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The Effect of Lattice Discreteness on the Linewidth of the Electron Spin Resonance of Local Moments in Metals

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Abstract

Solutions of the Hasegawa–Bloch equations for the electron spin resonance of local moments in metals are obtained which take account of the discrete nature of the lattice. The most notable result derived is that the resonance linewidth is proportional to $T-\theta$ for any degree of bottlenecking, where θ is the paramagnetic Curie temperature calculated with lattice discreteness. An alternative derivation of the appropriate Hasegawa–Bloch equations is also given.

1. Introduction

The coupled phenomenological mean field equations of Hasegawa (1959) and their various improvements and extensions have proved to be of great value in gaining an understanding of the spin dynamics of local moments in metals (Orbach *et al.* 1974; Taylor 1975; Barnes 1982). Recently, an accurate analytical approximate solution to Hasegawa's equations has been obtained (Stewart 1980) which allows the influence of the physical parameters in the equations on the resonance line position and width to be seen in a transparent manner. In particular, the solution showed that above the Curie temperature θ' the linewidth was proportional to $T-\theta'$ for any degree of bottlenecking. However, the equations to which the above solution was obtained did not take account of the discrete nature of the crystal lattice (Barnes 1974; Stewart 1975). In Section 2 of the present paper we review the problem of the lattice discreteness and provide an alternative derivation of the appropriate Hasegawa–Bloch equations. In Section 3 we show that the solution of the equations which involve discreteness may be obtained very simply from the solutions of those which do not, and that the linewidth remains proportional to $T-\theta$, where θ is now the paramagnetic Curie temperature of the system with lattice discreteness taken into account. Once again the result holds for an arbitrary degree of bottlenecking. In Section 4 we discuss the experimental verification of the predictions of the theory.

2. Destination Vectors

The Hasegawa–Bloch equations we use have the form

$$\partial \boldsymbol{M}_{\mathrm{d}}/\partial t = -\gamma \boldsymbol{M}_{\mathrm{d}} \times \boldsymbol{H}_{\mathrm{d}} - (\delta_{\mathrm{ds}} + \delta_{\mathrm{dL}})(\boldsymbol{M}_{\mathrm{d}} - \chi_{\mathrm{d}}^{0} \boldsymbol{H}_{\mathrm{d}}) + \delta_{\mathrm{sd}}(\boldsymbol{M}_{\mathrm{s}} - \chi_{\mathrm{s}}^{0} \boldsymbol{H}_{\mathrm{s}}), \qquad (1)$$

$$\partial \boldsymbol{M}_{\mathrm{s}}/\partial t = -\gamma \boldsymbol{M}_{\mathrm{s}} \times \boldsymbol{H}_{\mathrm{s}} - (\delta_{\mathrm{sd}} + \delta_{\mathrm{sL}})(\boldsymbol{M}_{\mathrm{s}} - \chi_{\mathrm{s}}^{0} \boldsymbol{H}_{\mathrm{s}}) + \delta_{\mathrm{ds}}(\boldsymbol{M}_{\mathrm{d}} - \chi_{\mathrm{d}}^{0} \boldsymbol{H}_{\mathrm{d}}), \qquad (2)$$

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where the destination vectors for the s and d magnetizations are $\chi_s^0 \boldsymbol{H}_s$ and $\chi_d^0 \boldsymbol{H}_d$ respectively (see Barnes 1982 for a detailed discussion of the various relaxation parameters). It is the purpose of this section to determine expressions for these destination vectors. The physical system we consider is a metal of volume V consisting of N atoms with N^* atoms carrying localized moments (the d system) lying on a Bravais lattice L. Other symbols used have been defined before by Stewart (1975, 1980); these papers should be consulted for further details.

The driving fields $\boldsymbol{H}_d$ and $\boldsymbol{H}_s$ and the associated destination vectors are obtained from the instantaneous fields acting on the appropriate system (Stewart 1975). The first of these is

$$\boldsymbol{H}_s = \boldsymbol{H} + \lambda \boldsymbol{M}_d + \lambda_s \boldsymbol{M}_s, \tag{3}$$

where we have arbitrarily added the term $\lambda_s \boldsymbol{M}_s$ to simulate the effect of exchange enhancement of the conduction electrons. We shall see shortly that it is more profitable to absorb it into other parameters.

The driving field $\boldsymbol{H}_i$ acting on local moment i is (Stewart 1975; equation 8)

$$\boldsymbol{H}_i = \boldsymbol{H} + \lambda \boldsymbol{M}_s + \frac{2}{g_s g_d \mu_\beta^2 N} \sum_{\boldsymbol{q}}{}' J(\boldsymbol{q}) \exp(\mathrm{i}\boldsymbol{q} \cdot \boldsymbol{R}_i) \langle \boldsymbol{m}(\boldsymbol{q}) \rangle, \tag{4}$$

where the prime on the wavevector sum indicates that the $\boldsymbol{q} = \boldsymbol{0}$ term is excluded; this $\boldsymbol{q} = \boldsymbol{0}$ term is explicitly written as $\lambda \boldsymbol{M}_s$. The expression for λ is $(2V/g_s g_d \mu_\beta^2 N) J(\boldsymbol{0})$. The key assumption in the derivation is that while the $\boldsymbol{q} = \boldsymbol{0}$ component of the conduction electron s magnetization is retained as a variable in the theory, the $\boldsymbol{q} \neq \boldsymbol{0}$ components are calculated on the basis that the local moment system is static. The validity of this approximation was discussed by Barnes (1974), who argued that the dynamic part of the indirect exchange interaction was carried by a process which, at finite temperature, had the character of a diffusive mode of the conduction electrons with a characteristic diffusion length d much longer than the lattice spacing. Viewing the situation in the extended zone scheme of reciprocal space, we see that modes with $q \geqslant 2\pi d^{-1}$ will be propagated but that modes with $q \leqslant 2\pi d^{-1}$ will not. Since, in a periodic lattice, only modes with $\boldsymbol{q}$ equal to a reciprocal lattice vector $\boldsymbol{G}$ will be excited, it appears that the different way of treating the modes with $\boldsymbol{q} = \boldsymbol{0}$ and $\boldsymbol{q} \neq \boldsymbol{0}$ is plausible, since the modes with $q \approx 2\pi d^{-1}$ lie in between $\boldsymbol{q} = \boldsymbol{0}$ and the nearest reciprocal lattice vectors and are well separated from the latter. However, in systems in which the wavevector is not conserved, such as random alloys or amorphous metals, important modes could have $q \leqslant 2\pi d^{-1}$ and the above assumption may be questionable.

Nonetheless, proceeding in this way, we can use the relation $\langle \boldsymbol{m}(\boldsymbol{q}) \rangle = \chi^0(\boldsymbol{q}) \boldsymbol{H}(\boldsymbol{q})$ for $\boldsymbol{q} \neq \boldsymbol{0}$, where $\chi^0(\boldsymbol{q})$ is the conduction electron susceptibility (Stewart 1975). The Fourier component $\boldsymbol{H}(\boldsymbol{q})$ of the effective field on the conduction electrons is (for $\boldsymbol{q} \neq \boldsymbol{0}$)

$$\boldsymbol{H}(\boldsymbol{q}) = -\frac{2V}{g_s \mu_\beta N} J(-\boldsymbol{q}) \left(\exp(-\mathrm{i}\boldsymbol{q} \cdot \boldsymbol{R}_i) \langle \boldsymbol{S}_i \rangle + \sum_{n \neq i} \exp(-\mathrm{i}\boldsymbol{q} \cdot \boldsymbol{R}_n) \langle \boldsymbol{S}_n \rangle \right), \tag{5}$$

where we have explicitly separated the effect due to local moment i (the self-polarization term) from the other terms. Equation (5) is then substituted into (4) to give

$$H_i = H + \lambda M_s + M_d\left(\frac{2V}{g_s g_d \mu_\beta^2 N}\right)^2 \sum_q{}' \,|J(q)|^2 \chi^0(q) N^{*-1}$$
$$\times \left((\langle S_i\rangle/\langle S\rangle) + \sum_{n\neq i} \exp\{\mathrm{i}q\,.(R_i - R_n)\}\right), \tag{6}$$

where we have assumed that all local moments have the same average spin $\langle S\rangle$. The sum over n gives $N^*\delta_{q,G}-1$, and omitting the self-polarization term (Stewart 1978) and letting $N^* \to \infty$, we obtain

$$H_d = H + \lambda M_s + \alpha M_d\,, \tag{7}$$

where $H_d = H_i$ is the same for each local moment and

$$\alpha = \left(\frac{2V}{g_s g_d \mu_\beta^2 N}\right)^2 \left(\sum_G |J(G)|^2 \chi^0(G) - N^{*-1} \sum_q |J(q)|^2 \chi^0(q) - |J(0)|^2 \chi^0(0)\right). \tag{8}$$

Since the first two terms in equation (8) are equal to θ/C, where θ is the paramagnetic Curie temperature and C is the Curie constant of the local moments (Stewart 1975), we arrive at an expression equivalent to that given by Barnes (1974, 1982).

3. Solution of Discrete Lattice Equations

We now need to find solutions to the Hasegawa–Bloch equations (1)–(3) and (7) which take account of the discrete nature of the lattice. First we note that the terms in (3) and (7) proportional to λ_s and α will have no effect on the torque terms because of the existence of the vector cross product. Further, by defining the exchange enhanced susceptibilities $\chi_d = \chi_d^0(1-\alpha\chi_d^0)^{-1}$ and $\chi_s = \chi_s^0(1-\lambda_s\chi_s^0)^{-1}$ (note that the symbols χ_d and χ_s were defined differently by Stewart 1980), we obtain

$$\partial M_d/\partial t = -\gamma M_d \times (H + \lambda M_s) + \delta_{sd}(1-\lambda_s\chi_s^0)\{M_s - \chi_s(H+\lambda M_d)\}$$
$$-(\delta_{ds}+\delta_{dL})(1-\alpha\chi_d^0)\{M_d - \chi_d(H+\lambda M_s)\}\,, \tag{9}$$

$$\partial M_s/\partial t = -\gamma M_s \times (H + \lambda M_d) + \delta_{ds}(1-\alpha\chi_d^0)\{M_d - \chi_d(H+\lambda M_s)\}$$
$$-(\delta_{sL}+\delta_{sd})(1-\lambda_s\chi_s^0)\{M_s - \chi_s(H+\lambda M_d)\}\,. \tag{10}$$

We note that exchange enhancement may also affect other parameters implicitly: $\chi^0(q)$ will become enhanced to $\chi(q) = \chi^0(q)\{1-\lambda_s\chi^0(q)\}^{-1}$ where λ_s is an interaction parameter [$\chi_s = \chi(0)$], the relaxation rates will become renormalized (Barnes 1982), and the dynamical range of the indirect exchange interaction will become shorter. We note also that if for $\alpha = 0$ and $\lambda_s = 0$ the detailed balance condition $\chi_s^0/\chi_d^0 = \delta_{ds}/\delta_{sd}$ holds, then for α and λ_s nonzero the detailed balance relation $\chi_s/\chi_d = \delta_{ds}(1-\alpha\chi_d^0)/\delta_{sd}(1-\lambda_s\chi_s^0)$ is valid also.

Since equations (9) and (10) have the same mathematical structure as those with $\alpha = \lambda_s = 0$, the solutions to the former may be obtained from the solutions to the latter by making an appropriate redefinition of the parameters. In particular, the static susceptibility $\chi = (M_d + M_s)/H$ is given by

$$\chi = \chi_s + \chi_d^0(1+\lambda\chi_s)^2(1-\theta/T)^{-1}, \tag{11}$$

where

$$\theta/T = \chi_d^0(\alpha+\lambda^2\chi_s). \tag{12}$$

It is convenient in equations (11) and (12) to use the enhanced conduction electron susceptibility χ_s, since this is what is measured in the non-magnetic host, whereas the bare local moment susceptibility would often be known *a priori* to follow a Curie law. Expressions for the g shift Δg and linewidth DH of the resonance of the dynamic susceptibility become (see Stewart 1980; equations 28 and 29)

$$\Delta g = \frac{B^2 + L^2\chi_s/\chi_d - 2\lambda\chi_s BL}{\{1+B+2\lambda\chi_s+(1+L)\chi_s/\chi_d\}^2 + D^2}\Delta g_0, \tag{13}$$

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

$$DH = \frac{\{L(1+B)+B\}\{1+B+2\lambda\chi_s+(1+L)\chi_s/\chi_d\} + (B+L)D^2/(1+2\lambda\chi_s+\chi_s/\chi_d)}{\{1+B+2\lambda\chi_s+(1+L)\chi_s/\chi_d\}^2+D^2}$$
$$\times K_0(T-\theta), \tag{14}$$

where $K_0 = \delta_{ds}/\gamma T$ is a constant independent of temperature, because the Korringa relaxation rate δ_{ds} is itself proportional to temperature, $B = \delta_{sL}/\delta_{sd}$, $L = \delta_{dL}/\delta_{ds}$, θ is given by equation (12), and $D = \gamma\lambda\chi_s H(1+2\lambda\chi_s+\chi_s/\chi_d)/\delta_{ds}(1-\theta/T)$. We see that the solutions for nonzero α and λ_s are obtained from the solutions with these quantities zero simply by letting $\chi_s^0 \rightarrow \chi_s$ and $\chi_d^0 \rightarrow \chi_d$ and taking θ to be the paramagnetic Curie temperature with the discreteness of the lattice incorporated. One particular conclusion we can come to from equation (14) is that the linewidth is predicted to be proportional to $T-\theta$, where θ is the measured paramagnetic Curie temperature. We also note that for $\chi_s/\chi_d \ll 1$ the lineshape will have the non-Lorentzian form given by equation (32) of Stewart (1980).

4. Discussion

Equations (1)–(3) and (7) were proposed by Cottet *et al.* (1968) who showed, in the unbottlenecked limit only, that they gave rise to a linewidth proportional to $T-\theta$. However, Cottet *et al.* were not able to provide any microscopic basis for their equations and in particular were unable to give an explanation for the origin of the term in α. Around the same time (Dupraz *et al.* 1970) it became apparent that the equations which did not take account of the discreteness of the lattice gave a linewidth proportional to $T-\theta'$ in the bottlenecked limit, where θ' is the lattice average Curie temperature. Subsequently, Barnes (1974) showed that the term in α arose from the discreteness of the lattice and that consequently the linewidth would be expected to be proportional to $T-\theta$ in both the bottlenecked and unbottlenecked limits. Solutions to the equations which did not take account of the discreteness of the lattice (Stewart 1980) produced a linewidth proportional to $T-\theta'$ for any degree of bottlenecking. In the present paper it has been shown that a linewidth proportional to $T-\theta$ is expected for any degree of bottlenecking when the discreteness of the lattice is taken into account. Of course, although the solutions obtained here follow rigorously from the Hasegawa–Bloch equations, these equations themselves have not yet been justified in a system with a large concentration of magnetic atoms.

Experimentally, it has not yet been fully clarified whether the linewidth does have a $T-\theta$ dependence. An investigation of this question by Thân-Trong *et al.* (1981) on $Gd_{1-x}Dy_xAl_2$ and $Gd_{1-x}Sm_xAl_2$ showed that when the linear part of the linewidth is extrapolated back to the transition temperature (which is a function of x) a constant small positive intercept is obtained. From equation (14), which in the appropriate limit has the form

$$DH = \{B/(1+B)\}K_0(T-\theta)+\gamma^{-1}\delta_{\mathrm{dL}}(1-\theta/T), \tag{15}$$

we might associate this intercept with δ_{dL}, although other causes may be important (Barnes 1982). However, the equations developed in the present paper cannot be directly applied to these two compounds since they contain the extra magnetic component Dy or Sm. A calculation by Chiu and Stewart (1982) showed that even in this case one would expect a linewidth proportional to $T-\theta'$; however, this calculation did not take account of the discrete nature of the lattice. The question of the $T-\theta$ variation of the linewidth therefore remains open and it would be valuable to carry out measurements on materials with negative θ, as in this case $|\alpha|$ would be largest and any effect it had upon the resonance presumably most noticeable.

References

Barnes, S. E. (1974). *J. Phys.* F **4**, 1535.
Barnes, S. E. (1982). *Adv. Phys.* **30**, 801.
Chiu, L. B., and Stewart, A. M. (1982). *J. Phys. Chem. Solids* **43**, 167.
Cottet, P., Donzé, P., Dupraz, J., Giovannini, B., and Peter, M. (1968). *Z. Angew. Phys.* **24**, 249.
Dupraz, J., Giovannini, B., Orbach, R., Riley, J. D., and Zitkova, J. (1970). 'Magnetic Resonance' (Eds C. K. Coogan *et al.*), pp. 197–226 (Plenum: New York).
Hasegawa, H. (1959). *Prog. Theor. Phys. (Kyoto)* **21**, 483.
Orbach, R., Peter, M., and Shaltiel, D. (1974). *Arch. Sci.* **27**, 141.
Stewart, A. M. (1975). *Aust. J. Phys.* **28**, 685.
Stewart, A. M. (1978). *Phys. Rev.* B **17**, 2388.
Stewart, A. M. (1980). *Aust. J. Phys.* **33**, 1049.
Taylor, R. H. (1975). *Adv. Phys.* **24**, 681.
Thân-Trong, N., Chiu, L. B., Elliston, P. R., Stewart, A. M., and Taylor, K. N. R. (1981). *J. Phys.* F **11**, 1123.

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