

Conduction Electron Spin Resonance II: Transition Metal Dilute Alloys*

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A study of EPR in transition metal dilute alloys is presented. Phenomenological equations to describe the dynamics of the magnetization are proposed. They reproduce the resonance condition that is obtained from quantum mechanical derivations and satisfy, without any *ad hoc* hypothesis, several physical properties not accounted for by previous models. A recalculation of the γ -shift in RPA is done, using a new approach. The cross-relaxation times are also obtained.

É apresentado um estudo de Ressonância Paramagnética Eletrônica em ligas metálicas diluídas. São propostas equações fenomenológicas para descrever a dinâmica das magnetizações. Elas reproduzem a condição de ressonância que é obtida por cálculos quânticos e verificam, sem nenhuma hipótese *ad hoc*, várias propriedades físicas não satisfeitas por modelos anteriores. Usando um novo tratamento, é recalculado o deslocamento do fator g . Os tempos de relaxação cruzados são também obtidos.

1. Introduction

In a previous paper¹, it was presented a brief description of magnetic resonance in metallic materials, with emphasis in non-magnetic dilute alloys. A phenomenological equation of motion for the magnetization was then proposed which reproduced all the characteristic results obtained from quantum mechanical derivations. Now, we address ourselves to a discussion of EPR in transition metals dilute alloys. The transmission technique in Ref. 1 seems to be very successful for this kind of experiments, which provide a satisfactory way to study localized moments^{2,3}. Within the general problem of magnetism in metals, that of localized moments in transition metal dilute alloys has recently been the object of particular interest and controversy^{4,5,6}. The most successful microscopic model has been that of Anderson⁵.

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For the sake of completeness, we present in the following section a brief review of Anderson's theory and the Schieffer-Wolff⁷ canonical transformation that shows the equivalence of Anderson hamiltonian with Kondo hamiltonian⁸, a form that will be used later on in Sections 4 and 5.

The experimental data of **EYR** of dilute magnetic alloys^{2,3} has been interpreted on the basis of Hasegawa's phenomenological equations⁹. These are Bloch-type equations for the magnetization, which involve relaxation of the magnetization towards thermal equilibrium. Sasada and Hasegawa¹⁰ performed a detailed quantum mechanical (RPA) calculation for the transverse magnetization using Anderson's hamiltonian. They obtain a resonance condition which agrees with the one obtained in the macroscopic approach.

However, Hasegawa^{9,10} makes quite a point of the fact that a severe shortcoming of the phenomenological treatment resides into the question about the destination of the relaxing magnetization. The equations involving thermal equilibrium destination agree with the results of the microscopic theory, but in order to satisfy the requirement of positive absorption, it is necessary to introduce an ad hoc hypothesis regarding the detailed balance condition¹⁰. Macroscopic equations involving instantaneous equilibrium destination^{10,11} correctly satisfy the requirement of positive energy absorption, for another particular detailed balance condition, but in that case it is not obtained the proper resonance condition¹⁰.

The present authors have proposed¹² an alternative phenomenological equation for the magnetization which involves transverse relaxations normal to the instantaneous external fields and to the instantaneous Weiss molecular fields. However, the final result is formally equivalent to that derived assuming relaxation to the thermal equilibrium values of the magnetization^{9,10}. The proposed model, without any additional hypothesis correctly produces a transverse spin-susceptibility that, for equal values of the gyromagnetic factors of both conduction and localized spins, (a) gives a resonance condition which can be identified term by term with the RPA result¹⁰, (b) satisfies the requirements of positive energy absorption, (c) satisfies the isotropy condition and (d) the exchange part of the relaxation terms has no effect on the resonance line shape. We present these results in detail in Section 3. In Section 4, we present a recalculation of the transverse spin susceptibility of the electron gas in dilute magnetic alloys, using a Green function technique

in conjunction with the use of Schrieffer-Wolff⁷ form of Anderson's hamiltonian⁵. Finally, in Section 5 we present the calculation of the transverse relaxation times that govern the cross-relaxation between the spin subsystems.

2. Localized Moments

In Anderson's own words, his microscopic model for the description of localized moments⁵, is a mathematical formulation and an extension of Friedel's ideas⁴. Essentially, he performs a self-consistent calculation to determine in which conditions localized moments appear.

Anderson's hamiltonian, in the second quantization formalism, reads

$$\begin{aligned}
 H = & \sum_{\mathbf{k}\sigma} \varepsilon_{\mathbf{k}\sigma} C_{\mathbf{k}\sigma}^{\dagger} C_{\mathbf{k}\sigma} + \sum_{j\sigma} E_{\sigma} d_{j\sigma}^{\dagger} d_{j\sigma} + \\
 & + U \sum_j d_{j\uparrow}^{\dagger} d_{j\uparrow} d_{j\downarrow}^{\dagger} d_{j\downarrow} + \sum_{\mathbf{k}aj} V_{\mathbf{k}j} (C_{\mathbf{k}\sigma}^{\dagger} d_{j\sigma} + d_{j\sigma}^{\dagger} C_{\mathbf{k}\sigma}).
 \end{aligned} \tag{2-1}$$

The first term is the unperturbed energy of the free electron system, and the second the unperturbed energy of electrons in d-states on impurity atoms. The third term is the Coulomb repulsive energy of two d-electrons on the same impurity, (the index j numerates the atomic impurities) and finally the last is the hybridization one-electron energy of s and d-electrons, being the fundamental term in Anderson's hamiltonian.

This hamiltonian is treated within the Hartree-Fock approximation and it is shown⁵ that localized moments will arise for small V_{kj}/U ratios. Schrieffer and Wolff⁷ analyze Anderson's model in the more favorable conditions for the existence of localized moments. Because of the inconvenience of dealing with the hybridization term in perturbation theory, [small denominators $(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}'}) \simeq 0$ appear in the terms in four order in V], they propose a canonical transformation that makes the hamiltonian (2-1) to take, in the new variables, a form similar to Kondo's hamiltonian⁸, containing a s-d-exchange-type energy. Since the Kondo effect^{8,13} seems to imply a condensation at very low temperature with the production of a localized conduction electron spin polarization that compensates that of the impurity, Schrieffer and Wolff conclude that there is no localized moments at zero temperature. However, there exists a critical temperature (generally of the order of 1⁰K or lower) above which such a localized conduction

electron polarization collapses and localized impurity moments can appear. We restrict ourselves to consider our system at temperatures above Kondo temperature.

Schrieffer-Wolff hamiltonian is obtained performing the transformation

$$H = e^S H_A e^{-S} \quad (2-2)$$

on hamiltonian (2-1), where

$$S = \sum_{kaj} \{B_\sigma(\mathbf{k}) + [A_\sigma(\mathbf{k}) - B_\sigma(\mathbf{k})] d_{j\sigma}^\dagger d_{j\sigma}\} (C_{k\sigma}^\dagger d_{j\sigma} - d_{j\sigma}^\dagger C_{k\sigma})$$

and

$$A_\sigma(\mathbf{k}) = V_k/(\varepsilon_{k\sigma} - E_\sigma - U), \quad B_\sigma(\mathbf{k}) = V_k/(\varepsilon_{k\sigma} - E_\sigma). \quad (2-3)$$

This transformation eliminates terms containing V in first order, to obtain

$$\begin{aligned} H = & \sum_{k\sigma} \varepsilon_{k\sigma} C_{k\sigma}^\dagger C_{k\sigma} + \sum_{j\sigma} E_\sigma d_{j\sigma}^\dagger d_{j\sigma} + \\ & + \sum_j U d_{j\uparrow}^\dagger d_{j\uparrow} d_{j\downarrow}^\dagger d_{j\downarrow} + \sum_{\mathbf{k}\mathbf{k}'} J_{\mathbf{k}\mathbf{k}'} (\psi_{\mathbf{k}}^\dagger \mathbf{S} \psi_{\mathbf{k}}) \cdot (\psi_{\mathbf{j}}^\dagger \mathbf{S} \psi_{\mathbf{j}}), \end{aligned} \quad (2-4)$$

where

$$\begin{aligned} J_{\mathbf{k}\mathbf{k}'} = & V_{\mathbf{k}'j} V_{j\mathbf{k}} [(\varepsilon_{\mathbf{k}} - E - U)^{-1} + (\varepsilon_{\mathbf{k}'} - E - U)^{-1} - \\ & - (\varepsilon_{\mathbf{k}} - E)^{-1} - (\varepsilon_{\mathbf{k}'} - E)^{-1}], \end{aligned} \quad (2-5)$$

$$\psi_{\mathbf{k}} = \begin{pmatrix} C_{k\uparrow} \\ C_{k\downarrow} \end{pmatrix} \quad \psi_{\mathbf{j}} = \begin{pmatrix} d_{j\uparrow} \\ d_{j\downarrow} \end{pmatrix}$$

and $2\mathbf{S} = \mathbf{a}$, with σ the Pauli matrices.

The Hamiltonian (2-4) clearly shows an exchange-type form with an effective exchange interaction $J_{\mathbf{k}\mathbf{k}'}$ that contains square powers of the hybridization potential V . Some other terms that are present in H after performing transformation (2-2) have been dropped⁷ because they do not give any substantial contribution in the calculation of Sections 4 and 5.

3. Phenomenological Equation of Motion

One of the existing phenomenological theories is due to Hasegawa⁹, who was the first to introduce the idea of considering the **conduction**

and the localized electrons as two different spin systems, each with its own resonant frequency and relaxation times but coupled through a spin-spin-type interaction.

According to Hasegawa, one can describe the dynamics of the spin system through a system of two equations of motion, one for each spin subsystem, coupled by means of Bloch-type relaxation terms. Figure 1 describes the relaxation scheme devised by Hasegawa. One serious difficulty involved in this treatment is that, for certain values of the parameters, Hasegawa's solution produces negative absorption. [cf. Hasegawa's Eq.(52) for $\lambda\chi_d < 1$]. This drawback can be overcome by redefining the "destination" values of the magnetization of the subsystems, namely, M_s and M_d , that appear in the detailed balance condition, $M_s/T_{sd} = M_d/T_{ds}$ as $M_s = \chi_s^0 H_{ext}$ and $M_d = \chi_d' H_{ext}$, where T_{sd} and T_{ds} are the cross relaxation times, χ_s^0 and χ_d' the static susceptibilities for the s-spins and for the "clothed" d-spins and H_{ext} the total external field¹⁰.

Another phenomenological model is that of Cottet *et al.*¹¹, which is based on instantaneous equilibrium destination, and the detailed balance condition $\chi_s^0/T_{sd} = \chi_d^0/T_{ds}$ needs to be introduced in order to satisfy the requirement of positive absorption. The resonance condition obtained from this model does not agree with that of Sasada and Hasegawa¹².

We propose a new system of phenomenological equations, a brief account of which has been given elsewhere¹², that involves particular mechanisms of relaxation associated with spinlattice and the cross spin-spin relaxations. Our results present a different phenomenological model that besides satisfying the physical requirements of positive absorption, isotropy condition and no effect of exchange contributions on the line shape, it correctly reproduces the expected resonance condition¹³ and is not burdened with the necessity of introducing any ad hoc condition of detailed balance.

We write for the equations of motion for the coupled s- and d-magnetizations

$$\dot{\mathbf{M}}_s = \gamma_s \mathbf{M}_s \times [\mathbf{H}_e + \lambda \mathbf{M}_d + (2A/M_{s0}^2) V^2 \mathbf{M}_s] + \mathbf{R}_s, \quad (3-1a)$$

$$\dot{\mathbf{M}}_d = \gamma_d \mathbf{M}_d \times (\mathbf{H}_e + \lambda \mathbf{M}_s) + \mathbf{R}_d. \quad (3-1b)$$

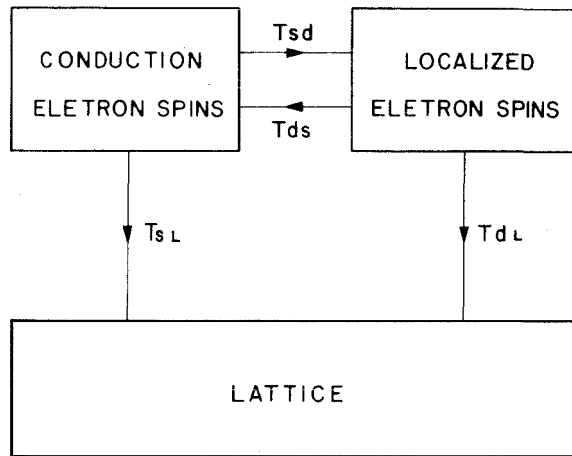


Fig. 1 – Hasegawa's relaxation scheme for coupled two spin systems.

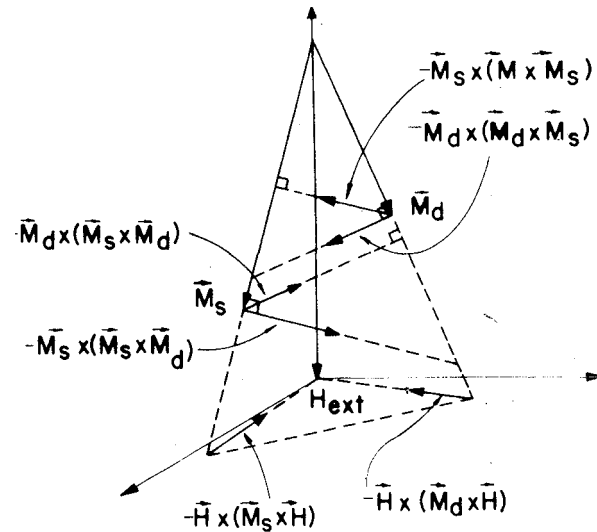


Fig. 2 – The relaxation vectors as proposed in the text

Here, γ_s and γ_d are the gyromagnetic factors of s- and d-spins. The field $\mathbf{H}_e$ includes the static and r.f. external fields. The internal fields are¹⁴ $\mathbf{H}_s = A_m \mathbf{M}_s + \lambda \mathbf{M}_d + (2A/M_{s0}^2) \nabla^2 \mathbf{M}_s$, and $\mathbf{H}_d = \lambda_{dd} \mathbf{M}_d + \lambda \mathbf{M}_s$. The coupling constants between magnetic moments are A_m , λ_{dd} and $\lambda_{sd} = \lambda_{ds} = \lambda$, M_{s0} , M_{d0} are the static magnetizations, and A is known as the stiffness parameter¹⁵. Finally, the vectors, $\mathbf{R}$, and $\mathbf{R}_d$ are the relaxation terms, whose choice decides the destination of the relaxing magnetizations. We propose for this relaxation terms

$$\mathbf{R}_s = \mathbf{R}_{sL} + \mathbf{R}_{ss} + \mathbf{R}_{sd}, \quad (3-2a)$$

$$\mathbf{R}_d = \mathbf{R}_{dL} + \mathbf{R}_{ds}, \quad (3-2b)$$

where the direct relaxation vectors are

$$\mathbf{R}_{sL} = (H_e^2 T_{sL})^{-1} \mathbf{H}_e \times (\mathbf{M}_s \times \mathbf{H}_e), \quad (3-3a)$$

$$\mathbf{R}_{dL} = (H_e^2 T_{dL})^{-1} \mathbf{H}_e \times (\mathbf{M}_d \times \mathbf{H}_e), \quad (3-3b)$$

$$\mathbf{R}_{ss} = (2A/M_{s0}^2 T_{ss}) \mathbf{M}_s \times (\mathbf{M}_s \times \nabla^2 \mathbf{M}_s), \quad (3-3c)$$

and the cross-relaxation vectors are

$$\begin{aligned} \mathbf{R}_{sd} = & (\Gamma_{sd}/M_{s0}^2) \mathbf{M}_s \times (\mathbf{M}_s \times \mathbf{M}_d) + \\ & + (\Gamma_{ds}/M_{d0}^2) \mathbf{M}_d \times (\mathbf{M}_s \times \mathbf{M}_d) \end{aligned} \quad (3-3e)$$

$$\begin{aligned} \mathbf{R}_{ds} = & (\Gamma_{ds}/M_{d0}^2) \mathbf{M}_d \times (\mathbf{M}_d \times \mathbf{M}_s) + \\ & + (\Gamma_{sd}/M_{s0}^2) \mathbf{M}_s \times (\mathbf{M}_d \times \mathbf{M}_s), \end{aligned} \quad (3-3f)$$

where Γ_{sd} and Γ_{ds} are phenomenological constants.

The relaxation terms (3-3a) and (3-3b) are of the type used (in the case of a single component system) by Codrington *et al.*¹⁶. These terms imply a transverse relaxation perpendicular to the instantaneous total external field, with relaxation time T_{sL} and T_{dL} , and they reproduce for the transverse part of $\mathbf{M}$, in which we are interested, a Bloch-Wangsness relaxation term¹⁷. The term $\mathbf{R}_{ss}$ of Eq. (3-3c) will result in a spin diffusion term, as discussed in Ref. 1. The cross terms $\mathbf{R}_{sd}$ and $\mathbf{R}_{ds}$ are a generalization of Landau-Lifshitz-type relaxation terms¹⁸, which are expected to be relevant in case of systems with strong exchange interactions, as is the present case. They account for transverse relaxations which have components perpendicular to the instantaneous exchange fields $\lambda \mathbf{M}_s$ and $\lambda \mathbf{M}_d$. Figure 2 describes the different relaxation contributions. It is worth mentioning that Wangsness¹⁸ has shown that in the steady state attained by a system of magnetic moments in a resonant experiment the rate of entropy production is a minimum.

Such a condition is satisfied by equations of motion containing either Codrington et al. or Wangsness-Bloch relaxation terms. Such a proof is presently not available for equations containing Landau-Lifshitz relaxation.

The two kinds of relaxation mechanisms so far introduced correctly satisfy

- i) for $\gamma_s = \gamma_d$ and when $T_{sL}^{-1} = T_{dL}^{-1} = 0$, one finds $R + \mathbf{R}_d = 0$, what means that exchange interactions do not modify the resonance line in the absence of other interactions;
- ii) the absorption coefficient is positive, as shown in Appendix A;
- iii) the isotropy condition is verified, as shown in Appendix B;
- iv) the resonance condition, to be obtained below, agrees with that of Sasada and Hasegawa, as will be shown with the help of Table 1.

The transverse susceptibility is derived in the linear approximation from Eqs.(3-1a) and (3-1b). Introducing $\mathbf{m} = \mathbf{M} - \mathbf{M}_0$, one finds

$$\dot{\mathbf{m}}_{s+} = i\Omega_{ss} \mathbf{m}_{s+} + i\Omega_{sd} \mathbf{m}_{d+} + i\beta_s h_+ + D^* \nabla^2 \mathbf{m}_{s+} \quad (3-4a)$$

$$\dot{\mathbf{m}}_{d+} = i\Omega_{dd} \mathbf{m}_{d+} + i\Omega_{ds} \mathbf{m}_{s+} + i\beta_d h_+, \quad (3-4b)$$

where

$$\Omega_{ss} = -\gamma_s (H_0 + \lambda M_{d0}) - i (T_2^{-1} + T_{sd}^{-1}), \quad (3-5a)$$

$$\Omega_{dd} = -\gamma_d (H_0 + \lambda M_{s0}) - i (T_{dL}^{-1} + T_{ds}^{-1}), \quad (3-5b)$$

$$\Omega_{sd} = \gamma_s \lambda M_{s0} + i T_{ds}^{-1}, \quad (3-5c)$$

$$\Omega_{ds} = \gamma_d \lambda M_{d0} + i T_{sd}^{-1}, \quad (3-5d)$$

$$D^* = 2A T_{ss}^{-1} - i \gamma_s A M_{s0}, \quad (3-5e)$$

$$T_{sd}^{-1} = \Gamma_{ds} + (M_{d0}/M_{s0}) \Gamma_{sd}, \quad (3-5f)$$

$$T_{ds}^{-1} = \Gamma_{sd} + (M_{s0}/M_{d0}) \Gamma_{ds}, \quad (3-5g)$$

$$\beta_s = \gamma_s M_{s0} + i(M_{s0}/H_0) T_{sL}^{-1}, \quad (3-5h)$$

$$\beta_d = \gamma_d M_{d0} + i(M_{d0}/H_0) T_{dL}^{-1}. \quad (3-5i)$$

Here, we have defined the transverse relaxation time $T_2^{-1} = T_s^{-1} + \tau_2^{-1}$, where τ_2 is an orbital relaxation time which appears due to the random distribution of magnetic and non-magnetic impurities^{9,18}.

MICROSCOPIC RPA Calculation (5)	PHENOMENOLOGICAL MODELS	
	Thermal Equilibrium Destination (4,5)	Present Model
$c\mu\Delta n$	$\overline{M}_d$	M_{do}
$2\mu^2 N(0)H_0$	$\overline{M}_s$	M_{so}
$P/2\mu^2$	λ	λ
$Qc\Delta n$	T_{2sd}^{-1}	$T_{sd}^{-1} = \Gamma_{ds} + \Gamma_{sd} \frac{M_{do}}{M_{so}}$
$QN(0)2\mu H_0$	T_{2ds}^{-1}	$T_{ds}^{-1} = \Gamma_{sd} + \Gamma_{ds} \frac{M_{so}}{M_{do}}$
0	T_{dL}^{-1}	T_{dL}^{-1}
τ_1^{-1}	T_{sL}^{-1}	$T_2^{-1} = T_{sL}^{-1} + \tau_2^{-1}$
0	0	$D^* = 2AT_{ss}^{-1} - i\gamma_s AM_{so}$
Detailed Balance Condition $\rightarrow$	$\overline{M}, T_{sd}^{-1} = \overline{M}_s T_{ds}^{-1}$	$M_{so} T_{sd}^{-1} = M_{do} T_{ds}^{-1}$

Table 1

Taking the time and Fourier transform of Eqs. (3-4a) and (3-4b), one finds a system of algebraic equations, linear in $m_{s+}(\mathbf{q}, \omega)$ and $m_{d+}(\mathbf{q}, \omega)$, which can be easily solved to obtain the transverse susceptibility:

$$\begin{aligned}\chi_+(\mathbf{q}, \omega) &= \frac{m_{s+}(\mathbf{q}, \omega) + m_{d+}(\mathbf{q}, \omega)}{h_+(\mathbf{q}, \omega)} = \\ &= \frac{\beta_s(\omega - \Omega_{dd}) + \beta_d(\omega - \Omega_{ss} - iq^2 D^*) + \beta_d \Omega_{sd} + \beta_s \Omega_{ds}}{(\omega - \Omega_{ss} - iq^2 D^*)(\omega - \Omega_{dd}) - \Omega_{sd} \Omega_{ds}}.\end{aligned}\quad (3-6)$$

In the absence of magnetic impurities, one recovers, for the transverse magnetization of conduction electrons, the results presented in Ref. 1, and the results of microscopic calculations¹⁸. The parameter D^* is the complex spin diffusion constant.

The resonance condition is given by the poles of the transverse susceptibility, *i.e.* by the solutions of

$$(o - R_s - iq^2 D^*)(o - \Omega_{dd}) - \Omega_{sd} \Omega_{ds} = 0. \quad (3-7)$$

Replacing the values of the parameters given in Eqs.(3-5), one finds that our result quite agrees with the results of the R.P.A. calculation of Ref. 10, and the model involving relaxation to thermal equilibrium, once the identification of coefficients, as shown in Table I, is made.

Furthermore, from Eqs. (3-5f) and (3-5g), one finds the detailed balance condition

$$M_{s0} T_{sd}^{-1} = M_{d0} T_{ds}^{-1}. \quad (3-8)$$

It should be observed that, for example for CuMn alloys, it is expected $1 > \lambda_{dd} \chi_d^0 \gg \lambda \chi_s^0 \gg \lambda_{ss} \chi_s^0$, and then Eq.(3-8) becomes approximately

$$\chi_s^0 T_{sd}^{-1} = \chi_d T_{ds}^{-1}, \quad (3-9)$$

where χ_s^0 is the bare or free electron static susceptibility and χ_d the dressed d-electron susceptibility $\chi_d = \chi_d^0 / [1 - \lambda_{dd} \chi_d^0]$ (see Appendix C).

Neglecting spacial dispersion in Eq.(3-7) (*i.e.*, putting $q = 0$), one can find the roots of Eq.(3-7). There are two solutions, $o+$ and $o-$, the latter corresponds to both spin magnetization precessing out of phase and being strongly damped and not observable. The other root, once it is taken into account that and since the exchange interaction is large and then

has the real part

$$\omega_{\text{res}} = \frac{\omega_s^0 + \chi_r \omega_d^0}{1 + \chi_r} \quad (3-10)$$

Here, $\omega_s^0 = \gamma_s H_0$ and $\omega_d^0 = \gamma_d H_0$ and $\chi_r = \chi_d^0 / \chi_s^0 = \text{Const.} / T$. On the other hand, the imaginary part of ω_+ is

$$T_{2\text{eff}}^{-1} = \frac{T_{sL}^{-1} + \chi_r T_{dL}^{-1}}{1 + \chi_r} \quad (3-11)$$

The temperature dependence of ω_{res} and $T_{2\text{eff}}$ occurs through χ_r . At low temperatures ω_{res} tends to ω_d^0 and $T_{2\text{eff}}$ tends to T_{dL} , and at high temperatures, *i.e.*, when $\chi_s^0 \gg \chi_d^0$, ω_{res} approximates ω_s^0 and $T_{2\text{eff}}$ tends to T_{sL} . These results are in accord with those originally obtained by Hasegawa⁹ and with the microscopic calculations^{10,19}.

4. Macroscopic Theory

The first quantum mechanical calculation of the spin susceptibility of the coupled s-d-spin system is due to Caroli *et al.*¹⁹. Using Anderson model⁵ and by means of an arduous calculation they obtain the y-shift in the R.P.A. Their result agrees with that of the phenomenological theory⁸. Sasada and Hasegawa¹⁰ extended the work of Caroli *et al.*, including spin-lattice relaxation of conduction electrons, cross relaxation and spin-orbit scattering of the conduction electrons. They performed a careful study of the bottleneck condition and investigated in detail the relation between microscopic and phenomenological equations.

In this section we reconsider the calculation of the transverse susceptibility using a Green function technique and starting from Schrieffer-Wolff version of Anderson hamiltonian, what we repute to be easier mathematical handling, and to provide a clearer connection with Hasegawa's description.

The transverse magnetic susceptibility is given by²⁰

$$\chi_+(\mathbf{q}, \omega) = -2\pi \langle\langle M_+(\mathbf{q}) | M_-(\mathbf{q}); \omega \rangle\rangle, \quad (4-1)$$

where $\langle\langle \dots | \dots; \omega \rangle\rangle$ is the Fourier transform of the retarded Green function.

The total magnetization of the two spin subsystems is

$$\mathbf{M}(\mathbf{r}, t) = \mu_s \psi_s^\dagger(\mathbf{r}, t) \boldsymbol{\sigma} \psi_s(\mathbf{r}, t) + \mu_d \psi_d^\dagger(\mathbf{r}, t) \boldsymbol{\sigma} \psi_d(\mathbf{r}, t), \quad (4-2)$$

where μ_s and μ_d are the magnetic moments of s- and d-electrons respectively and $\psi_{s,d}^\dagger$ are the second quantization operators

$$\psi_s(\mathbf{r}, t) = \sum_{\mathbf{k}\sigma} \psi_{\mathbf{k}\sigma}(\mathbf{r}) C_{\mathbf{k}\sigma}(t), \quad (4-3a)$$

and

$$\psi_d(\mathbf{r}, t) = \sum_{j\sigma} \phi_\sigma(\mathbf{r} - \mathbf{R}_j) d_{j\sigma}(t). \quad (4-3b)$$

Replacing (4-3a) and (4-3b) in Eq.(4-2), we obtain for the transverse part of the magnetization,

$$\begin{aligned} M_+(\mathbf{r}, t) = & 2\mu_s \sum_{\mathbf{k}\mathbf{k}'} \psi_{\mathbf{k}\uparrow}^*(\mathbf{r}) \psi_{\mathbf{k}'\downarrow}(\mathbf{r}) C_{\mathbf{k}\uparrow}^\dagger(t) C_{\mathbf{k}'\downarrow}(t) + \\ & + 2\mu_d \sum_{jj'} \phi_{j\uparrow}^*(\mathbf{r}) \phi_{j'\downarrow}(\mathbf{r}) d_{j\uparrow}^\dagger(t) d_{j'\downarrow}(t). \end{aligned} \quad (4-4)$$

Taking space-Fourier transform, it results for the $q=0$ component

$$M_+(0, t) = 2\mu_s \sum_{\mathbf{k}} C_{\mathbf{k}\uparrow}^\dagger(t) C_{\mathbf{k}\downarrow}(t) + 2\mu_d \sum_j d_{j\uparrow}^\dagger(t) d_{j\downarrow}(t). \quad (4-5)$$

In order to use Schrieffer-Wolff transformation of Anderson hamiltonian, it is necessary to express the magnetization in terms of the new variables. With S given in Eq.(2.3) one finds

$$\begin{aligned} \tilde{M}_+(0, t) = & e^S M_+ e^{-S} = 2\mu_s \sum_{\mathbf{k}} C_{\mathbf{k}\uparrow}^\dagger C_{\mathbf{k}\downarrow} + 2\mu_d \sum_j d_{j\uparrow}^\dagger d_{j\downarrow} + \\ & + 2(\mu_d - \mu_s) \sum_{\mathbf{k}} B(\mathbf{k})(C_{\mathbf{k}\uparrow}^\dagger d_{j\downarrow} + d_{j\uparrow}^\dagger C_{\mathbf{k}\downarrow}) + \\ & + 2(\mu_d - \mu_s) \sum_{\mathbf{k}j} D(\mathbf{k})(n_{j\uparrow} C_{\mathbf{k}\uparrow}^\dagger d_{j\downarrow} + n_{j\downarrow} d_{j\uparrow}^\dagger C_{\mathbf{k}\downarrow}), \end{aligned} \quad (4-6)$$

where $D(\mathbf{k}) = A(\mathbf{k}) - B(\mathbf{k})$. Let us observe that for $\mu_s = \mu_d$ the transformed magnetization of Eq.(4-6) has the same form than the magnetization given by Eq.(4-5). Since $|\mu_d - \mu_s|/\mu_s \approx 0.01$, we neglect the last two terms in Eq.(4-6). The transverse susceptibility is then given by

$$\begin{aligned} \chi_+(\omega) = & -4\pi\mu_s \sum_{\mathbf{k}\mathbf{k}'} \langle\langle C_{\mathbf{k}\uparrow}^\dagger C_{\mathbf{k}\downarrow} | C_{\mathbf{k}'\downarrow}^\dagger C_{\mathbf{k}'\uparrow}; \omega \rangle\rangle - \\ & -4\pi\mu_d \sum_{jj'} \langle\langle d_{j\uparrow}^\dagger d_{j\downarrow} | d_{j'\downarrow}^\dagger d_{j'\uparrow}; \omega \rangle\rangle. \end{aligned} \quad (4-7)$$

The Green functions that appear in this equation satisfy the coupled equations of motion²⁰

$$\omega \langle\langle C_{\mathbf{k}\uparrow}^\dagger C_{\mathbf{k}\downarrow} | C_{\mathbf{k}'\downarrow}^\dagger C_{\mathbf{k}'\uparrow}; \omega \rangle\rangle = \frac{1}{2\pi} \langle [C_{\mathbf{k}\uparrow}^\dagger C_{\mathbf{k}\downarrow}, C_{\mathbf{k}'\downarrow}^\dagger C_{\mathbf{k}'\uparrow}] \rangle +$$

$$+ \langle\langle [C_{k\uparrow}^\dagger C_{k\downarrow}, H] | C_{k'\downarrow}^\dagger C_{k'\uparrow}; \omega \rangle\rangle, \quad (4-8a)$$

$$\begin{aligned} \omega \langle\langle d_{j\uparrow}^\dagger d_{j\downarrow} | d_{j'\downarrow}^\dagger d_{j'\uparrow}; \omega \rangle\rangle &= \frac{1}{2\pi} \langle [d_{j\uparrow}^\dagger d_{j\downarrow}, d_{j'\downarrow}^\dagger d_{j'\uparrow}] \rangle + \\ &+ \langle\langle [d_{j\uparrow}^\dagger d_{j\downarrow}, H] | d_{j'\downarrow}^\dagger d_{j'\uparrow}; \omega \rangle\rangle, \end{aligned} \quad (4-8b)$$

where H is given in Eq.(2-4).

We treat the commutators in Eqs.(4-8a) and (4-8b) in the Random Phase Approximation^{20,21} to obtain

$$\begin{aligned} \omega \langle\langle C_{k\uparrow}^\dagger C_{k\downarrow} | C_{k'\downarrow}^\dagger C_{k'\uparrow}; \omega \rangle\rangle &= \frac{\delta_{kk'}}{2\pi} [\langle n_{k\uparrow} \rangle - \langle n_{k\downarrow} \rangle] + \\ &+ (\varepsilon_{k\downarrow} - \varepsilon_{k\uparrow}) \langle\langle C_{k\uparrow}^\dagger C_{k\downarrow} | C_{k'\downarrow}^\dagger C_{k'\uparrow}; \omega \rangle\rangle + \\ &+ \sum_j \frac{1}{2} J_{jk} [\langle n_{j\uparrow} \rangle - \langle n_{j\downarrow} \rangle] \langle\langle C_{k\uparrow}^\dagger C_{k\downarrow} | C_{k'\downarrow}^\dagger C_{k'\uparrow}; \omega \rangle\rangle + \\ &+ \sum_j \frac{1}{2} J_{jk} [\langle n_{k\downarrow} \rangle - \langle n_{k\uparrow} \rangle] \langle\langle d_{j\uparrow}^\dagger d_{j\downarrow} | d_{j'\downarrow}^\dagger d_{j'\uparrow}; \omega \rangle\rangle, \end{aligned} \quad (4-9a)$$

$$\begin{aligned} \omega \langle\langle d_{j\uparrow}^\dagger d_{j\downarrow} | d_{j'\downarrow}^\dagger d_{j'\uparrow}; \omega \rangle\rangle &= \frac{\delta_{jj'}}{2\pi} [\langle n_{j\uparrow} \rangle - \langle n_{j\downarrow} \rangle] + \\ &+ (\tilde{E}_\downarrow - \tilde{E}_\uparrow) \langle\langle d_{j\uparrow}^\dagger d_{j\downarrow} | d_{j'\downarrow}^\dagger d_{j'\uparrow}; \omega \rangle\rangle + \\ &+ \sum_k \frac{1}{2} J_{kk} [\langle n_{k\uparrow} \rangle - \langle n_{k\downarrow} \rangle] \langle\langle d_{j\uparrow}^\dagger d_{j\downarrow} | d_{j'\downarrow}^\dagger d_{j'\uparrow}; \omega \rangle\rangle \\ &+ \sum_k \frac{1}{2} J_{kk} [\langle n_{j\downarrow} \rangle - \langle n_{j\uparrow} \rangle] \langle\langle C_{k\uparrow}^\dagger C_{k\downarrow} | C_{k'\downarrow}^\dagger C_{k'\uparrow}; \omega \rangle\rangle, \end{aligned}$$

where $\tilde{E}_\sigma = E_\sigma - U_n$, and $(\dots)$ are statistical average values.

Following Anderson⁵, we replaced the products of two matrix elements V_{kj} by its average

$$V_{kj} V_{k'j} \simeq \langle V_{kj} V_{k'j} \rangle = V^2 \delta_{kk'} \quad (4-10a)$$

and therefore

$$J_{kk} = 2V^2 \cdot [(\varepsilon_k - E - U)^{-1} - (\varepsilon_k - E)^{-1}], \quad (4-10b)$$

where we have neglected the spin contribution.

Let us notice the *equivalence* between the terms in equations (4-9a) and (4-9b) with that of the phenomenological equations (3-4a) and (3-4b), with J_{kk} having the role of λ and omitting the relaxation terms.

Introducing the notation

$$\Omega_d(\omega) = \omega + \tilde{E}_\uparrow - \tilde{E}_\downarrow - \sum_k J_{kk} m_k/2, \quad (4-11a)$$

$$\Omega_k(\omega) = \omega + \varepsilon_{k\uparrow} - \varepsilon_{k\downarrow} - \sum_j J_{kk} m_j/2, \quad (4-11b)$$

$$m_j = [\langle n_{j\uparrow} \rangle - \langle n_{j\downarrow} \rangle], \quad (4-11c)$$

$$m_k = [\langle n_{k\uparrow} \rangle - \langle n_{k\downarrow} \rangle], \quad (4-11d)$$

system (4-9a) and (4-9b) becomes

$$\Omega_k(\omega) \chi_{kk'}(\omega) + \sum_{jj'} M_d \chi_{jj'}(\omega) = m_k, \quad (4-12a)$$

$$\Omega_d(\omega) \chi_{jj'}(\omega) + \sum_{kk'} M_k \chi_{kk'}(\omega) = m_j, \quad (4-12b)$$

where

$$\chi_{jj'}(\omega) = -271 \langle\langle d_{j\uparrow}^\dagger d_{j\downarrow} \mid d_{j'\downarrow}^\dagger d_{j'\uparrow}; \omega \rangle\rangle,$$

$$\chi_{kk'}(\omega) = -2\pi \langle\langle C_{k\uparrow}^\dagger C_{k\downarrow} \mid C_{k'\downarrow}^\dagger C_{k'\uparrow}; \omega \rangle\rangle.$$

Performing the summation $\sum_{kk'}$ on both sides of Eq.(4-12a) and $\sum_{jj'}$ on both sides of Eq. (4-12b), we are left with a system of two algebraic equations with two unknown variables. The roots of the secular equation

$$\Omega_d(\omega) - \sum_{kj} J_{kk}^2 m_k m_j \Omega_k^{-1}(\omega)/4 = 0 \quad (4-13)$$

produce the resonance frequencies. Using Anderson's notation

$$x = (\varepsilon_F - \tilde{E})/U, \quad y = I/\Delta, \quad A = \pi V^2 \rho_s,$$

where ρ_s is the density of s-electron states at the Fermi level, Eq. (4-13) becomes

$$\omega - \omega_d^0 + A(x, y) \frac{\omega_s^0(\omega - \omega_s^0)}{\omega - \omega_s^0 + \omega_s^0 A(x, y) \chi_r} = 0, \quad (4-14)$$

with $A(x, y) = \pi y x (1 - x)^{-1}$, $\omega_s^0 = \varepsilon_{k\downarrow} - \varepsilon_{k\uparrow}$, $\omega_d^0 = \tilde{E}_\downarrow - \tilde{E}_\uparrow$ and $\chi_r = \chi_d^0/\chi_s^0 = \chi_r(1^\circ K)/T$. For values appropriate to CuMn, i.e. $y \sim 10$, $x \sim 1/2$ and the impurity concentration ~ 100 ppm, produces $\chi_r(T=1^\circ K) \sim 3$ and then $\omega - \omega_s^0$ can be neglected in the denominator

in Eq. (4-14). The y-shift coincides with Eq. (3-10), becoming independent of the parameters of Anderson's hamiltonian. From these results we can identify λ as

$$\lambda = \frac{J_{\text{eff}}}{\mu^2} = [U/(\pi\mu^2xy)](1-x)^{-1}$$

Putting $U \sim 4\text{eV}$, one finds, for the x and y values given above, that $J_{\text{eff}} \sim 0.5 \text{ eV}$, which grossly coincides with the expected values for the exchange integral of Mn in noble metal hosts²².

In conclusion of this Section, we may say that Schrieffer-Wolff transformation of Anderson's hamiltonian makes quite clear how the covalent mixing of s - and d -electrons results in an effective exchange interaction.

The latter can be large for small values of x , *i.e.* when the renormalized d -level is near the Fermi level. Hasegawa's type of equations for the magnetization should appear, as in fact was shown here.

5. Cross Relaxation Times

We proceed in this section to an evaluation of the transverse relaxation times T_{sd} and T_{ds} . For that purpose, we use the formalism due to van Hove²³ in order to deal with the extension at finite temperatures of the "Golden Rule"²⁴. In the present case, we have

$$T_{ds}^{-1} = \int_{-\infty}^{\infty} dt \exp(-i\omega_d^0 t) \langle H_{j\downarrow j\uparrow}^{\text{exch.}}(0) H_{j\uparrow j\downarrow}^{\text{exch.}}(t) \rangle, \quad (5-1a)$$

$$T_{sd}^{-1}(\mathbf{k}) = \sum_{\mathbf{k}'} \int_{-\infty}^{\infty} dt \exp(-i\omega_{\mathbf{k}\mathbf{k}'} t) \langle H_{\mathbf{k}'\downarrow \mathbf{k}\uparrow}^{\text{exch.}}(0) H_{\mathbf{k}\uparrow \mathbf{k}'\downarrow}^{\text{exch.}}(t) \rangle, \quad (5-1b)$$

where $H_{nm'}^{\text{exch.}}$ is the exchange energy part of hamiltonian (2-4) when matrix element between states $|n\rangle$ and $|n'\rangle$ have been taken and $\omega_d^0 = E_{\downarrow} - E_{\uparrow}$ and $\omega_{\mathbf{k}\mathbf{k}'} = \varepsilon_{\mathbf{k}\downarrow} - \varepsilon_{\mathbf{k}'\uparrow}$. Furthermore, $\langle \dots \rangle$ stands for statistical average at temperature T .

According to Zubarev²⁰, Eqs. (5-1) can be rewritten as

$$\begin{aligned} T_{ds}^{-1} &= -4\pi [\exp(\beta\omega_d^0) - 1]^{-1} \text{Im} \langle\langle H_{j\downarrow j\uparrow}^{\text{exch.}} | H_{j\uparrow j\downarrow}^{\text{exch.}}; \omega_d^0 \rangle\rangle, \\ T_{sd}^{-1}(\mathbf{k}) &= -4\pi \sum_{\mathbf{k}'} [\exp(\beta\omega_{\mathbf{k}\mathbf{k}'}) - 1]^{-1} \text{Im} \langle\langle H_{\mathbf{k}'\downarrow \mathbf{k}\uparrow}^{\text{exch.}} | H_{\mathbf{k}\uparrow \mathbf{k}'\downarrow}^{\text{exch.}}; \omega_{\mathbf{k}\mathbf{k}'} \rangle\rangle. \end{aligned}$$

Replacing the interaction hamiltonians by their explicit form one finds

$$T_{ds}^{-1} = -\pi [\exp(\beta\omega_d^0) - 1]^{-1} \text{Im} \sum_{\mathbf{k}\mathbf{k}'\mathbf{p}\mathbf{p}'} J_{\mathbf{k}\mathbf{k}'} J_{\mathbf{p}\mathbf{p}'} \langle\langle C_{\mathbf{k}\downarrow}^\dagger C_{\mathbf{k}'\uparrow} | C_{\mathbf{p}\uparrow}^\dagger C_{\mathbf{p}'\downarrow}; \omega \rangle\rangle, \quad (5-2a)$$

$$T_{sd}^{-1}(\mathbf{k}) = -\pi \sum_{\mathbf{k}'} [\exp(\beta\omega_{\mathbf{k}\mathbf{k}'}^0) - 1]^{-1} \text{Im} \sum_{j,j'} J_{\mathbf{k}\mathbf{k}'}^2 \langle\langle d_{j\uparrow}^\dagger d_{j\downarrow} | d_{j'\downarrow}^\dagger d_{j'\uparrow}; \omega_{\mathbf{k}\mathbf{k}'} \rangle\rangle. \quad (5-2b)$$

One easily calculates the Green functions involved in Eqs. (5-2) obtaining

$$\langle\langle C_{\mathbf{k}\downarrow}^\dagger C_{\mathbf{k}'\uparrow} | C_{\mathbf{p}\uparrow}^\dagger C_{\mathbf{p}'\downarrow}; \omega \rangle\rangle = \frac{\delta_{\mathbf{k}\mathbf{p}'} \delta_{\mathbf{k}'\mathbf{p}}}{2\pi} \frac{\langle n_{\mathbf{k}\downarrow} \rangle - \langle n_{\mathbf{k}'\uparrow} \rangle}{\omega - \varepsilon_{\mathbf{k}'\uparrow} + \varepsilon_{\mathbf{k}\downarrow}} \quad (5-3a)$$

$$\langle\langle d_{j\uparrow}^\dagger d_{j\downarrow} | d_{j'\downarrow}^\dagger d_{j'\uparrow}; \omega \rangle\rangle = \frac{\delta_{jj'}}{2\pi} \frac{\langle n_{j\uparrow} \rangle - \langle n_{j\downarrow} \rangle}{\omega - \bar{\varepsilon}_\downarrow + \bar{\varepsilon}_\uparrow} \quad (5-3b)$$

Assuming $KT \gg \omega_d^0$, isotropic surfaces of constant free electron energy ε_k and $T \ll T_F$, $T_{sd}(k)$ has a constant value $T_{sd}(k_F)$ given by

$$T_{sd}^{-1} = \frac{\pi}{2} \frac{K_B T}{\omega_d^0} J^2 \omega_s^0 \sum_{\mathbf{k}} \frac{\partial n_{\mathbf{k}}}{\partial \varepsilon_{\mathbf{k}}} \delta(\omega_d^0 + \omega_{\mathbf{k}F\mathbf{k}F}) \quad (5-4a)$$

and

$$T_{ds}^{-1} = \frac{\pi}{2} \frac{K_B T}{\omega_s^0} J^2 \sum_j (n_{j\uparrow} - n_{j\downarrow}) \delta(\omega_d^0 + \omega_{\mathbf{k}F\mathbf{k}F}), \quad (5-4b)$$

where $J = J_{\mathbf{k}F\mathbf{k}F}$.

Using $\sum_{\mathbf{k}} \rightarrow \int d\varepsilon g(\varepsilon)$ and $\Sigma(n_{j\uparrow} - n_{j\downarrow}) = \chi_d^0 H_0 / \mu_d$ one finds

$$T_{sd}^{-1} = \frac{\pi}{6} c S(S+1) J^2 \rho_s \quad (5-5a)$$

and

$$T_{ds}^{-1} = \pi K_B T J^2 \rho_s^2, \quad (5-5b)$$

where ρ_s is the s-electron density of states at the Fermi level, and S the d-electron spin quantum number.

It is interesting to note that the results thus obtained formally agree with that of Overhauser²⁵ for relaxation of conduction electrons interacting with nuclear spins, and that of Heitler and Teller²⁶ for the relaxation of nuclear spins.

Let us observe that the expression for T_{sd} is valid only when the Zeemann splitting is smaller than the thermal energy KT . For typical values for CuMn with $c = 100$ ppm, one finds $T_{sd} \sim 10^{-11}$ sec. For comparison, typical relaxation times for spin lattice relaxation times are $T_{sL} \sim 10^{-9}$ sec and $T_{dL} \sim 10^{-8}$ sec. Therefore [cf. Eq. (3-11)], the resonance linewidth is determined by the spin-lattice relaxation times and then there is no experimental access to the cross relaxation times.

6. Conclusions

This work presents a study of EPR in transition metal dilute alloys. We present a phenomenological equation for the dynamics of the magnetization. This equation reproduces the quantum mechanical results and, without any additional ad *hoc* hypothesis, satisfies the main physical requirements of positive absorption, isotropy conditions and no exchange effects on the resonance line. Several relaxation mechanisms are introduced, involving transverse relaxation normal to the instantaneous external fields, and to the instantaneous Weiss molecular fields. The orbital relaxation time was introduced on a phenomenological basis, and inhomogeneity in the itinerant s-electron magnetization became responsible for the appearance of a spin diffusion term.

Finally, we consider some microscopic aspects of the problem. Calculations were performed utilizing Schrieffer-Wolff transformation of Anderson hamiltonian what makes clearer the connection with exchange-type interaction theories, and the phenomenological equations. Using the Bogoliubov-Tyablikov Green function technique, as described in the already classical paper by Zubarev, and the RPA, the equation of motion for the magnetization was obtained and from it, the resonance condition. Next we evaluated the cross relaxation times, which unfortunately cannot be determined from the EPR experimental data.

To conclude, it should be remarked that these microscopic calculations were within the Hartree-Fock approximation. A more precise treatment would be the utilization of the unrestricted Hartree-Fock approximation. Since this implies in leaving unspecified the spin orientation in d-orbitals, it should be a very convenient way to treat the particular problem in the theory of magnetism that we discussed here.

Appendix A: The Energy Absorption

The absorption coefficient is given by $-\omega \text{Im}\chi_+(\omega)$. Therefore, it is enough to verify that $\text{Im}\chi_+(\omega) < 0$ to show that absorption is definite positive. From Eq. (3-6), one finds

$$\chi_+(\omega) = N(\omega) \cdot D^*(\omega) / |D(\omega)|^2, \quad (\text{A-1})$$

where

$$N(\omega) = \beta_s(\omega - \Omega_{dd}) + \beta_d(\omega - \Omega_{ss}) + \beta_d\Omega_{sd} + \beta_s\Omega_{ds}, \quad (\text{A-2a})$$

$$D(\omega) = (\omega - \Omega_{dd})(\omega - \Omega_{ss}) - \Omega_{sd}\Omega_{ds}. \quad (\text{A-2b})$$

To find the sign of the imaginary part of the susceptibility, it suffices to study the sign of the numerator in (A-1). Replacing the values of the different parameters as given by Eq. (3-5), one finds for $\text{Im}ND^*$ a polynomial in T_{sd}^{-1} , T_{ds}^{-1} , T_{sL}^{-1} and T_{dL}^{-1} . The linear terms in T_{sL}^{-1} and T_{dL}^{-1} result

$$-\omega | \chi_s^0 T_{sL}^{-1} + \chi_d^0 T_{dL}^{-1} | | \omega + \gamma(H_0 + \lambda M_{s0} + \lambda M_{d0}) | < 0.$$

The linear terms in T_{sd}^{-1} and T_{ds}^{-1} vanish identically. Finally, the cubic terms in the inverse of the relaxation times result

$$\begin{aligned} & -\omega | \chi_s^0 T_{sL}^{-1} + \chi_d^0 T_{dL}^{-1} | (T_{sd}^{-1} + T_{ds}^{-1})^2 + T_{sd}^{-1} T_{dL}^{-1} + T_{ds}^{-1} T_{sL}^{-1} | + \\ & + (T_{sd}^{-1} + T_{ds}^{-1}) T_{sL}^{-1} T_{dL}^{-1} (\chi_s^0 + \chi_d^0) + (\chi_s^0 T_{dL}^{-1} + \chi_d^0 T_{sL}^{-1}) T_{sL}^{-1} T_{dL}^{-1} < 0. \end{aligned}$$

Others cubic terms in the inverse of the relaxation times, but not proportional to ω , vanish identically.

Therefore, $\text{Im}\chi_+(\omega)$ is definite negative and thus we have proved that power absorption, as derived from our phenomenological equations, is definite positive for all values of the parameters.

Appendix B: The Isotropy Condition

The isotropy condition means that in the static limit $\omega \rightarrow 0$, the susceptibility reduces to the longitudinal one, regardless the spin lattice relaxation. This fact implies the equality between the adiabatic and isothermal susceptibility¹⁰.

Taking $\omega \rightarrow 0$ in the Eq. (3-6) and neglecting the spin diffusion terms, we obtain

$$\chi_+(q, 0) = \frac{\beta'_s(\Omega_{ds} - \Omega'_{dd}) + \beta'_d(\Omega_{sd} - \Omega'_{ss})}{\Omega'_{dd}\Omega'_{ss} - \Omega_{sd}\Omega_{ds}} \quad (\text{B-1})$$

where the superscript (') means that we drop the spin lattice relaxation times in the Eqs. (3-5). Using these expressions in (B1) we have

$$\begin{aligned} \chi_+(q, 0) = & \\ = & \frac{\gamma_s \gamma_d (H_0 + \lambda M_{s0} + \lambda M_{d0}) (M_{s0} + M_{d0}) + i (T_{sd}^{-1} + T_{ds}^{-1}) (\gamma_s M_{s0} + \gamma_d M_{d0})}{\gamma_s \gamma_d H_0 (H_0 + \lambda M_{s0} + \lambda M_{d0}) + i H_0 (\gamma_d T_{sd}^{-1} + \gamma_s T_{ds}^{-1}) + i \lambda (\gamma_s - \gamma_d) (M_{d0} T_{ds}^{-1} - M_{s0} T_{sd}^{-1})}. \end{aligned} \quad (\text{B-2})$$

Using the detailed balance condition given by the Eq.(3-8), we finally obtain:

$$\chi_+(q, 0) = \frac{M_{s0} + M_{d0}}{H_0} = \chi_s^0 + \chi_d^0. \quad (\text{B-3})$$

Appendix C: The Static Susceptibilities

In the static limit, one finds

$$M_{s0} = \chi_s^0 (H_0 + \lambda_{ss} M_{s0} + \lambda M_{d0}), \quad (\text{C-1a})$$

$$M_{d0} = \chi_d^0 (H_0 + \lambda_{dd} M_{d0} + \lambda M_{s0}). \quad (\text{C-1b})$$

Solving this inhomogeneous system of linear equations, one finds

$$\chi_s = M_{s0}/H_0 = \chi_s^0 \left[1 - (\lambda_{dd} - \lambda) \chi_d^0 \right] / D \quad (\text{C-2a})$$

$$\chi_d = M_{d0}/H_0 = \chi_d^0 \left[1 - (\lambda_{ss} - \lambda) \chi_s^0 \right] / D, \quad (\text{C-2b})$$

where

$$D = (1 - \lambda_{ss} \chi_s^0) (1 - \lambda_{dd} \chi_d^0) - \lambda^2 \chi_s^0 \chi_d^0. \quad (\text{C-3})$$

Neglecting $\lambda_{ss} \chi_s^0$ (case of strong paramagnetism), and taking into account that (e.g. in CuMn) $\lambda \mu^2 \sim J_{\text{eff}} \sim 0.5$, $\lambda \chi_s^0 = \lambda \mu^2 \rho_s = J_{\text{eff}} \rho_s \sim 0.1$ ($\rho_s \sim 0.2 \text{ eV}^{-1}$), it results to a good approximation for χ_s and χ_d :

$$\chi_s \simeq \chi_s^0 \quad (\text{C-4a})$$

$$\chi_d \simeq \chi_d^0 / (1 - \lambda_{dd} \chi_d^0). \quad (\text{C-4b})$$

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