LBL-32483
UC-404

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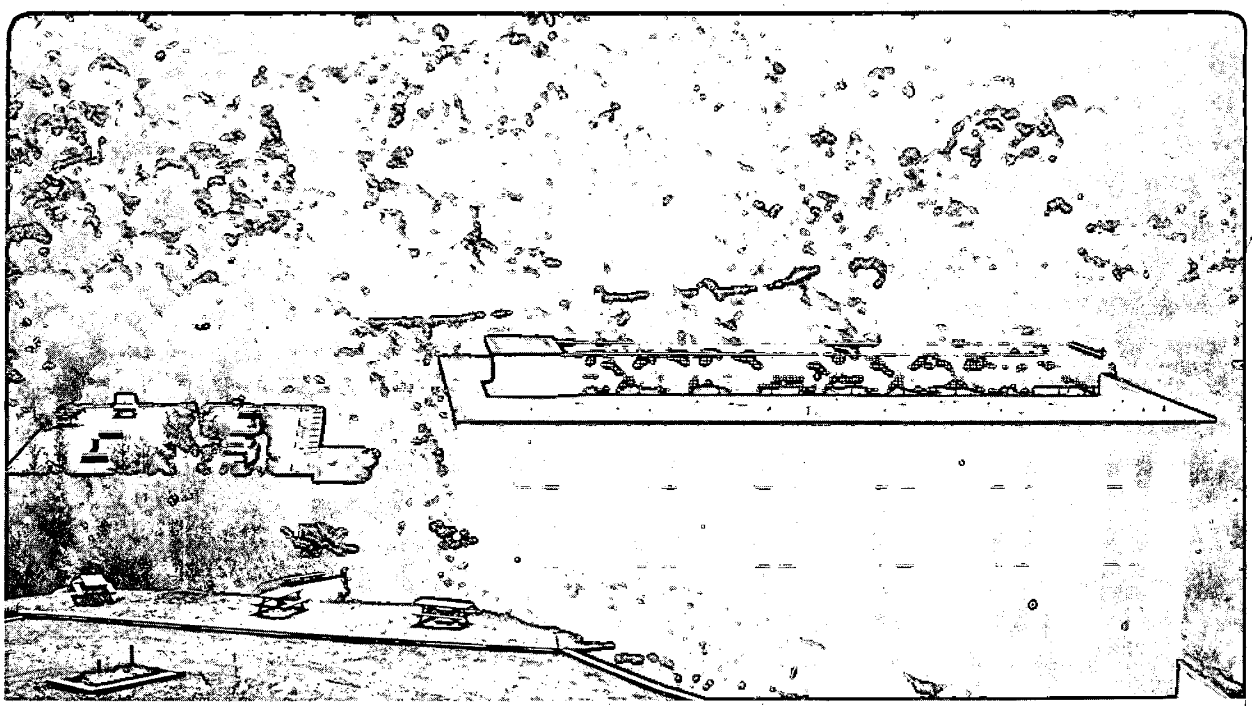
Materials Sciences Division

To be presented at the Symposium on Magnetic Ultrathin Films, Multilayers, and Surfaces, Lyon, France, September 7-10, 1992, and to be published in the Proceedings

A Boltzmann-Equation Approach to the Magnetoresistance of Ferromagnetic-Normal Metallic Multilayers

L.M. Falicov and R.Q. Hood

June 1992



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A BOLTZMANN-EQUATION APPROACH TO THE MAGNETORESISTANCE OF FERROMAGNETIC-NORMAL METALLIC MULTILAYERS*

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*This work was supported in part by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division of the U. S. Department of Energy under Contract No. DE-AC03-76SF00098.

Symposium on Magnetic Ultrathin Films,
Multilayers and surfaces. Lyon 1992

Contributed paper # 27

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ABSTRACT

The Boltzmann equation is solved for a system consisting of a ferromagnetic - normal - ferromagnetic metallic trilayer. The in-plane conductance of the film is calculated for two configurations: the ferromagnetic layers aligned (i) parallel and (ii) antiparallel to each other. The results explain the giant negative magnetoresistance encountered in these systems when an initial antiparallel arrangement is changed into a parallel configuration by application of an external magnetic field. A large negative magnetoresistance requires, in general, considerable asymmetry in the interface scattering for the two spin orientations.

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Ferromagnetic-normal metallic superlattices and sandwiches[1,2] display a number of interesting properties, such as a varying interlayer magnetic coupling[3] and a giant negative magnetoresistance (MR) effect.[4] In systems such as $(Fe/Cr)_n$, the magnetic moment of each Fe layer is arranged with respect to the neighboring layers either in a parallel fashion, or in an antiparallel one, depending on the thickness of the Cr spacers and on the quality of the Fe/Cr interfaces.[3,5,6]

When the conditions are such that the consecutive moments are arranged antiparallel to each other, the application of an external magnetic field to the sample results in two effects: (1) the moments rearrange themselves into a completely parallel arrangement in fields of the order of 1 T; and (2) the sample decreases its resistance -- negative MR -- in all directions (in-plane in particular) by varying amounts which can be as small as a few percent, and as large as 50% (for Fe/Cr at liquid Helium temperatures). This latter is known as the *giant magnetoresistance effect*[4,7–12] (GMR).

The present calculation,[13] an extension of the Fuchs-Sondheimer theory,[14,15] uses a Stoner description[16] of the itinerant ferromagnetic Fe layers: it introduces different potentials for majority and minority spins. Band-structure and electron-density effects are included only by means of a constant, metal- and spin-dependent potential, and an isotropic effective mass for each spin in each layer. The different potentials in neighboring layers results in coherent potential scattering (*i.e.,* refraction) of electrons as they traverse the interface. The angular-dependent effects are treated by a quantum-mechanical matching of the electron wave functions at the interfaces. Impurity scattering at the interface and interfacial roughness are also a source of spin-dependent scattering, and they contribute to the present model through a single spin-dependent parameter.

Figure 1 shows the system and defines the axes and geometric parameters. Both the current and the time-independent electric field are in the $\hat{x}$ direction. A sandwich consists of three flat layers (labeled 1, 2 and 3) of infinite extent in the $\hat{x}$ and the $\hat{y}$ directions of thicknesses d_1, d_2, and d_3. The structures investigated have identical ferromagnetic materials in layers 1 and 3 and a normal metal in layer 2. The symbols α and β are used to denote the surfaces of layers 1 and 3 with the vacuum, respectively, and A and B denote the 1-2 and 2-3 interfaces, respectively.

For a given sandwich the conductivity was calculated for both antiparallel alignment, denoted $\sigma_{\uparrow\downarrow}$, and for parallel alignment, denoted $\sigma_{\uparrow\uparrow}$, of the ferromagnetic moments of layers 1 and 3. The magnetoresistance ($\Delta\rho$ / ρ), is defined by

$$\frac{\Delta\rho}{\rho} \equiv \frac{\rho_{\uparrow\downarrow} - \rho_{\uparrow\uparrow}}{\rho_{\uparrow\downarrow}} = \frac{\sigma_{\uparrow\uparrow} - \sigma_{\uparrow\downarrow}}{\sigma_{\uparrow\uparrow}} \quad , \tag{1}$$

where $\rho_{\mu,\nu} = (\sigma_{\mu,\nu})^{-1}$. Note that this quantity varies between zero and one (or 0 and 100%) whenever the resistance decreases upon the application of an external magnetic field.

The conductivity for both alignments is obtained by adding the contributions of the spin-up and the spin-down electrons, calculated separately. This is the two-current model,[17] which provides a good description of electron transport in magnetic $3d$ metals. Spin-flip processes, which mix the two currents, are neglected. It is known that their effect is small at low temperatures.[17]

The theory includes 20 parameters:
three effective masses m_M, m_m, and m_s;
three constant potentials V_M, V_m, and V_s;
three relaxation times τ_M, τ_m, and τ_s;
three thicknesses d_1, d_2, and d_3;
four free-surface scattering parameters $P_{\alpha M}$, $P_{\alpha m}$, $P_{\beta M}$ and $P_{\beta m}$;
and four interface scattering parameters S_{AM}, S_{Am} S_{BM}, and S_{Bm}.

In all these symbols the subindex M (m) indicates majority-spin (minority-spin) electrons in the ferromagnetic layers; s indicates the electrons in the spacer layer.

At the two outer α and β surfaces [Figure 2 (a)], the specularity factors, $P_{\alpha\sigma}$ and $P_{\beta\sigma}$ -- for the respective surfaces and for electrons of spin σ -- take values between zero (completely diffusive scattering) and one (completely specular scattering) and provide a measure of the surface roughness. At the A and B interfaces [Figure 2 (b)], the parameters $S_{A\sigma}$ and $S_{B\sigma}$, which also vary between zero and one, are factors that indicate the degree of potential scattering for spin σ. The scattering follows the reflection-refraction laws when $S = 1$ and is completely diffusive when $S = 0$.

The results presented here include only the cases for which the *relaxation times* are identical $\tau \equiv \tau_m = \tau_M = \tau_s$. (The *mean free paths* of the minority and the majority spins within the ferromagnetic layers 1 and 3 and for the spacer metal are still different, however, since the Fermi velocities are different.) The discussion of the results is also confined to the situation $d_F \equiv d_1 = d_3$ and $d_s \equiv d_2$, since this is the most common case. At the outer surfaces all Ps are taken to be identical

$P \equiv P_{\alpha M} = P_{\alpha m} = P_{\beta M} = P_{\beta m}$. The spin dependence of these parameters is caused mostly by magnetic impurities, which are taken not to be present at the outer (identical) surfaces. The interfaces are also assumed to be identical $S_M \equiv S_{AM} = S_{BM}$; $S_m \equiv S_{Am} = S_{Bm}$.

Results are given for Fe-Cr. In these metals the isotropic effective mass is assumed to be independent of the material and spin orientation with a value $m_M = m_m = m_s = 4.0 \times$ free-electron mass. With this effective mass the potentials, with respect to the Fermi energy E_F chosen to be at $E_F = 0$, are $V_M = -8.23\ eV$, $V_m = -5.73\ eV$ for Fe; $V_s = -5.77\ eV$ for Cr.

Figure 3 shows the potential energies: V_M, V_m, and V_s for Fe-Cr for the spin-up and spin-down electrons for both the parallel and the antiparallel configurations.

The parameters that remain to be specified are altogether six: (A) two geometric parameters d_F and d_s; (B) one relaxation time τ, which depends on bulk sample properties; (C) one outer-surface scattering parameter P (the roughness of the outer surfaces); and (D) two interface scattering parameters S_M, S_m (diffuse scattering versus potential scattering at the interfaces for the majority and the minority spins respectively).

Even with these specifications, the phenomena under consideration are complicated functions of the 6 variables, and the task of describing these dependencies is not simple. In general terms, and with exceptions, it is found that $(\Delta\rho / \rho)$ is a strong function of the surface and interface parameters P, S_M, and S_m, and a relatively weak function of the thicknesses and the mean free path. For example, as P, S_M, and S_m vary between 0 and 1, the calculated $(\Delta\rho / \rho)$ varies between 0 and 92.7% for Fe-Cr trilayers when values of $d_F = d_s = 10.0$ Å and $\tau = 5.0 \times 10^{-13}\ s$ are chosen. Figure 4 shows the regions in this three-dimensional "surface and interfacial" parameter space where $(\Delta\rho / \rho)$ is greater than 20% for these values of d_F, d_s, and τ. With this choice of τ, the mean free paths are: (i) 4,250 Å for the majority-spin and 3,540 Å for the minority-spin electrons in Fe; and (ii) 3,560 Å for electrons in Cr. These values correspond to mean free paths which are orders of magnitude larger than the film thicknesses, *i.e.,* the clean-film limit, where surface and interface effects are supposed to be paramount.

It was found in general that:

(A) $(\Delta\rho\ /\ \rho)$ increases with increasing values of P, except in the region where $S_M \approx S_m \approx 1$.

(B) $(\Delta\rho\ /\ \rho)$ is in general small (only a few percent) when $S_M = S_m$, except when both parameters are very close to 1.

(C) $(\Delta\rho\ /\ \rho)$, as a function of d_F, exhibits a variety of behaviors which include (i) a monotonic decrease with increasing d_F; (ii) an initial increase followed by a decrease (a single maximum); (iii) a decrease, followed by an increase and a subsequent decrease (a minimum followed by a maximum); in all cases the asymptotic value as $d_F \to \infty$ is zero.

(D) $(\Delta\rho\ /\ \rho)$, as a function of increasing d_s, exhibits either (i) a continuous monotonic decrease, or, most commonly, (ii) a single maximum at a value of d_s of the order of d_F; the asymptotic value as $d_s \to \infty$ is also zero.

(E) $(\Delta\rho\ /\ \rho)$, as a function of the relaxation time τ, either (i) increases monotonically and saturates at a maximum value, or, most commonly, (ii) increases to a maximum, and then *very gradually* decreases.

Figure 4 shows how, for specific values of d_F, d_s, and τ, the quality of surfaces and interfaces influences the MR. As the surface scattering parameter P increases from 0 to 1, *i.e.,* as the scattering becomes less diffuse (or equivalently the surface roughness decreases) the MR in general increases. It is also evident from this figure that the region of large MR is close either to the plane $S_M = 1$, or to the plane $S_m = 1$, and away from the plane $S_M = S_m$. There is a very large asymmetry between S_M and S_m in Fe-Cr.

It is interesting to note that when $P = 1$, the MR of the trilayer becomes identical to that of an infinite multilayer or superlattice. A specular-scattering event makes the electron traverse the same ferromagnetic layer for a second time in the opposite direction or, equivalently, "continue" through a mirror-image of the film. Therefore, if for both surfaces $P = 1$, then as far as the MR is concerned, a trilayer

$$vacuum \mid d_F \mid d_s \mid d_F \mid vacuum$$

is exactly equivalent to an infinite, periodic superstructure

$$\cdots \mid 2d_F \mid d_s \mid 2d_F \mid d_s \mid 2d_F \mid d_s \mid 2d_F \mid \cdots$$

As seen above, the MR increases in general with P, because the number of interfaces where magnetic scattering can occur "increases" as P increases. When realistic values are chosen for the parameters, the MR is found to increase by as much as an order of magnitude when P increases from 0 to 1. This fact can be reinterpreted as an increase in the MR as the number of magnetic interfaces encountered by an electron within its bulk mean-free path increases.

Experimentally it is found that the more layers a sample has, the larger the MR. The (liquid He temperature) MR in Fe-Cr trilayers prepared by molecular-beam-epitaxy methods is found to be a few percent, while the MR is found to be nearly 50% Fe-Cr in *multilayers* prepared by the same method at the same temperature.[4]

As clearly seen in Figure 4, a large MR requires, in general, a large difference in interface scattering for the different spins. When $S_M = S_m$ (with some exceptions) the MR is found to be not more than a few percent. Therefore a large MR cannot be explained as being caused solely by different densities of electrons with different spins, which vary from layer to layer. What is required is a spin imbalance *and* a spin-dependent scattering mechanism at the interface, *i.e.*, $S_M \neq S_m$. When such a spin-dependent scattering mechanism exists, for example when magnetic impurities are present at the interfaces, the MR is profoundly influenced by spatial variations in the density of electron spins. This is the main cause of the GMR effect in ferromagnetic multilayers.

ACKNOWLEDGMENTS

We acknowledge helpful discussions with A. da Silva and P. Lévy. RQH acknowledges the support of the Department of Education. This research was supported, at the Lawrence Berkeley Laboratory, by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Science Division, U.S. Department of Energy, under contract No. DE-AC03-76SF00098.

REFERENCES

1 *Thin Film Growth Techniques for Low-Dimensional Structures,* edited by R. F. C. Farrow, S. S. P. Parkin, P. J. Dobson, J. H. Neave, and A. S. Arrott (Plenum, New York, 1987).

2 *Synthetic Modulated Structures,* edited by L. L. Chang and B. C. Giessen (Academic, New York, 1985); *Metallic Superlattices: Artificially Structured Materials,* edited by T. Shinjo and T. Takada, Studies in Physical and Theoretical Chemistry Vol. 49 (Elsevier, Amsterdam, 1987); B. Y. Jin and J. B. Ketterson, Adv. Phys. 38 (1989) 189.

3 S. S. P. Parkin, Phys. Rev. Lett. 67 (1991) 3598.

4 M. N. Baibich, J. M. Broto, A. Fert, F. Nguyen Van Dau, F. Pétroff, P. Etienne, G. Creuzet, A. Friederich, and J. Chazelas, Phys. Rev. Lett. 61 (1988) 2472.

5 S. S. P. Parkin, N. More, and K. P. Roche, Phys. Rev. Lett. 64 (1990) 2304.

6 J. Unguris, R. J. Celotta, and D. T. Pierce, Phys. Rev. Lett. 67 (1991) 140.

7 E. E. Fullerton, D. M. Kelly, J. Guimpel, I. K. Schuller, and Y. Bruynseraede, Phys. Rev. Lett. 68 (1992) 859.

8 P. Baumgart, B. A. Gurney, D. R. Wilhoit, T. Nguyen, B. Dieny, and V. Speriosu, J. Appl.. Phys. 69 (1991) 4792.

9 P. M. Levy, S. Zhang, A. Fert, Phys. Rev. Lett. 65 (1990) 1643.

10 A. Barthélémy, A. Fert, M. N. Baibich, S. Hadjoudj, F. Pétroff, P. Etienne, R. Cabanel, S. Lequien, F. Nguyen Van Dau, and G. Creuzet, J. Appl. Phys. 67 (1990) 5908.

11 J. Barnaś, A. Fuss, R. E. Camley, P. Grünberg, and W. Zinn, Phys. Rev. B 42 (1990) 8110.

12 R. E. Camley and J. Barnaś, Phys. Rev. Lett. 63 (1989) 664.

[13] R. Q. Hood and L. M. Falicov, Phys. Rev. B 46 (1992) [to appear].

[14] K. Fuchs, Proc. Cambridge Philos. Soc., 34 (1938) 100.

[15] E. H. Sondheimer, Adv. Phys., 1 (1952) 1.

[16] E. C. Stoner, Proc. R. Soc. London A, 165 (1938) 372.

[17] A. Fert and I. A. Campbell, J. Phys. F: Metal Phys. 6 (1976) 849; I. A. Campbell and A. Fert, in *Ferromagnetic Materials,* edited by E. P. Wohlfarth (North-Holland, Amsterdam, 1982), Vol. 3, p. 769.

FIGURE CAPTIONS

Figure 1

Schematic diagram of the ferromagnetic-normal-ferromagnetic metallic trilayer. Axes and thicknesses are defined.

Figure 2

Schematic diagrams of the scattering processes at (a) the vacuum-metal free surface and (b) the metal-metal interface. The parameters P and S_σ define the fractions controlled by the potentials. In (b) $S_\sigma R$ is the probability of specular scattering; $S_\sigma T$ is the probability of transmission (refraction) into the other metal. The isotropic, diffuse scattering parts are $(1-P)$ and $(1-S_\sigma)$, respectively.

Figure 3

Schematic diagrams of the potentials for the spin $\uparrow$ and spin $\downarrow$ electrons in the parallel ($\uparrow\uparrow$) and the antiparallel ($\uparrow\downarrow$) configurations of an Fe-Cr-Fe trilayer.

Figure 4

The region in three-dimensional parameter space (P, S_M, S_m) where $(\Delta\rho / \rho) > 0.2$ for the parameters corresponding to Fe-Cr and $d_F = d_s = 10\,\text{Å}$, and $\tau = 5.0 \times 10^{-13}\,s$. The three variables vary between 0 and 1.

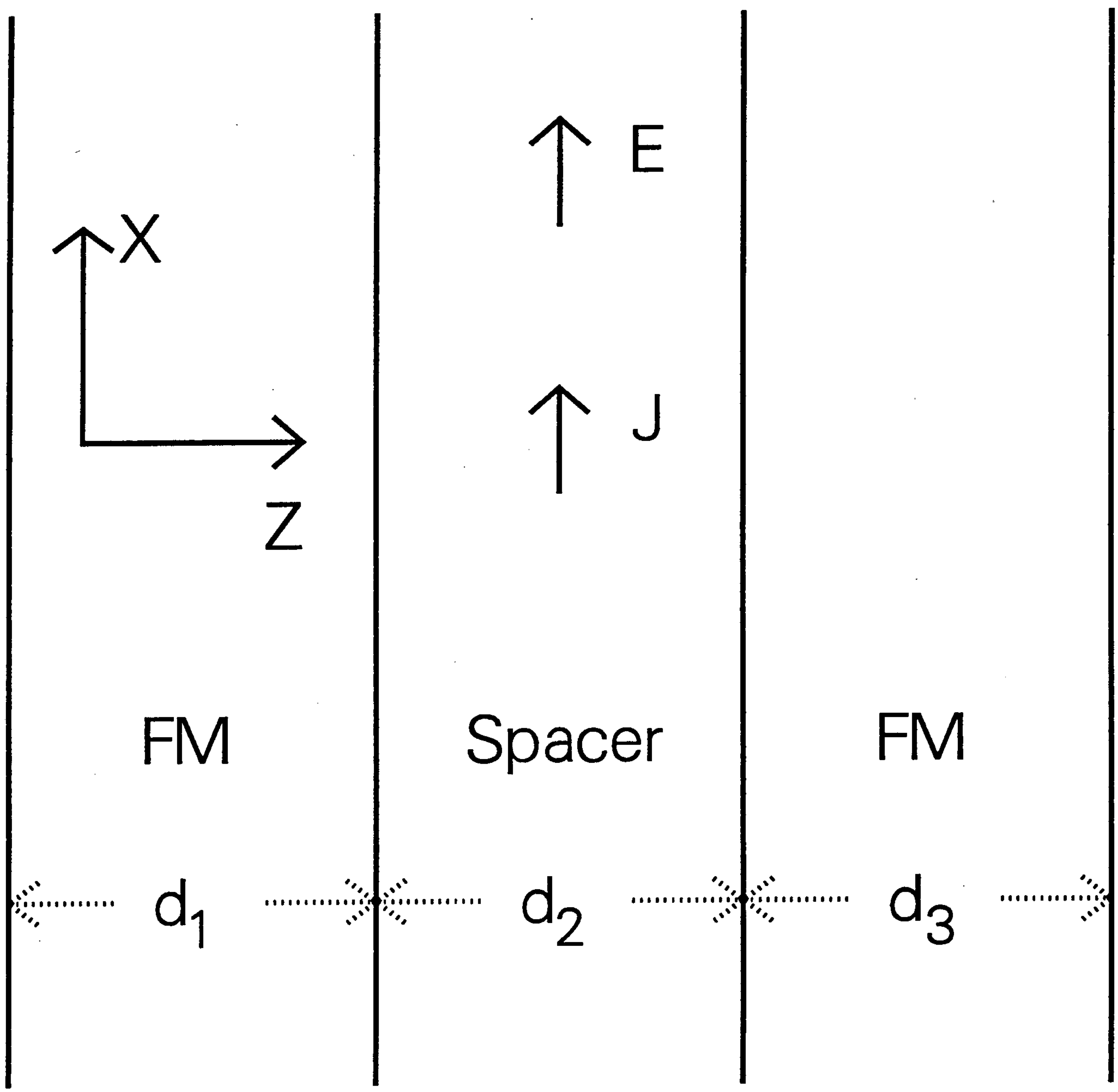

Figure 1

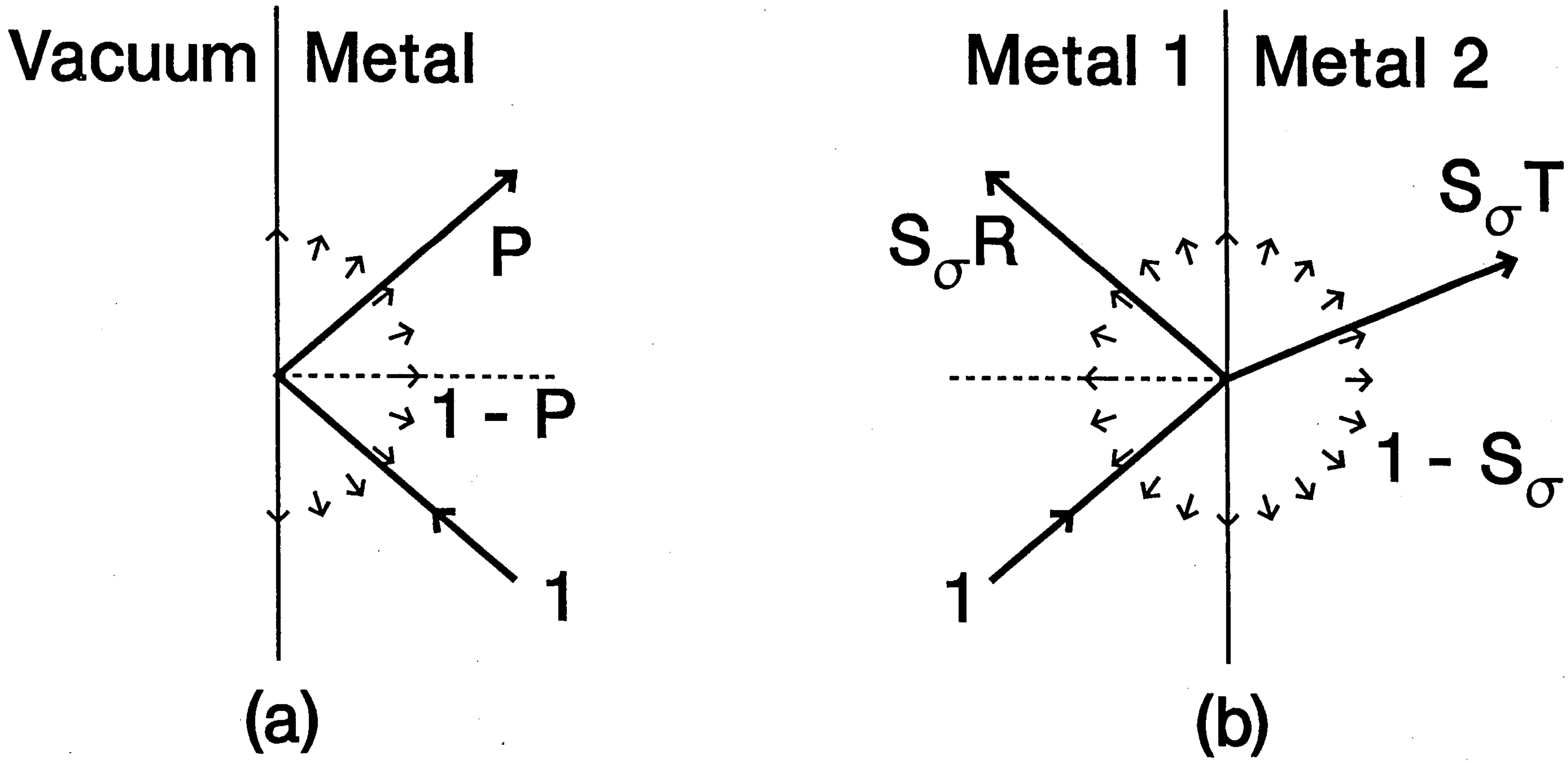
Vacuum
Metal
P
1 - P
1
(a)
Metal 1
Metal 2
$S_\sigma R$
$S_\sigma T$
$1 - S_\sigma$
1
(b)

Figure 2

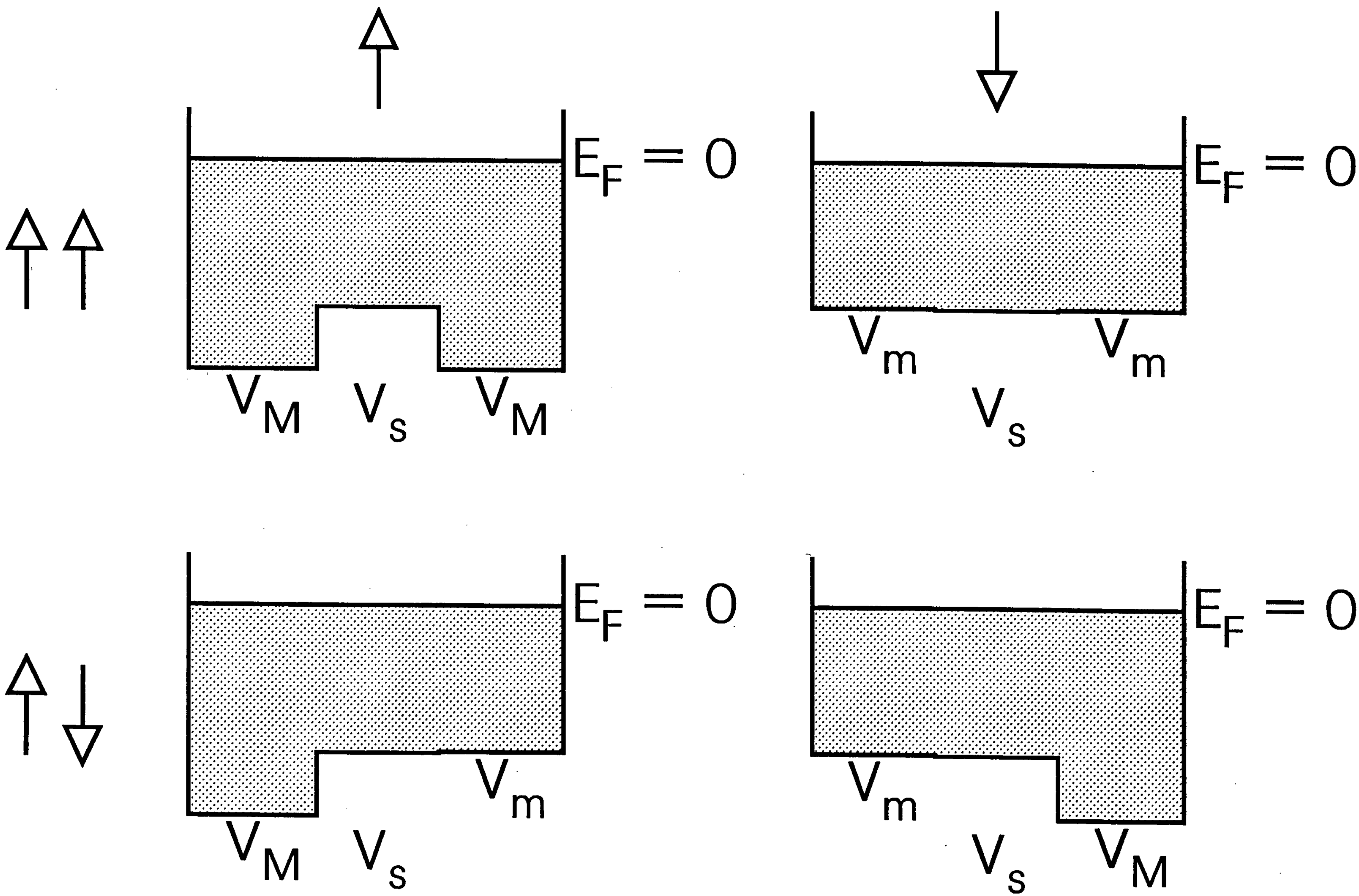

Figure 3

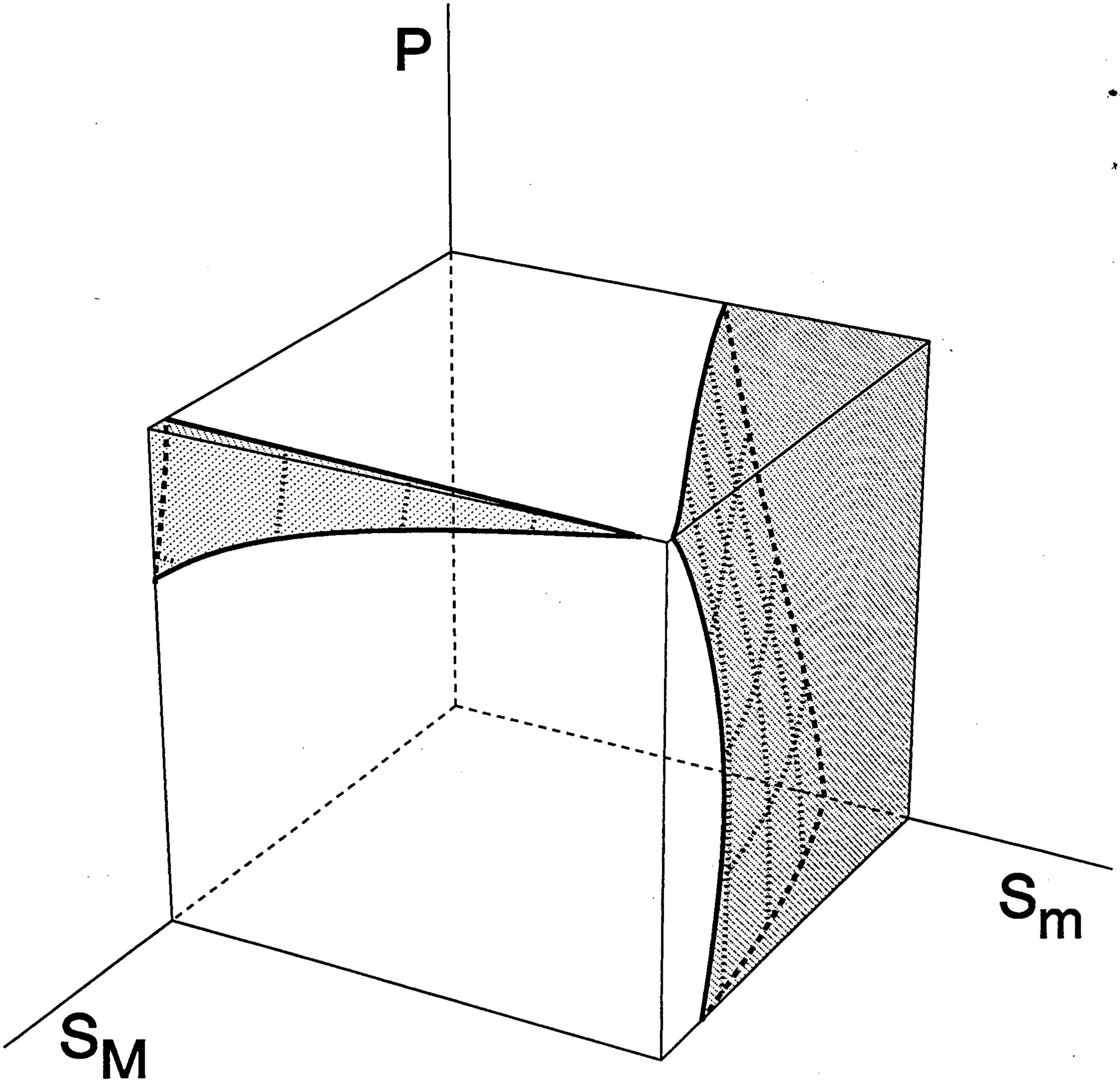

Figure 4

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