

Simple Theory of the 'Negative Residual Linewidth' in Reflection Spectroscopy of Magnetically Concentrated Local Moment Systems*

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Abstract

A mathematically improved analytical approximation is obtained to the solutions of the phenomenological equations of H. Hasegawa that describe the electron spin resonance of local moments in metals and the bottleneck effect that frequently occurs. The approximation shows that at all temperatures above the Curie point θ the linewidth is proportional to $T - \theta$ for any degree of bottlenecking, and not to temperature T . The solution also reveals that the g shift remains explicitly independent of temperature down to the Curie point. The expressions obtained take into account explicitly the direct local moment spin lattice relaxation rate and show that it is effective in breaking the bottleneck.

Introduction

The phenomenological theory of the bottleneck in the electron spin resonance (e.s.r.) of local moments in metals due to Hasegawa (1959) and its various improvements and extensions (Giovannini 1967; Cottet *et al.* 1968; Dupraz *et al.* 1970; Barnes *et al.* 1971) has proved to be of great importance in gaining an understanding of the spin dynamics of metals (Taylor 1975). However, available solutions of the equations of motion (see also Schultz *et al.* 1967; Monod and Schultz 1968; Pifer and Longo 1971, 1972) are unsuitable for providing a transparent resolution of one of the major problems, identified by Taylor (1975), in the reflection spectroscopy of magnetically concentrated materials: the 'negative' residual linewidth. In the present paper we show that a suitable approximation to the analytical solution of Hasegawa's equation is able to resolve this problem.

Equations of Motions

In this and the following section we will define the notation used, set the work in context, and correct an error in the literature. The physical system that we consider consists of a set of S state local moments denoted by d, a conduction electron system s and a lattice L. The d and s systems have similar gyromagnetic ratios γ and the various relaxation rates are given by the δ_{ij} . The equations of motion we need to solve are

$$\partial \mathbf{M}_d / \partial t = -\gamma \mathbf{M}_d \times (\mathbf{H} + \lambda \mathbf{M}_s) - (\delta_{ds} + \delta_{dL})(\mathbf{M}_d - \bar{\mathbf{M}}_d) + \delta_{sd}(\mathbf{M}_s - \bar{\mathbf{M}}_s), \quad (1)$$

$$\partial \mathbf{M}_s / \partial t = -\gamma \mathbf{M}_s \times (\mathbf{H} + \lambda \mathbf{M}_d) - (\delta_{sL} + \delta_{sd})(\mathbf{M}_s - \bar{\mathbf{M}}_s) + \delta_{ds}(\mathbf{M}_d - \bar{\mathbf{M}}_d). \quad (2)$$

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In these equations H is the total applied magnetic field and λ the molecular field coefficient that couples the s and d systems. The M are the instantaneous magnetizations and the \bar{M} are the values of the instantaneous *equilibrium* magnetizations to which the systems relax. These quantities are given by

$$\bar{M}_d = \chi_d^0(H + \lambda M_s), \quad \bar{M}_s = \chi_s^0(H + \lambda M_d), \quad (3a, b)$$

where χ_i^0 is the static susceptibility. There is also a detailed balance condition

$$\chi_s^0/\chi_d^0 = \delta_{ds}/\delta_{sd}. \quad (4)$$

These phenomenological equations have been justified from the s-d Hamiltonian at high enough temperatures with varying degrees of complexity and rigour by Barnes and Zitkova-Wilcox (1973), Zitkova-Wilcox (1973), Langreth and Wilkins (1972) and Smith (1973). The derivations are restricted to the case of low concentration and might not be fully applicable to the situation considered here.

Formal Solutions

Static Solution

The values of the magnetizations obtained under a static field, for example the steady field in an e.s.r. experiment, are derived by putting $M = \bar{M}$ in equations (3). Hence, we get

$$M_d/H = \chi_d = \chi_d^0(1 + \lambda\chi_s^0)(1 - \theta/T)^{-1}, \quad (5)$$

where

$$\theta/T = \lambda^2\chi_s^0\chi_d^0. \quad (6)$$

The parameter θ is the paramagnetic Curie temperature of the system if χ_d^0 obeys a Curie law and χ_s^0 is a Pauli susceptibility. We shall only consider the behaviour of the system in the paramagnetic phase.

An equation similar to (5) holds with the suffixes s and d interchanged, and lastly we obtain

$$\chi = (M_d + M_s)/H = \chi_d^0(1 + 2\lambda\chi_s^0 + \chi_s^0/\chi_d^0)(1 - \theta/T)^{-1}. \quad (7)$$

Dynamic Solution

We take the applied field to be

$$H = \hat{z}H + \hat{x}h \cos \omega t + \hat{y}h \sin \omega t, \quad (8)$$

and the magnetization of component i to be

$$M_i = \hat{z}M_i + \hat{x}m_i \cos \omega t + \hat{y}m_i \sin \omega t. \quad (9)$$

When these two expressions are substituted into the equations of motion (1) and (2), we obtain

$$\begin{pmatrix} \Delta\omega - \varepsilon_d & \zeta_d \\ \zeta_s & \Delta\omega - \varepsilon_s \end{pmatrix} \begin{pmatrix} m_d \\ m_s \end{pmatrix} = -h \begin{pmatrix} \eta_d \\ \eta_s \end{pmatrix}, \quad (10)$$

where

$$\Delta\omega = \omega - \gamma H, \quad (11)$$

$$\varepsilon_d = \gamma\lambda M_s + i\delta_{ds}(1 + L + \lambda\chi_d^0), \quad \varepsilon_s = \gamma\lambda M_d + i\delta_{sd}(1 + B + \lambda\chi_s^0), \quad (12a, b)$$

$$\zeta_d = \gamma\lambda M_d + i\delta_{sd}\{1 + \lambda\chi_s^0(1 + L)\}, \quad \zeta_s = \gamma\lambda M_s + i\delta_{ds}\{1 + \lambda\chi_d^0(1 + B)\}, \quad (13a, b)$$

$$\eta_d = \gamma M_d + iL\chi_d^0 \delta_{ds}, \quad \eta_s = \gamma M_s + iB\chi_s^0 \delta_{sd}. \quad (14a, b)$$

The bottleneck factor B and the lattice relaxation factor L are respectively

$$B = \delta_{sL}/\delta_{sd}, \quad L = \delta_{dL}/\delta_{ds}. \quad (15a, b)$$

The formal solution to equation (10) is the frequency and field dependent susceptibility

$$\chi(\omega, H) = (m_d + m_s)/h = N(\omega)/D(\omega), \quad (16)$$

where

$$N(\omega) = \eta_d(\varepsilon_s + \zeta_s - \Delta\omega) + \eta_s(\varepsilon_d + \zeta_d - \Delta\omega), \quad (17)$$

$$D(\omega) = (\Delta\omega - \varepsilon_d)(\Delta\omega - \varepsilon_s) - \zeta_d \zeta_s. \quad (18)$$

The resonant frequencies are given by

$$D(\omega) = 0. \quad (19)$$

Our quantity $\chi(\omega, H)$ is the same as the $[\chi^+(-\omega)]^*$ of Giovannini (1967) with ζ_1 and ζ_2 reversed. The expression (19) vanishes at the roots of the K_{20} of Pifer and Longo (1971, 1972).

Resonant Frequencies

In solving the quadratic equation

$$\Delta\omega^2 - \Delta\omega(\varepsilon_d + \varepsilon_s) + \varepsilon_d \varepsilon_s - \zeta_d \zeta_s = 0, \quad (20)$$

previous workers manipulated the coefficients into a form that enabled the roots to be expanded in powers of λ . We have found it preferable to evaluate the coefficients directly as they stand. Thus, we get

$$\varepsilon_d + \varepsilon_s = i\delta_{sd}\{1 + B + 2\lambda\chi_s^0 + (1 + L)\chi_s^0/\chi_d^0 - iD\}, \quad (21)$$

where the dynamical factor D is given by

$$D = \gamma\lambda(M_d + M_s)/\delta_{sd} \quad (22)$$

or

$$D = \gamma\lambda\chi_s^0 H(1 + 2\lambda\chi_s^0 + \chi_s^0/\chi_d^0)/\delta_{ds}(1 - \theta/T). \quad (23)$$

Since the term in the first set of parentheses in equation (23) will be close to unity for the usual experimental conditions of reflection spectroscopy, the dynamical factor is essentially equal to the ratio of the lineshift to the linewidth at any temperature in the unbottlenecked limit, as we shall confirm later. Further, after a certain amount of algebraic manipulation we obtain

$$\varepsilon_d \varepsilon_s - \zeta_d \zeta_s = -(1 - \theta/T)\delta_{sd}^2\{L + B(1 + L) - i(B + L)D/(1 + 2\lambda\chi_s^0 + \chi_s^0/\chi_d^0)\}\chi_s^0/\chi_d^0. \quad (24)$$

At this stage we can expand the square root in the solution of equation (21),

$$\Delta\omega_x = \frac{1}{2}(\varepsilon_d + \varepsilon_s)[1 \pm \{1 - 4(\varepsilon_d \varepsilon_s - \zeta_d \zeta_s)/(\varepsilon_d + \varepsilon_s)^2\}^{\frac{1}{2}}], \quad (25)$$

to get the frequency shift of the observable resonance

$$\Delta\omega_1 = (\varepsilon_d \varepsilon_s - \zeta_d \zeta_s)/(\varepsilon_d + \varepsilon_s). \quad (26)$$

Combining equations (21) and (24) gives the expansion parameter in equation (25),

$$-4(1 - \theta/T) \frac{\chi_s^0}{\chi_d^0} \frac{\{L + B(1+L) - i(B+L)D/(1 + 2\lambda\chi_s^0 + \chi_s^0/\chi_d^0)\}}{\{1 + B + 2\lambda\chi_s^0 + (1+L)\chi_s^0/\chi_d^0 - iD\}^2}. \quad (27)$$

Under the usual conditions of reflection e.s.r. we will have $L \ll 1$, $D \lesssim 1$ and $2\lambda\chi_s^0 \lesssim 1$. The expansion parameter will therefore be small in any of the following cases: (a) $B+L \rightarrow 0$ the bottlenecked limit, (b) $B \rightarrow \infty$ the unbottlenecked limit, (c) $\chi_s^0/\chi_d^0 \ll 1$ or (d) $T \rightarrow \theta$. We note that the coupling parameter λ only appears explicitly in condition (d). In a usual reflection experiment at least one of these conditions is likely to be valid, and the errors created by using only the first term in the expansion of equation (25), the only approximation made in this paper, will be negligible.

From equation (26) we may then obtain the g shift defined by $\Delta g = g \operatorname{Re} \Delta\omega_1/\gamma H$ and the linewidth defined by $DH = \operatorname{Im} \Delta\omega_1/\gamma$:

$$\Delta g = \frac{B^2 + L^2\chi_s^0/\chi_d^0 - 2\lambda\chi_s^0 BL}{\{1 + B + 2\lambda\chi_s^0 + (1+L)\chi_s^0/\chi_d^0\}^2 + D^2} \Delta g_0, \quad (28)$$

where $\Delta g_0 = g\lambda\chi_s^0$, and

$$DH = \frac{\{B(1+L) + L\}\{1 + B + 2\lambda\chi_s^0 + (1+L)\chi_s^0/\chi_d^0\} + (B+L)D^2/(1 + 2\lambda\chi_s^0 + \chi_s^0/\chi_d^0)}{\{1 + B + 2\lambda\chi_s^0 + (1+L)\chi_s^0/\chi_d^0\}^2 + D^2} \times K_0(T - \theta), \quad (29)$$

where $K_0 = \delta_{ds}/\gamma T$ is a constant which is independent of temperature because the Korringa relaxation rate δ_{ds} is itself proportional to temperature.

Discussion

The results of this paper, equations (28) and (29), may be compared with previous results by letting the quantities L , χ_s^0/χ_d^0 and $2\lambda\chi_s^0$ become much less than unity. The latter quantity is typically 0.1 for rare earth systems and five or ten times larger for 3d systems. We thus obtain

$$\Delta g = \frac{B^2}{(1+B)^2 + D^2} \Delta g_0, \quad (30)$$

$$DH = \frac{B(1+B+D^2)}{(1+B)^2 + D^2} K_0(T - \theta). \quad (31)$$

These two expressions are identical to those of Hasegawa (1959) except for one important feature. In Hasegawa's result the linewidth is proportional to T ; in ours it is proportional to $T - \theta$. Experimental data (Thân-Trong *et al.* 1976, 1977) favour

the latter dependence. Further, if we have shown that important terms in λ^2 occur in the expression for DH , we have also shown that they do not occur in the expression for Δg and that this quantity is independent of temperature above the Curie point apart from the implicit dependencies of B and D .

Finally, using the approximations of this section, we are able to obtain a simple expression for the dynamic susceptibility itself, equation (16):

$$\chi(\omega, H) = \frac{M_d(\gamma H + \Delta\omega_1)}{H(\gamma H + \Delta\omega_1 - \omega)}, \quad (32)$$

where $\Delta\omega_1$ is given by equations (30) and (31). This non-Lorentzian form of susceptibility is associated with relaxation to the instantaneous internal field (Spencer and Orbach 1968). For magnetically concentrated materials the spin-flip mean free path is much shorter than the skin depth and conduction electron diffusion effects are unimportant. The magnetic susceptibility is therefore a local quantity, and the skin effect will cause the measured absorption to have the form of the sum of the real and imaginary parts of equation (32).

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