinetic coefficients b_{ij} can be expressed in accordance with (8) in terms of the equilibrium correlation functions of the magnetic moment fluxes of the metals. Expression (8) for the b_{ij} is analogous to the corresponding expressions for the transport coefficients in the Kubo theory.[9]

If we restrict the discussion to second-order perturbation theory in $\mathscr{H}_T$, we can express the correlation function in (8) in terms of the products of the imaginary parts of one-particle retarded Green's functions $G_{i\sigma}^R(k,\omega)$ of the conduction electrons from the two metals.[10] Ignoring the dependence of the matrix elements T_{pq} on the absolute values of the momenta, and assuming $\omega\tau_{tr}\ll1$, we then find

$$\mathrm{Re}\,b_{ij}(\omega)=(2\delta_{ij}-1)\frac{\pi g_ig_j\mu_B^2}{4S\chi_j}\langle T_{pq}^2\rangle_\Omega\frac{\mathrm{sh}(\beta\omega/2)}{\omega}\times\int_{-\infty}^{\infty}\frac{d\omega'}{\mathrm{ch}(\beta\omega'/2)\,\mathrm{ch}[\beta(\omega'-\omega)/2]}[N_1^+(\omega'-\omega)N_2^-(\omega')+N_1^-(\omega')N_2^+(\omega'-\omega)],$$
$$\mathrm{Im}\,b_{ij}(\omega)=-\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{\mathrm{Re}\,b_{ij}(\omega')}{\omega-\omega'}d\omega'.\qquad(9)$$

Here g_i is the g-factor, μ_B is the Bohr magneton,

$$\langle T_{pq}^2\rangle_\Omega=V_1V_2\int\frac{d\Omega_p}{4\pi}\int\frac{d\Omega_q}{4\pi}|T_{pq}|^2$$

is the expectation value of the square of the tunnel matrix element over the Fermi sphere, and

$$N_i^\sigma(\omega)=-\frac{1}{\pi}\int\frac{d^3k}{(2\pi)^3}\mathrm{Im}\,G_{i\sigma}^R(k,\omega)$$

is the state density of the conduction electrons of metal i with spin $\sigma=\pm1/2$.

The kinetic coefficients $b_{ij}(\omega)$ are thus expressed in terms of the state densities of the metals. We can show that the Re b_{ij} determine the broadening of the conduction-electron spin-resonance line, while the Im b_{ij} determine its shift. The off-diagonal terms of the matrix of coefficients b_{ij}—which determine the mixing of the magnetizations of the two metals—are related by the Onsager relation[9]

$$b_{12}\chi_2=b_{21}\chi_1.\qquad(10)$$

Since the Hamiltonian (3) conserves the total spin of the system, we can also write

$$b_{11}b_{22}-b_{12}b_{21}=0.\qquad(11)$$

a) We assume that the bimetallic system consists of normal metals (an $N-N$ junction) with a state density $N_i(0)$ which is constant near the Fermi level on scales larger than $\beta^{-1}(=T)$ and ω. It then follows from (9) that

$$\mathrm{Re}\,b_{ij}^{N-N}=(2\delta_{ij}-1)\frac{g_ig_j\mu_B^2}{4S\chi_je^2}\frac{1}{R_N},\qquad(12)$$

where R_N is the junction resistance in the normal state, given by[7]

$$\frac{1}{R_N}=4\pi e^2N_1(0)N_2(0)\langle T_{pq}^2\rangle_\Omega.\qquad(13)$$

The imaginary part of b_{ij}^{N-N} is of order $(\omega/D)\mathrm{Re}\,b_{ij}^{N-N}$ (D is the width of the conduction band), and the shifts of the resonant frequencies are inconsequential.

b) Now assuming that one of the metals is a superconductor, we have a situation in which the gap Δ in the energy spectrum causes the electronic excitations of the normal metal with energies below Δ to undergo Andreev reflection from the junction, and mixing of the magnetizations at the $N-S$ junction will be "frozen" at low temperatures. In fact, substituting in (9) the BCS state density in a magnetic field H_0

$$N_2^\pm(\omega)=N_2(0)\frac{|\omega_\mp|}{(\omega_\mp^2-\Delta^2)^{1/2}},\quad|\omega_\mp|>\Delta,\quad\omega_\mp=\omega\mp\frac{\omega_2}{2},$$

we find

$$\mathrm{Re}\,b_{ij}^{N-S}=(2\delta_{ij}-1)\frac{g_ig_j\mu_B^2}{4S\chi_je^2}\frac{1}{R_N}\left(\frac{2\pi\Delta}{T}\right)^{1/2}e^{-\Delta/T}\times\frac{\mathrm{sh}(\omega/2T)\,\mathrm{ch}[(\omega+\omega_2)/2T]}{\omega/2T};\quad\omega,T\ll\Delta,\qquad(14)$$

where $\omega_i=q_i\mu_BH_0$ is the Zeeman frequency of the conduction electrons. The width of the conduction-electron spin-resonance line of the normal metal due to the outflow of magnetization into the superconductor at a rate $\mathrm{Re}\,b_{11}^{N-S}$

thus falls off exponentially with the temperature, as has been found in experiments on the resonant transparency in the Cu-Nb system.[5] Furthermore, the strong frequency dependence of the state density of the superconductor gives rise to a significant imaginary part:

$$\operatorname{Im} b_{ij}^{N-S}(\omega)=(1-2\delta_{ij})\frac{g_i g_j \mu_B^2}{4S\chi_j e^2}\frac{1}{R_N}\frac{2\omega}{\pi\Delta}\left[1-\left(\frac{2\pi\Delta}{T}\right)^{1/2}e^{-\Delta/T}\right]; \tag{15}$$

$$\omega\ll T\ll\Delta.$$

The corresponding negative shift of the g-factor of the conduction electrons of the normal metal reaches a maximum at low temperatures; its value is on the order of $\omega/\Delta\approx 0.1$ of the broadening of the resonant line in the normal metal due to tunneling.

It follows from (12) and (14) that the coefficients $b_{ij}(\omega)$ are related to the parameters of the electron spectrum of the metals and the properties of the junction in the same way as the tunnel current across the junction.

3. SPECTRUM OF CONDUCTION-ELECTRON SPIN RESONANCE IN A BIMETALLIC SYSTEM

Solving Maxwell's equations and the Bloch equations under boundary conditions (1), we find an expression for the shape of the conduction-electron spin-resonance signal in a bimetallic plate. The general result is too lengthy to reproduce here, so we will discuss only some particular cases.

a) We first assume that the thicknesses of the metals, d_i, are small in comparison with the skin thickness δ_i and in comparison with the spin-diffusion depth $\delta_{ie}=(2D_i\tau_i)^{1/2}$ (τ_i is the spin-relaxation time of the electrons in the volume of the i-th metal). If the magnetization-equalization rate in the volume of the metal is considerably higher than the rate at which the magnetization escapes across the junction, i.e., if

$$D_i/d_i^2\gg \operatorname{Re}(b_{ii})/d_i,$$

then the resonant part of the surface impedance is given by

$$\Delta Z_{res}=\left(\frac{4\pi}{c}\right)^2\omega\frac{d_1\chi_1\omega_1(\xi_2-\beta_1)+d_2\chi_2\omega_2(\xi_1-\beta_2)}{\xi_1\xi_2-\beta_1\beta_2},$$

$$\xi_j=i(\omega_j-\omega)+\tau_j^{-1}+\frac{b_{jj}}{d_j},\quad \beta_1=\frac{b_{21}\delta_1^2}{d_1\delta_2^2},\quad \beta_2=\frac{b_{12}\delta_2^2}{d_2\delta_1^2}. \tag{16}$$

If the rate at which the nonequilibrium magnetizations of the two metals mix is small in comparison with the spin-relaxation rates in the volume and also in comparison with the separation of the resonant frequencies, i.e., if

$$\operatorname{Re} b_{21}/d_1+\operatorname{Re} b_{12}/d_2\ll\tau_1^{-1}+\tau_2^{-1},\quad |\omega_1-\omega_2|,$$

then the poles of expression (16) are

$$\omega_1^*=\left(\omega_1+\frac{\operatorname{Im} b_{11}}{d_1}\right)-i\left(\frac{1}{\tau_1}+\frac{\operatorname{Re} b_{11}}{d_1}\right),$$

$$\omega_2^*=\left(\omega_2+\frac{\operatorname{Im} b_{22}}{d_2}\right)-i\left(\frac{1}{\tau_2}+\frac{\operatorname{Re} b_{22}}{d_2}\right). \tag{17}$$

Expressions (17) determine the renormalization of the widths and g-factors of the resonant lines of the two metals due to electron tunneling.

In the opposite limit,

$$\operatorname{Re} b_{21}/d_1+\operatorname{Re} b_{12}/d_2\gg\tau_1^{-1}+\tau_2^{-1},\quad |\omega_1-\omega_2|,$$

which corresponds to a pronounced cross relaxation at the junction, the observed resonance signal is of a collective nature; i.e., it characterizes the resultant response of the dynamically coupled subsystems 1 and 2. The pole with which most of the absorption is related is

$$\omega^*=\frac{\omega_1\alpha_{12}+\omega_2\alpha_{21}}{\alpha_{12}+\alpha_{21}}-i\frac{\tau_1^{-1}\alpha_{12}+\tau_2^{-1}\alpha_{21}}{\alpha_{12}+\alpha_{21}}, \tag{18}$$

where $\alpha_{12}=(g_2/g_1)d_1\chi_1$ and $\alpha_{21}=(g_1/g_2)d_2\chi_2$. According to (18), the resonant frequency and the line width are averages with appropriate weights. The situation is reminiscent of the electron bottleneck which is observed in alloys with paramagnetic impurities, where the strong exchange coupling of the localized spins of the impurities and conduction electrons couples the motion of their magnetizations.[11]

b) The situation apparently of most interest for applications is that in which the conduction-electron spin-resonance signal in the second metal cannot be observed directly because, for example, the relaxation times τ_2 are too short. If the thicknesses of the metals satisfy the conditions $\delta_1\ll d_1\ll\delta_{1e}$ and $d_2\gg\delta_2$, the shape of the resonant signal is then described by

$$\Delta Z_{res}\approx-\left(\frac{4\pi}{c}\right)^2\frac{\delta_1^2}{2d_1}\frac{\omega\chi_1\omega_1}{\omega-\omega_1^*},$$

$$\omega_1^*=\omega_1-\frac{i}{\tau_1}-ib_{11}\left\{d_1\left[1+b_{22}\left(\frac{\tau_2}{D_2}\right)^{1/2}\operatorname{cth}\frac{d_2}{(D_2\tau_2)^{1/2}}\right]\right\}^{-1}. \tag{19}$$

Expression (19) corresponds to the ordinary absorption signal of the first metal with the one distinction that the resonant frequency and line width are renormalized by transitions of electrons into the second metal, followed by spin relaxation with a characteristic time τ_2 (if the electron does not manage to return to the first metal over this time).

The results expressed in Eqs. (9), (12), (14), and (19) suggest that it may be possible to use the spin-resonance method to study the electronic structure and spin-scattering processes in a wide variety of conducting media by making use of a junction between the medium of interest and a metal with good resonance lines (lithium or sodium, for example).

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Translated by Dave Parsons