Developments of the theory of spin susceptibility in metals[*]

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ABSTRACT

The calculation of spin susceptibilities in metals, in particular the Ruderman-Kittel-Kasuya-Yosida (RKKY) polarization due to a point coupling, is reviewed. In the one-dimensional case, the low coupling limit of the non-linear theory clarifies traditional perturbation approaches. Multilayer structures require theories with bounded metals. An explicit formula is derived for the indirect interaction between two ferromagnetic plates in a half space. A case, where an oscillatory amplitude decays with the first power of the distance to the boundary is discussed.

Key-words: Spin susceptibility; Multilayers.

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[*]Dedicated to Roberto Luzzi

I. BASIC THEORY

The seemingly simple subject of the theory of the spin susceptibility in metals is still developing. This short review describes certain aspects of the subject. It may serve as an introduction into the field. However, in no way does it intend to cite all important contributions.

The subject was opened by Pauli [1] 70 years ago. He considered a homogeneous magnetic field acting on the spins of a degenerate electron gas and calculated the resulting magnetization. The electrons have a magnetic moment of one Bohr magneton μ_B, which leads to an energy in the field B of $\mp \mu_B B$, where the minus or plus sign applies when the moment is aligned or opposed to the field. This shifts the two bands which are filled to the same Fermi energy ϵ_F. The resulting change in occupation produces a net magnetisation $2\nu(\epsilon_F)\mu_B^2 B$, where $\nu(\epsilon_F)$ is the number of states per unit volume for one type of spin. Kittel raised the question whether a localized action on the spins in a metal would also turn around some spins, which would fill the whole volume with a magnetization. The answer was given in the famous paper by Ruderman and Kittel [2]. Plane waves are not diagonal in a inhomogeneous potential, which was assumed to have the form of a three-dimensional Dirac δ-function:

$$H' = -\gamma \delta^{(3)}(\mathbf{x})\frac{\sigma_z}{2},$$

where $\{\mathbf{x}, \sigma_z/2\}$ are the position and spin variables of a conduction electron. Perturbed states were formed using first order perturbation theory. With these states, and assuming no change in their occupation, the spin density as a function of the distance $r = |\mathbf{x}|$ becomes:

$$R(r) = \frac{\gamma m}{32\pi^3\hbar^2}\frac{\sin(2k_F r) - 2k_F r \cos(2k_F r)}{r^4} \tag{1}$$

where k_F is the radius of the Fermi sphere. $R(r)$ oscillates with increasing r with an amplitude which decays as r^{-3}.

The spacial integral over $R(r)$ gives $\gamma\nu(\epsilon_F)/2$. This corresponds to the Pauli result for the spin polarization due to a homogeneous coupling γ. For $r \to 0$, the function $R(r)$ diverges, indicating that a higher order approximation would not be meaningful. In fact, an operator as in Eq. (I) does not have an exact scattering theory [3]. It is obvious that it cannot be treated rigorously in a Schrödinger equation: let the negative three dimensional Dirac operator be represented by a cubic well of width a and debth a^{-3}. A Schrödinger wave squeezed into this well has kinetic energy $\sim a^{-2}$ and negative potential energy $\sim a^{-3}$, which dominates infinitely for $a \to +0$. Therefore, whenever such operators are used in solid state theory, it is understood that the treatment is limited to the lowest order Born approximation.

The method was also studied by Kasuya [4]. Yosida [5] was intrigued by the fact, that in the Ruderman-Kittel calculation no shift in the occupation of the states was applied, and yet, a net magnetization did result. He showed that by integrating over all virtual states and not excluding the same state, the normalization of the perturbed states was violated in such a way that this compensated the first mistake. In this way the method and result, often denoted by the initials RKKY, were consolidated.

It is straightforward to extend the theory to the Fermi-Dirac statistics at finite temperatures T [6]. At intermediate temperatures, when the width of the filled part of the band is comparable to $k_B T$, where k_B is the Boltzmann constant, the amplitudes of the oscillation is diminished. At hight temperature, for the Boltzmann gas, the magnetization has a Gaussian shape, where the reciprocal wavenumber of an electron with energy $k_B T$, i.e. $\hbar/\sqrt{2mk_B T}$, is the decay length.

II. SIMPLE APPLICATIONS

In the original paper [2], Eq. [1] represented the contact interaction between a nuclear spin and a conduction electron. The resulting indirect interaction between nuclear spins explained the linewidth of nuclear spin resonance in metals. The same form, Eq. [1], can

be used to describe the exchange coupling between the spin of a magnetic ion and the spins of conduction electrons. The δ-function approximates the situation where the diameter of the magnetic shell of the ion is small compared to the wavelength of an electron at the Fermi energy. The spin polarization due to one ion interacts with another ion and thereby produces an indirect coupling between the two ions. Considering the ions in the hexagonal lattice of the heavy rare earth metals, De Gennes [7] summed this coupling energy over all pairs of ions and found that he could explain the Curie temperatures of all the elements from Gd to Lu with just one coupling constant. This works also for the light rare earth metals [8]. In the liquid state of Gd [9], the radial distribution of the neighbours to an ion gives a theoretical Curie temperature which, while superior to that of the solid phase, is below the melting point. The experimental discovery [10], that thermally emitted electrons from ferromagnetic Fe have spin polarization zero, was explaned [11] by the decay of the electron spin polarization in the electron gas outside the metal.

III. EXTENSIONS OF THE THEORY

Kittel [12] applied the RKKY method to the one-dimensional metal and obtained a polarization with a constant term, i.e. an infinite range. Yafet [13] remarked that this unlikely result could be avoided by integrating first over the occupied states and then over the virtual states. The two-dimensional problem was only solved in 1986 by Béal-Monot [14]. The problem of the interaction per unit surface η of two parallel ferromagnetic plates embedded in a metal, also depends on just one variable, the distance a between the plates. η can be obtained by integrating over the couplings between a surface element of one plate with all surface elements of the other plate. The result is [13,15]:

$$\eta(v) = \Gamma\left[-\frac{\pi}{4} + \frac{Si(v)}{2} + \frac{\sin(v) - v\cos(v)}{2v^2}\right]. \tag{2}$$

with $v = 2k_F a$. Again, $\eta(v)$ oscillates, but for large v the amplitudes decay as $\sim v^{-2}$, and for $v \to 0$ the finite value $\eta(0) = -\Gamma\pi/4$ is obtained. Here $\Gamma = \beta\beta'\mathbf{I} \cdot \mathbf{I}'k_F^2 m/(8\pi^2\hbar^2)$, where $\mathbf{I}$ and $\mathbf{I}'$ are the directions of the magnetizations in the two plates. β is the interaction strength per unit surface of a plate at position a_x and a conduction electron with variables $\{\,\mathbf{r}, \sigma\}$:

$$H'_\beta = -\beta\delta(r_x - a_x)\,\mathbf{I} \cdot \frac{\sigma}{2}. \tag{3}$$

IV. NON-LINEAR THEORY

The Hamiltonian, Eq. (3), contains a one-dimensional Dirac δ-function. This produces a change in the slope of the wavefunctions. When the potential is attractive, it has one bound state or, rather, one bound band in view of the quantum numbers k_y and k_z. The exact treatment of one plate in the electron gas was given by Bardasis et al. [16] and Yosida et al. [17]. These papers received little attention, possibly since their explicit aim was to disprove a theory which ascribed the indirect coupling between ferromagnetic plates to quantum well states. An exact theory of the indirect interaction between two plates was developed by Bruno [18] using the framework of multiple scattering theory. Actually, the bound and propagating states in the presence of two plates with couplings as given by Eq. (3) are analytic expressions, so that the direct solution of the quantum mechanical problem for two plates is also possible [19]. The bound bands give an important contribution, which, however, is canceled by terms from the running waves. When the two plates have equal coupling and parallel magnetization, there is a symmetric bound state, and, for large enough distances between the plates, also an antisymmetric bound state. The distance where this state ceases to exist is not visible in the plot of the energy versus separation of the plates. The propagating states are orthogonal to the bound states. At the crucial distance the bound state appears with zero energy and infinite range. The part of the Hilbert space which belongs to the bound state, is taken over from the propagating states

in a smooth way which escapes detection.

The non-linear effects are surprisingly small: if the coupling strength is caracterized by the binding energy ϵ_0 of the bound state of a plate, then for $\epsilon_0 = \epsilon_F$ which is an intermediate coupling, the nodes of the function $\eta(v)$ are only slightly changed and the amplitudes are reduced to about half the value of the perturbation result, Eq. (2). For a strong coupling, say $\epsilon_0 = 8\epsilon_F$, which applies to the exchange coupling of an ion to the degenerate electron gas of a semiconductor, the interaction has much shorter range in the variable $2k_F a$. In the non-linear theory, a change in the density of a spin up state at a point is not exactly compensated by the change in the density of the spin down state. A net charge density appears, and the corresponding Coulomb energy becomes relevant in the strong coupling range [20].

Since with the planar interaction, Eq. (3), a bound state appears at arbitrarily small coupling β, a perturbation expansion around the point $\beta = 0$ is not valid. The puzzling one-dimensional RKKY results [12,13] are clarified by the low coupling limit of the non-linear theory [21].

V. INHOMOGENEOUS SYSTEMS

When two ferromagnetic metals are separated by a spacer metal of variing thickness a, the interaction between the magnetisations oscillates as a function of a. Often more than one period of oscillation can be extracted from the measurement. In real metals the interaction between the plates can be a superposition of functions η with several values of k_F, which correspond to extremal distances of the Fermi surface of the spacer metal, say the thickness of an 'arm' between 'Fermi sheres'. In the spacer metal the electrons are often in a lower potential, so that bound states appear periodically as a function of the width of the well. Repeatedly it has been suggested that this produces the oscillations of the interaction. However, it has been shown [16,17,22,23] that the formation of quantum well states does not lead to any periodicity in the coupling between the ferromagnets.

Experiments with multilayer structures call for a theory of the spin susceptibility in confined media. A simple model uses an electron gas in a semi-space [24]. The wavefunctions then must have a node at $x = 0$, where x is the coordinate perpendicular to the boundary. Thus the RKKY procedure is repeated using instead of plane waves the set

$$\sqrt{2}\,\sin(k_x x)\,e^{i\mathbf{k_y}\mathbf{y}+\mathbf{k_z}\mathbf{z}}, \qquad k_x > 0. \tag{4}$$

A spin-polarization P_h results which can be expressed in terms of the RKKY result R(r) in homogeneous space, Eq. (1), as follows:

$$P_h = R(2k_F\rho_-) + R(2k_F\rho_+) - \frac{(\rho_- + \rho_+)^2}{2\rho_-\rho_+}R[k_F(\rho_- + \rho_+)] \tag{5}$$

with $\rho_\mp = \sqrt{(x \mp a)^2 + y^2 + z^2}$ where $\{a, 0, 0\}$ is the position of the point coupling. Thus ρ_- is the distance to this point, and ρ_+ the distance to the mirror position of the point outside the boundary. The first two terms in Eq. (5) give equal contributions on the boundary where $\rho_- = \rho_+$. However, there the electron density vanishes, and indeed at the boundary the first two terms are cancelled by the third expression.

An interesting problem is the indirect interaction between two ferromagnetic plates at distances a_x and b_x from the boundary. This can be obtained by integrating the polarization due to a point copling at $\{a_x, 0, 0\}$ over the plane at b_x. For the first two terms in Eq. (5) this integration is identical to that which leads from Eq. (1) to Eq. (2). This is also the case for the third term, since the integration can be done with the variable $t = y^2 + z^2$, and $\partial(\rho_- + \rho_+)/\partial t = (\rho_- + \rho_+)/(2\rho_-\rho_+)$. Thus the two plates couple as

$$\eta_h = \eta(2k_F|a_x - b_x|) + \eta[2k_F(a_x + b_x)] - 2\eta[k_F(|a_x - b_x| + a_x + b_x)]. \tag{6}$$

The first term is the coupling of two plates in an infinite medium, Eq. (2). The second term is such a coupling, however, with the second plate in the mirror position. The third term subtracts two couplings to a plate at the boundary.

The confinement of a semi-space produces a remarkable effect with just one plate at a distance a_x from the boundary. The ferromagnetism in the plate can have a preferred

direction. The anisotropy energy is measured with a homogeneous outside field, which couples to the integrated magnetization. Part of this is the integrated magnetization $\mathcal{P}(a)$ of the conduction electrons. This is obtained by integrating the density of the states at the position a_x over the Fermi surface, Eq. (A8) of [24]. For the waves of Eq. (4) the spin polarization becomes (Eq. (18) of Ref. [24])

$$\mathcal{P}(a_x) = \frac{\gamma k_F}{8\pi^2} \frac{2m}{\hbar^2} \left[1 - \frac{\sin(2k_F a_x)}{2k_F a_x} \right] \tag{7}$$

The experiment has been performed [25] with an atomically thin Co film at a variable distance a_x from the surface of a Cu single crystal. The result shows oscillations which belong to two well known extremal vectors k_F of the Cu Fermi surface. The amplitudes decay slowly with the first reciprocal power of the distance a_x. This is the slowest decay of an oscillating polarization ever observed.

VI. DEDICATION TO ROBERTO LUZZI

Jorge Helman and I had planned to dedicate an original work to Roberto Luzzi. With the premature death of Jorge on January 7, 1997, at the age of 56 years, that project remains still in its initial steps. I therefore decided to dedicate this historic account of a subject, in which the activity of Jorge left its mark. In this way Jorge Helman is present in this volume. It was his wish to express his simpathy and admiration to his friend Roberto Luzzi.

REFERENCES

[1] W. Pauli,Z. Physik, **41**, 81 (1927)

[2] M.A. Ruderman and C. Kittel, Phys.Rev. **96**,99 (1954).

[3] José Giambiagi, private communication.

[4] T. Kasuya, Progr. Theoret. Phys. **16**, 45 (1956).

[5] Key Yosida, Phys.Rev. **106**, 893 (1957).

[6] W. Baltensperger and A. de Graaf, Helv.Phys.Acta **34**, 780 (1961). (?)

[7] P.G. de Gennes, Comptes Rend. **247**, 1836 (1958).

[8] J. Chevalier and W. Baltensperger, Helv.Phys.Acta **34**, 859 (1961).

[9] R.M.Chavier, X.A.da Silva and W. Baltensperger, Phys.Letters **15**, 126 (1965).

[10] A. Vaterlaus, F. Milani and F. Meier, Phys. Rev. Lett. **65**, 3041 (1990).

[11] J.S.Helman and W. Baltensperger, Modern Physics Letters, B **5**, 1769 (1991).

[12] C. Kittel, in *Solid State Physics*, edited by F.Seitz, D. Turnbull and H. Ehrenreich, (Academic, New York, 1968), Vol.22, p.11.

[13] Y. Yafet, Phys.Rev. **B36**, 8835 (1987).

[14] M.T. Béal-Monod, Phys.Rev. **B36**, 8835 (1987).

[15] W. Baltensperger and H.S. Helman, Appl.Phys.Lett. **71**, 2954 (1990).

[16] A. Bardasis, D.S. Falk, R.A. Ferrell, M.S. Fullenbaum, R.E. Prange, and D.L. Mills, Phys.Rev.Lett., **14**, 398 (1965).

[17] Key Yosida and A. Okiji, Phys.Rev.Lett., **14**, 301 (1965).

[18] P. Bruno, Europhys.Lett., **23**, 615 (1993), J. Magn. & Magn. Mater, **121**, 248 (1993)

and Phys.Rev. B **52**, 411 (1995).

[19] J.S. Helman and W. Baltensperger, Phys.Rev. B, **53**, 275 (1996).

[20] W. Baltensperger and H.S. Helman, unpublished.

[21] W. Baltensperger and H.S. Helman, Phys.Rev. B **54**,59 (1996).

[22] P. Bruno, Phys. Rev. Lett. **72**, 3627 (1994).

[23] P.Bruno, M.D.Stiles and Y. Yafet, Phys. Rev. Lett. **74**, 3087 (1995).

[24] J.S. Helman and W. Baltensperger, Phys.Rev. B**50**,12682-91 (1994).

[25] Ch. Würsch, C. Stamm, S. Egger, D. Pescia, W. Baltensperger and J.S. Helman, unpublished.