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Conduction Electron Spin Resonance* – I

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In this article, we give a brief description of magnetic resonance in metallic materials. In transmission conduction-electron spin-resonance experiments, the transmitted field is directly related to the magnetization. We derive this magnetization using an equation of motion which is an appropriate generalization of the equation of Codrington, Olds and Torrey. The proposed equation reproduces all the characteristic results obtained from quantum mechanical derivations.

Neste artigo, damos uma breve descrição de ressonância magnética em materiais metálicos. Nas experiências de ressonância de spin em elétrons de condução, com técnica de transmissão, o campo transmitido é diretamente relacionado com a magnetização. Deduzimos essa magnetização usando uma equação de movimento que é a generalização apropriada da equação de Codrington, Olds e Torrey. A equação proposta reproduz todos os resultados característicos das derivações quânticas.

1. Introduction

J. Winter [Comments in Solid State Physics, (1968)] remarked that the first experiments on spin resonance in paramagnetic metals were performed more than a decade ago by experimental groups at Berkeley[1] and a detailed theory was then developed by Dyson and later generalized by Azbel *et al.*[2]. However, only recently the interest in this field has been renewed as a result of the improvement and development of new techniques[3–11]. One is the transmission conduction electron spin resonance technique (TCESR), when a thin slab of the metallic sample separates two microwave cavities. The thickness L of the slab is typically much larger than the skin depth δ (6 of Cu, at 9.2 GHz, is of the order of 10^{-4} cm). An externa1 source of power is coupled to one of the cavities, the transmission cavity, and detection is made in the second cavity, the reception cavity. If properly

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insulated, power will be transmitted from one cavity to the other through the sample.

When the external field reaches the resonant condition, the spin system absorbs power on the side of the transmission, cavity. A transverse magnetization is generated in the sample and spreads out of the skin depth carried by the conduction electrons. This magnetization will eventually arrive at the other side of the sample and radiation will be generated in the reception cavity, where it is detected.

However, while this technique is not more sensitive than the standard one, it has several advantages. In the reflection type experimental setup, the sample is mounted in the microwave cavity. At resonance the sample produces an additional loss in the cítvity and a detectable modification in the Q-factor. But this technique is sensitive to every loss that can occur in the cavity, whether it results from the mechanism of interest as well as other ones. The signal could be obscured by spurious effects. This difficulty is eliminated in TCESR experiments, when no signal is transmitted unless under resonant conditions. Futhermore, the transmission technique is more stable than the reflection technique. In the latter case, the cavity forms a branch of a bridge circuit which, for sensitivity reasons works with a high degreí: of balance. Fluctuations and instabilities in the bridge circuit will produce noise superimposed on the signal unless one uses a low input power. This then limits the lowest detectable signal. In the transmission technique the balance of the circuit is represented by the insulation between cavities created by the sample. Furthermore, to obtain maximum sensitivity with both methods, one must modulate the signal in such a way 'that it could be amplified and detected with low noise level techniques. In reflection type experiments, one modulates the external magnetic field. In transmission type experiments, the transmitted field is modulated. This is much smaller than the external field, additional noise can be avoided and, therefore, sensitivity is increased.

2. Spin Resonance in Metals

Transmission experiments in Na and K allowed observation, ïor the first time, of spin-waves in these materials[12] which appear as side bands on the main spin resonance line. Platznian and Wolff[13] were able to describe these spin waves within the framework of the Fermi-liquid theory. A good amount of information on many-body effects,

i.e., on the effective interaction between electrons, can be derived from these experiments[14].

On the other hand, conduction electron spin-wave studies in ferromagnetic materials have a larger and richer history. However, the microscopic description of the effects of spatial non-uniformity of the magnetization, in ferromagnetic metals, lacks a completely general treatment. Contrary to the case of a non-magnetic metal, the calculation of the spin susceptibility and, therefore, of spin wave spectrum and spin diffusion, of a metallic ferromagnet in a Fermi-liquid formulation, involves quasi-particle states not too close to the Fermi surface. The lifetime of these states is finite and the results obtained within the framework of a Landau-quasi-particle picture is questionable[15]. However, it gives a first approximation and, at least, a qualitative description of the problem.

The discussion of the spin magnetization in a metallic sample can be accomplished in two different ways: from a microscopic point of view or through phenomenological arguments. In a previous paper[16], we have presented, using the time dependent Hartree-Fock approximation, a derivation of a unified theory for spin waves in an impure electron gas which is valid in both the paramagnetic and ferromagnetic state[17]. In this paper, we want to introduce a macroscopic description of magnetic resonance in a normal metal.

The study of magnetic resonance, spin waves and spin diffusion can be done by establishing the time and spatial dependence of the spin magnetization. The transmitted field, in TCESR experiments, is directly related to this magnetization as shown by Platzman and Lampe[18]. To obtain the transmitted field, it is necessary to solve Maxwell equations with the proper boundary conditions. Let us consider Fig. 1.

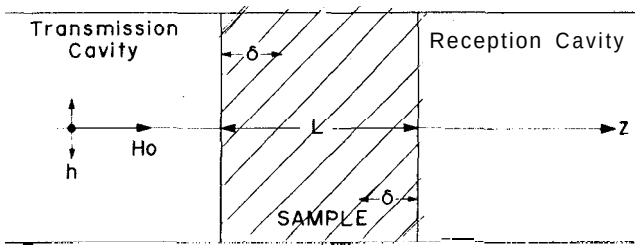

Fig. 1 - Description of the cavity in a TCESR experiment considered in the text.

The tangential components of E and H are continuous at $Z = \mathrm{L}$. Furthermore, the magnetization should satisfy

$$(\mathrm{n}\,.\,\mathrm{V})\,\mathbf{M}(\mathbf{r}, \mathrm{t}) = 0,$$

at $\mathrm{z} = 0$ and $z = \mathrm{L}$; n is a unit vector normal to the sample surface[19]. Let us approximate the r.f. field by a delta-function of amplitude h_0 (Ref. 18).

The magnetization propagating in z-direction is given by

$$\mathbf{M}(z, t) = \mathrm{h}_0 \int \frac{dq}{2\pi} \int \frac{d\omega}{2\pi}\, \chi(q, \omega) \exp\{i(\omega t - zq)\}, \tag{1}$$

where χ is the spin susceptibility. Since χ is an even function of q, only the real part, cos qz, contributes to Eq. (1). The boundary condition

$$\partial \mathbf{M}(z)/\partial z \,|_{z=0} = 0$$

then implies that $\mathrm{q} = n\pi/L$, with n an integer. Hence

$$\mathbf{M}(z, t) = (h_0/2\pi) \sum_{n=-\infty}^{\infty} \chi(n\pi/L, \omega) \cos(n\pi z/L). \tag{2}$$

On the other hand, at $z = \mathrm{L}$ one has the continuity of the tangential components of E and H: $E_t^{<} = E_t^{>}$ and $H_t^{<} = \mathrm{H}$ where $<$and$>$ indicate the values of the inner and outer fields at the sample-vacuum interphase. In vacuum (reception cavity), in cgs units, $\mathrm{E}' = \mathrm{H}'$.

The interna1 fields, E' and H', will be the superposition of the incident fields, $E_{\mathrm{in}}^{<}$ and $H_{\mathrm{in}}^{<}$, which carries the magnetization and of the refiected fields, $E_{\mathrm{re}}^{<}$ and $H_{\mathrm{re}}^{<}$.

From Maxwell equations for plane waves one, has

$$B_q = (c_q/\omega)\, E_q \tag{3}$$

and therefore

$$B_{q\,\mathrm{in}} = H_{q\,\mathrm{in}} + 4\pi M_q = (cq/\omega)\, E_{q\,\mathrm{in}}\,. \tag{4}$$

Using

$$B_{q\,\mathrm{in}} = \mu(q, \omega)\, H_{q\,\mathrm{in}}, \tag{5}$$

together with Eq. (4), one obtains

$$4\pi M_q = [\mu(q, \omega) - 1]\, H_{q\,\mathrm{in}} \tag{6a}$$

and

$$E_{q\,\mathrm{in}} = (\omega\mu/cq)\, H_{q\,\mathrm{in}} = Z(q, \omega)\, H_{q\,\mathrm{in}}\,. \tag{6b}$$

On the other hand, using Eq. (3) and the dispersion law,

$$c^2q^2 = \omega^2\varepsilon(q, o), \tag{7}$$

one finds

$$E_q = Z(q, \omega)\, H_q, \tag{8}$$

where the function

$$Z(q, \omega) = \mu(q, \omega)/\varepsilon^{1/2}(q, \omega) \tag{9}$$

is the same of Eq. (6b).

Since[20] $Z \sim q\delta$, this quantity grows with increasing wavenumber. The fields (E,, H_{in}) carried by the magnetization are important for small q, whereas the reflected fields (E,, , H,,) will contain components of small wavelength (they will be rapidly varying within the skin depth). With these considerations in mind, we can write

$$4\pi M \sim H_{in}^{<} \tag{10}$$

and

$$E_{,,} = \int dq\, Z(q, o) H_{,\,re}\, e^{iq_L}. \tag{11}$$

The surface impedance of the material, Q, is defined as[2']

$$Q = E^{<}/H^{<}$$

and then we will aproximately have

$$Q = E_{re}^{<}/H_{re}^{<} = (1/H_{re}^{<}) \int dq\, Z(q\omega)\, H_{q\,re}\, e^{iqL}, \tag{12}$$

since Z will be negligible for the small values of q characteristic of the incident fields.

Finally, using the boundary conditions at the interphase, we obtain

$$H_{in}^{<} + H; = E_{in}^{<} + E; \sim QH_{re}^{<} \tag{13}$$

and

$$H_{re}^{<} = H_{in}^{<}/(Q - 1). \tag{14}$$

Hence

$$H' = H_{in}^{<} + H_{re}^{<} = H_{in}^{<}[1 + (Q - 1)^{-1}] \tag{15}$$

or

$$H_{\text{transmitted}} \equiv H^{>} \simeq \frac{4\pi Q}{1-Q} M(L). \tag{16}$$

Eq. (16) tells us that the transmitted field is directly proportional to the magnetization tangential to the outer surface of the sample.

3. The Spin Magnetization

A most convenient way to discuss the time and spatial dependence of the spin magnetization is in terms of a Landau-Lifshitz-type equation[22]. The equation of motion for the magnetization is

$$\partial \mathbf{M}/\partial t = -\gamma(\mathbf{M} \times \mathrm{H}) + \mathrm{R}, \tag{17}$$

where

$$\mathrm{H} = \mathbf{H}_0 + \mathrm{h} + \lambda \mathbf{M} + (2A/M_0^2)\,\mathrm{V}^2\mathrm{M} \tag{18}$$

and $\gamma = |e|/mc$. The effective field H is a functional derivative of the energy with respect to the variation of the magnetization δM (Ref. 23), and contains, besides the constant applied field $\mathbf{H}_0$ and the r.f. field h, the term due to exchange interaction among electrons[24]. The constant A is usually referred to as the stiffness parameter[25], λ is the coupling constant between magnetic moments and M_0 is the static magnetization. The vector $\mathbf{R}(\mathbf{r}, t)$ represents a relaxation term due to energy dissipation.

A shortcoming of a phenomenological treatmerit resides in the question on the "destination" of the relaxing magnetizations, i.e., the proper choice of R. Usually, it has been considered either relaxation to thermal equilibrium, $\mathbf{M}_0 = \mathrm{X}\,\mathbf{H}_0$ (Ref. 26),

$$\mathbf{R}_{\mathrm{T}} = (M_z - M_0)\,\hat{z}/T_1 + \mathbf{M}_{\perp}/T_2$$

or to the instant local equilibrium value (M) (Ref. 27),

$$\mathbf{R}_{\mathrm{I}} = |M_z - \langle M_z \rangle|\,\hat{z}/T_1 + [\mathbf{M}_{\perp} - \langle \mathbf{M}_{\perp} \rangle]/T_2 .$$

Both choices do not reproduce the results obtained from the microscopic theory. Wangsness[28] has shown that in the steady state attained by a system of magnetic moments, in a resonance experiment, the rate of entropy production is a minimum, and that a macroscopic equation of motion for the magnetization which satisfíes such a requirement can be Eq. (17) with

$$\mathbf{R} = \mathbf{R}_{\mathrm{I}} + \mathbf{R}'$$

where

$$R' = (1/T_3H^2)\,\mathbf{H} \times (M \times H).$$

The relaxation term R' has been introduced by Codrington *et al.*[29].

To deal with Eq. (17), in the case of metals, we propose a generalization of the latter type of relaxation term as

$$\mathbf{R} = (1/T_L H_e^2)[\mathbf{H}_e \times (\mathbf{M} \times \mathbf{H}_e)] + (2A/T_e M_0^2)\,[\mathbf{M} \times (\mathbf{M} \times \nabla^2\mathbf{M})], \quad (19)$$

where

$$H_e = \mathbf{H}_0 + h.$$

The first term implies a transverse relaxation perpendicular to the instantaneous externa1 field with a relaxation time T_L, and will reproduce, for the transverse part of M, in which we are interested, a Bloch-Wangsness relaxation term R,. Hence, T_L can be interpreted as the usual transverse relaxation time. The second term implies a transverse relaxation perpendicular to the instantaneous exchange field, with a relaxation time T_e due to exchange interactions. Furthermore, let us observe that, as shown in the work of Ref. 16, the transverse relaxation time T_L should be replaced in the case of a metal with random distribution non-magnetic impurities, by $1/T_2 = 1/T_L + 1/\tau_2$, where τ_2 is an orbital relaxation time.

So far, we have neglected anisotropy. This effect can be accounted for by the introduction of an anisotropy field, $\mathbf{H}_A$, in the expression for the effective field H. For simplicity, we choose $\mathbf{H}_A = \beta(\mathbf{M} \cdot \mathbf{n})\mathbf{n}$, which is an appropriate expression for an uniaxial crystal and where $\mathbf{n}$ is an unit vector along the direction of easiest magnetization, and β a parameter. We will only consider here the case of parallel n, $\mathbf{H}_0$ and $\mathbf{M}_0$ along z-direction.

Furthermore, we will look at the case of spin waves propagation along the static field. Then one needs only to consider the transverse magnetization. Eq. (17), completed with Eqs. (18) and (19), is nonlinear. However, since only the case of small disturbances of the system from equilibrium are going to be discussed in this paper, it is a valid approximation to consider Eq. (17) up to first order terms in h and $m = M - \mathbf{M}_0$.

Introducing a circularly polarized r-f-field $h_0 \exp\{i\omega t - q \cdot r)$, normal to the constant magnetic field, we obtain, for the circularly polarized magnetization,

$$(\omega - \omega_0^* - D^* q^2)\, m_+(\mathbf{q}\omega) = -\gamma M_0 h_0^*, \quad (20)$$

where

$$\omega_0^* = \omega_0 + (i/T_2), \tag{20a}$$

$$\omega_0 = \gamma(H_0 + \beta M_0), \tag{20b}$$

$$D^* = c + iD = (2\gamma A/M_0)[1 - i(M_0/\gamma T_e)], \tag{20c}$$

$$h_0^* = h_0[1 + (i/\omega_0 T_2)]. \tag{20d}$$

Eq. (20) has the form of a diffusion equation with a complex diffusion constant D* defined in Eq. (20c). The real part of D* is directly related to the stiffness parameter and is the basic parameter of the long wavelength magnon dispersion relation. The imagiriary part, D, is the so called spin diffusion constant. We stress here that a diffusion term has naturally appeared in the present formalism and need not be introduced as an ad hoc hypothesis[30].

As is well known[31], the spin wave dispersion relation follows from the poles of the spin susceptibility $\chi_+(q,\omega)$. According to Eq. (20), it is given by

$$\omega(\mathbf{q}) = \omega_0 + Cq^2 - i\Gamma_\perp(\mathbf{q}), \tag{21}$$

with the spin wave damping constant being

$$\Gamma_\perp(\mathbf{q}) = Dq^2 + (1/T_2). \tag{22}$$

The real and imaginary parts of D* are not independent but satisfy the relation

$$D = -i(M_0/\gamma T_e)C = -2A/T_e. \tag{23}$$

We can observe that since T_e is positive, D and A have opposite sign. The stiffness parameter A is positive in the ferromagnetic state and negative in the paramagnetic state, which just reflects the relative stability of one state with respect to the other[32]. Consequently, the diffusion constant D changes sign, from positive to negative, going from the paramagnetic to the ferromagnetic state[16]. This diffusion term is a mixed effect of electron transport and exchange effects. The latter is a negative contribution negligible in the paramagnetic phase. In the ferromagnetic phase, on the contrary, exchange diffusion overcomes the kinetic effect, making D negative. A positive value for D_{ferro} has been derived by other methods[33] but those calculations only take account of the positive kinetic contribution.

If the r-f field is suddenly removed at time $t = t_0$, one gets for the transverse magnetization at $t > t_0$

$$M_+(\mathbf{r},t) = M_+(\mathbf{r},t_0)\exp\{itR_e\omega(\mathbf{q}) - t\Gamma_\perp(\mathbf{q})\}. \tag{24}$$

This means that the magnetization is relaxing to equilibrium with a relaxation time $T_R = \Gamma_\perp^{-1}(q)$. The width of the resonance line at half height is given by damping parameter $\Gamma_\perp$, which is composed of contributions derived from the relaxation terms towards the external and exchange fields. In the paramagnetic phase both effects are additive, while in the ferromagnetic phase the diffusion mechanism is opposing the other relaxation processes and would produce line width narrowing. However, spin diffusion effects are largely reduced in the ferromagnetic phase[16,33], and gives results severa1 orders of magnitude smaller than the usual relaxation times, giving no noticeable experimental effects.

Finally, replacing the result of Eq. (20) into Eq. (16), one finds for'the transmitted field[18]

$$H, \sim [\cot(W/2) - \cot W]/W, \tag{25}$$

where

$$W = (\omega - \omega_0)(L^2/D^*) - (iL^2/D^*T_2).$$

If $L >> 6$, we can write approximately

$$H_t \sim (K \sinh KL)^{-1}, \tag{26}$$

where

$$D^*K^2 = -i[(\omega - \omega_0) + (i/T_2)].$$

The proportionality factor is essentially the surface impedance of the metal and Eq. (26) is the standard Dyson formula[2].

4. Conclusions

In this article, we presented some comments concerning Conduction Electron Spin Resonance experiments. We reviewed, in Section 2, the connection between transmitted field and magnetization. A generalized macroscopic equation of motion was introduced in Section 3. The generalization of the equation of Codrington, Olds and Torrey[29], here proposed, reproduces all the results obtained from quantum mechanical derivations[13,16]. Our equation (19) reflects the existence of two relaxation mechanisms, characterized by the relaxation times T_2 and T_e. The first involves transverse relaxation normal to the instant applied fields, the second to the instantaneous Weiss molecular field.

A study of CESR in transition metals dilute alloys along a similar line as presented here, removing inconsistencies of previous approaches, will be presented elsewhere[34].

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