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DYNAMICAL SUSCEPTIBILITY OF A DILUTE MAGNETIC ALLOY IN THE ANDERSON MODEL *

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ABSTRACT

We have derived spin transport equations in the magnetic limit of the Anderson model. The corresponding Bloch equations are found to differ in several ways from those obtained in the exchange model. For $g_e \neq g_d$, the magnetizations transferred between the localized and conduction electrons by the admixture are not equal and their difference alters the resonance condition. The relaxation terms are of the form « towards the local instantaneous field », the internal field experienced by the conduction electrons being proportional to the difference between the redistribution and admixture polarizations. Using the Schrieffer-Wolff transformation, our results are found to be compatible with the exchange model results.

INTRODUCTION

Numerous experimental data of *EPR* in dilute magnetic alloys have been analyzed following Hasegawa's [1] suggestion: The motion of the conduction and localized electron spins in magnetic field is described by two coupled Bloch-type equations. The coupling between the two spin systems is usually considered to be of the exchange, $JS \cdot \sigma$, type. Using the exchange model, several recent microscopic calculations [2-4] attempted to derive the spin transport equations. Though these calculations were performed to various degrees of accuracy and differ in some of their partial results, they all calculate the *ESR* g -shift to be proportional to the polarization of the conduction electrons ($\Delta g_d \approx \lambda M_e^z$) and confirm the essential correctness of the Hasegawa's original approach, with the form of the relaxation terms "towards the equilibrium values in the local (including internal) instantaneous field".

A more fundamental Anderson Hamiltonian [5] pictures the coupling between the two spin systems in terms of the mixing interaction V_{kd} . With respect to the exchange, Anderson model has the advantage of being able to distinguish between two contributions to the conduction electron polarization; the redistribution (adjustment of the electronic population due to the energy level shifts) and the admixture (transfer of the magnetization due to the mixing of the wave functions)

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effects. Though the latter effect has no analogy in the exchange model, and covalent mixing is important, there has been only one attempt made, by Sasada and Hasegawa [6], to derive Bloch equations in the Anderson model. This calculation assumed the density of states of the conduction electrons to be constant ($\rho(\varepsilon_k) = \text{constant}$) and the g -factors $g_e = g_d$. However, according to the compensation theorem [5], in the limit when $\rho(\varepsilon_k) = \text{constant}$, polarization by the redistribution is just equal (and opposite in sign) to polarization by the admixture effect. Sasada and Hasegawa obviously considered only this case when the two effects cancelled. In addition, and contrary to the exchange model results [2-4], these authors conclude that the spins relax towards their equilibrium values in the instantaneous external field only.

In order to get some insight into the redistribution and admixture polarizations of conduction electrons (e.g., how will the admixture affect the g -shift) and to resolve the discrepancy between the relaxation "destination", it was desirable to calculate the dynamic susceptibility of a dilute alloy for a general (non-constant) density of states of conduction electrons and $g_e \neq g_d$.

In this paper we consider a magnetic impurity described by Anderson Hamiltonian [5]. To distinguish between the redistribution and admixture effects we consider $\rho(\varepsilon_k) \neq \text{constant}$ and $g_e \neq g_d$. To obtain the dynamical "bottleneck" aspects, we include a term describing scattering of conduction electrons off non-magnetic impurities (distinct from the magnetic) by a potential which has a non-spin flip part as well as a spin-flip part from the spin-orbit interaction. We focus upon the admixture and redistribution effects and establish a relationship between spin transport equations in the Anderson (magnetic limit, $S = 1/2$) and exchange models.

MODEL AND RESULTS

Anderson Hamiltonian in the familiar notation [5] is written as

$$\mathcal{H} = \sum_{k\sigma} \varepsilon_{k\sigma} a_{k\sigma}^+ a_{k\sigma} + \sum_{i\sigma} E_{i\sigma} c_{i\sigma}^+ c_{i\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} + \frac{V}{\sqrt{N}} \sum_{k\sigma} e^{-ik \cdot R_j} (a_{k\sigma}^+ c_{i\sigma} + c.c.) + \mathcal{H}_{eo}, \quad (1)$$

where the first and second terms are the noninteracting Hamiltonians of the conduction and localized electrons. The third term correlates the localized electrons and the fourth term transfers electrons from the conduction onto localized levels. In the presence of the static magnetic field h_z , energies $\varepsilon_{k\sigma} = \varepsilon_k - \sigma(\omega_e/2)$, $E_{i\sigma} = E^0 - \sigma(\omega_d/2)$; $\sigma = \pm 1$ (or \uparrow, \downarrow) is the spin label. Index i labels positions of the magnetic impurities of concentration c .

$$\mathcal{H}_{eo} = \frac{1}{N} \sum e^{-i(k-k') \cdot R_j} a_{k'\alpha}^+ (A \hat{1}_{\alpha\beta} + iB [\hat{k} \times \hat{k}'] \cdot \sigma_{\alpha\beta}) a_{k\beta} \quad (2)$$

describes the scattering of the conduction electrons off non-magnetic impurities (of concentration c') by a potential which has a non spin-flip (A) as well as a spin-flip part (iB) from the spin-orbit interaction. $\hat{1}_{\alpha\beta}$ and $\sigma_{\alpha\beta}$ are the unit and Pauli spin matrices respectively.

The transverse dynamic susceptibility $\chi^+(\omega)$ is the Fourier transform of the retarded response function

$$i\theta(t) \frac{1}{N} \langle M^+(t), M^-(0) \rangle; \quad M^\pm = g_d \mu_B \sum_i S_i^\pm + (1/2) g_e \mu_B \sigma^\pm(0). \quad (3)$$

We have evaluated [8] the response function $\chi^{+-}(\omega)$ using the diagrammatic methods. The Coulomb interaction term $Un_\uparrow n_\downarrow$ correlating the localized electrons has been treated in the *RPA* approximation and we have assumed the ground state of the impurity to be magnetic in the sense of Anderson [5]. Interference between different impurities has not been considered, and we have assumed the non spin-flip scattering cross section to be larger than the spin-orbit scattering ($A^2 \gg B^2$). Though not necessary, the latter assumption is a convenient one; strong potential scattering redistributes electronic momenta rapidly over the Fermi surface and suppresses [4] thus the angular dependence of the spin-orbit scattering. The susceptibility equations we have derived [8] are, in the Hartree-Fock magnetic limit, exactly equivalent to the following set of linearized spin transport equations

$$\begin{aligned} \frac{d}{dt} \tilde{M}_d^A &= g_d^A [\tilde{M}_d^A \times (h + \lambda^A \tilde{M}_e^A)] + g_d^A \alpha [\tilde{M}_d^A \times h] \\ &\quad - \delta_{de}^A \delta \tilde{M}_d^A + (g_d^A/g_e^A) \delta_{ed}^A \delta \tilde{M}_e^A \end{aligned} \quad (4a)$$

$$\begin{aligned} \frac{d}{dt} \tilde{M}_e^A &= g_e^A [\tilde{M}_e^A \times (h + \lambda^A \tilde{M}_d^A)] - g_e^A \alpha [\tilde{M}_e^A \times h] \\ &\quad - (\delta_{ed}^A + \delta_{e0} + D\nabla^2) \delta \tilde{M}_e^A + (g_e^A/g_d^A) \delta_{de}^A \delta \tilde{M}_d^A \end{aligned} \quad (4b)$$

where $\tilde{M}_i^A(t)$ and $h(t)$ are the instantaneous values of the magnetization and magnetic field respectively, δ_{ij}^A are the cross-relaxation rates ($i, j = e, d$ designate the conduction and localized spins, superscript A means values as evaluated in the Anderson model),

$$\delta_{ed}^A = \pi \rho_F J^2 (1 + A); \quad \delta_{de}^A = \pi \rho_F^2 J^2 \omega_d^A (1 + \alpha + \lambda^A \chi_e^{z,A}). \quad (5)$$

δ_{e0} ($\delta_{e0} = (16\pi/9) c' \rho_F B^2$) is the usual electron-lattice relaxation rate due to the spin orbit interaction and the diffusion constant $D = v_F^2 \tau/3$, where τ ($\tau^{-1} = 2\pi c' \rho_F A^2$) the usual transport time. The exchange field parameter $\lambda^A = 2J/g_e^A g_d^A$ with

$$J = V_{kd}^2 (E_{d\downarrow} - E_{d\uparrow})/E_{d\uparrow} E_{d\downarrow}; \quad J < 0, \quad (6)$$

which is the value given by the well-known Schrieffer-Wolff transformation [9]. ρ_F is the density of states of the conduction electrons at the Fermi surface, $\omega_d^A = g_d^A h^z$ and the static susceptibility $\chi_e^{z,A} = M_e^{z,A}/h^z$. The departures from the equilibrium values in the local instantaneous fields are

$$\delta \tilde{M}_d^A = \tilde{M}_d^A - \chi_d^{0,A} [h(1 + \alpha) + \lambda^A \tilde{M}_e^A] \quad (7a)$$

$$\delta \tilde{M}_e^A = \tilde{M}_e^A - \chi_e^0 [h + \beta \tilde{M}_d^A] \quad (7b)$$

where $\chi_e^0 = g_e^2 \rho_F / 2$ and where we have used the low temperature definition

$$\chi_d^{0,A} = M_d^{z,A} / [h_z(1 + \alpha) + \lambda^A M_e^{z,A}] \quad (8)$$

All alternations of the Bloch equations due to the admixture effect are described by parameters α and β

$$\alpha = A(1 - g_e^A/g_d^A) \quad (9)$$

$$\beta = g_e^A(\rho_F J - A) / g_d^A \chi_e^0(1 + A) \quad (10)$$

where the admixture parameter A has its usual value [10]

$$A = (V^2/N) \sum_k \{ [(f_k - 1)/(\epsilon_k - E_{d\uparrow})^2] - f_k / (\epsilon_k - E_{d\downarrow})^2 \}. \quad (11)$$

DISCUSSION

Equations 4a and 4b differ in several ways from the Bloch equations derived in the exchange model:

The driving torque terms of equations 4a and 4b contain an additional torque term for the quantity $(g_d^A - g_e^A) A \tilde{M}_d^A$, which is the difference between the magnetizations transferred between the localized and conduction electrons by the admixture. This term arises since, for $g_e^A \neq g_d^A$, mixing does not conserve the magnetization transferred between the two species of electrons. It is straightforward to verify that, if the dynamical bottleneck is broken, equation 4a yields the resonance position at $\omega_{res}^A = \omega_d^A (1 + \alpha + \lambda^A \chi_e^{z,A})$. That is, unlike in the exchange model (where $\Delta g_d^{Ex} h^z = \omega_d^A \lambda^{Ex} \chi_e^{z,Ex}$), the g -shift is comprised from three parts,

$$\Delta g_d^A h^z = (\omega_{res}^A - \omega_d^A) = \omega_d^A \lambda^A \chi_e^{z,A} - \omega_e^A A + \omega_d^A A. \quad (12)$$

The first and second terms in equation 12 are the polarizations of the conduction electrons due to the redistribution (around the Fermi surface) and the admixture effects respectively and the last term describes the delocalization of the impurity. The polarization redistribution is proportional to effective exchange constant J of the Schrieffer-Wolff transformation (equation 6) and the difference between the

two admixed magnetizations (mixing conserves the spin but not the magnetization) is proportional to $(g_e^A - g_d^A)$. We thus conclude: (a) Even if the density of states of conduction electrons is constant (and the total internal polarization ($\Delta M_e \approx J\rho_F - A = 0$), there is a finite g -shift in ESR ¹,

$$\Delta g_d^A h^z = \omega_d^A A = (g_d^A/g_e^A) \omega_d^A \lambda^A \chi_e^{o,A}, \quad (13)$$

due to the delocalization effect. Its magnitude is enhanced from that contributed by the exchange field by ratio g_d^A/g_e^A and proportional to the unenhanced Pauli susceptibility $\chi_e^{o,A}$. (b) For $g_e^A = g_d^A$, the dephasing introduced by the "high frequency" term $\omega_d^A \alpha = (\omega_d^A - \omega_e^A) A$ (the difference between the two admixed magnetizations) vanishes. The g -shift is entirely due to the exchange field

$$\Delta g_d^A h^z = \omega_d^A \lambda^A \chi_e^{z,A}. \quad (14)$$

The admixture effect alters (as compared to the exchange model results) also the relaxation terms. In the departure δM_d^A , equation 7a, the destination vector for the localized spin relaxation contains an additional term proportional to the difference between the two admixed magnetizations, $(g_d^A - g_e^A) A \chi_d^{o,A} h$. The conduction electron destination vector in the departure δM_e^A , equation 7b, is also modified (the β parameter), by the admixed polarization $g_e^A A M_d^A$. The internal polarization which determines the relaxation of the conduction electrons is given by the difference between the redistribution and admixture effects and vanishes, in accordance with the compensation theorem, for $\rho(\epsilon_k) = \text{constant}$ (for $\rho(\epsilon_k) = \rho_F$, $\rho_F J \equiv A$, i.e. $\beta \equiv 0$). We wish to emphasize that both departure δM_d^A and δM_e^A (equations 7a and 7b) are of the form "relaxation towards the instantaneous internal field" and our results do differ, therefore, (even under the restrictions that $\rho(\epsilon_k) = \text{constant}$ and $g_e^A = g_d^A$) from those of Sasada and Hasegawa [6].

We should also check the analytical behavior of equations 4a and 4b. First, it is straightforward to show that for $\omega = 0$ both departures δM_d^A and δM_e^A (which identically vanish) and the torque terms of equations 4a and 4b yield independently

$$\begin{aligned} \chi_d^{+,A}(0) &= \chi_d^{o,A} [1 + \alpha + \lambda^A \chi_e^{+,A}(0)] = [\chi_d^{o,A}(1 + \alpha) + \lambda^A \chi_e^{o,A} \chi_d^{o,A}] / [1 - \lambda^A \beta \chi_e^{o,A} \chi_d^{o,A}] \\ \chi_e^{+,A}(0) &= \chi_e^{o,A} [1 + \beta \chi_d^{+,A}(0)] = [\chi_e^{o,A} + \beta(1 + \alpha) \chi_d^{o,A}] / [1 - \lambda^A \beta \chi_e^{o,A} \chi_d^{o,A}], \end{aligned} \quad (15)$$

i.e. the values which are just equal to the static enhanced susceptibilities $\chi_d^{z,A}$ and $\chi_e^{z,A}$.

Secondly, in order to determine the so-called "detailed balance" condition, we relate the cross-transfer susceptibilities $\chi_{de}^{+,A}(\omega)$ and $\chi_{ed}^{+,A}(\omega)$. The susceptibilities $\chi_{dd}^{+,A}$, $\chi_{ee}^{+,A}$, $\chi_{de}^{+,A}$ and $\chi_{ed}^{+,A}$ are identified as the diagonal and off diagonal components respectively of the total, 4×4 matrix, susceptibility function $\chi^{+,A}(\omega)$ (see equation 3).

¹ This is unlike in NMR, where the Knight shift is given by the polarization of the conduction electrons only.

Having evaluated $\chi_{de}^{+,A}(\omega)$ and $\chi_{ed}^{+,A}(\omega)$ we find that they are equal (i.e. the magnetization scattering-in and scattering-out rates are equal) for any ω only if the relation

$$\delta_{de}^A \chi_d^{0,A} / (g_d^A)^2 = \delta_{ed}^A \chi_e^{0,A} / (g_e^A)^2 \quad (16)$$

holds. Equation 16 is the so-called detailed balance condition; though its *algebraic* equality could have been calculated using Equation 5, the present derivation is physically a correct one. It reflects first the cross-transfer character and secondly the dynamical nature of this relation (at $\omega = 0$, $\chi_{de}^{+-}(0) \equiv \chi_{ed}^{+-}(0)$ regardless of any relation between the values of the scattering rates).

Finally, we check that the absorption (proportional to $\text{Im} \chi^{+,A}(\omega + i\delta)$) is positive definite. We have solved equations 4a and 4b for the total susceptibility $\chi^{+,A}(\omega + i\delta)$ and found that if the detailed balance relation holds, $\text{Im} \chi^{+,A}(\omega + i\delta)$ reduces to a quadratic form, which for $\omega > 0$ is positive definite. In leading orders of both the magnetization and the interaction parameters,

$$\text{Im} \chi^{+,A}(\omega + i\delta) \approx \omega [(g_d^A - g_e^A)^2 \delta_{ed}^A + |C| \delta_{e0}] \quad (17)$$

where C is a function of parameters defined by equations 5-11. The result is quite physical. The absorption is non-vanishing only if either from the two dephasing conditions is fulfilled: $\delta_{e0} \neq 0$ (in the absence of the direct impurity-lattice relaxation) or $g_e \neq g_d$.

SPIN TRANSPORT EQUATIONS IN THE ANDERSON AND EXCHANGE MODELS

There have been claims made recently [3] that the exchange model's and Sasada and Hasegawa's results [6] are identical; we shall find that these claims are incorrect and that it is the present result, equation 4, which is compatible with the spin transport equations derived in the exchange model.

In the magnetic limit, the Anderson and exchange Hamiltonians are related by Schrieffer-Wolff canonical transformation [9]:

$$\mathcal{H}_{Ex} \rightarrow e^S \mathcal{H}_A e^{-S}; \quad \psi_{Ex} \rightarrow e^S \psi_A \quad (18)$$

$$\text{where } S = \sum_{ks\alpha} V n_{d,-s}^\alpha \frac{c_{ks}^+ d_s}{\epsilon_k - E_\alpha} - H.C.$$

Using equation 18, the localized and conduction spin expectation values are, to order V^2 , transformed respectively as

$$\langle \varphi \rangle^A \simeq \langle \varphi \rangle^{Ex} + \langle S [S, \varphi]_- \rangle^{Ex} \simeq \langle \varphi \rangle^{Ex} (1 + A) \quad (19a)$$

$$\langle s \rangle^A \simeq \langle s \rangle^{Ex} + \langle S [S, s]_- \rangle^{Ex} \simeq \langle s \rangle^{Ex} - A \langle \varphi \rangle^{Ex}, \quad (19b)$$

where the superscripts A and Ex mean the averages evaluated in the space of the wave functions ψ_A and ψ_{Ex} respectively and the parameter A is the admixture

defined by equation 11. Notice, that the total spin remains conserved by the transformation. The total magnetization as a physical variable is also representation-independent,

$$M_d^A + M_e^A = M_d^{Ex} + M_e^{Ex}; \quad M_d^{A,Ex} = g_d^{A,Ex} \varphi^{A,Ex}; \quad M_e^{A,Ex} = g_e^{A,Ex} s^{A,Ex}. \quad (20)$$

States ψ_A and ψ_{Ex} are in general characterized by different g -factors. Using eqs. 19 and 20 we obtain the transformation of the g -factors,

$$g_e^A = g_e^{Ex} = g_e; \quad g_d^A (1 + \alpha) = g_d^{Ex}, \quad (21)$$

which, combined with equation 20 yields

$$M_d^A = M_d^{Ex} (1 + A) / (1 + \alpha); \quad M_e^A = M_e^{Ex} - A M_e^{Ex} (g_e / g_d^{Ex}). \quad (22)$$

Equations 4a and 4b are transformed from the ψ_A to the ψ_{Ex} space by using relations 22. We obtain:

$$\frac{d}{dt} \tilde{M}_d^{Ex} = \left(\frac{1 + \alpha}{1 + A} \right) \frac{d}{dt} \tilde{M}_d^A = g_d^{Ex} [\tilde{M}_d^{Ex} x (h + \lambda^{Ex} \tilde{M}_d^{Ex})] - \delta_{ed}^{Ex} \delta \tilde{M}_d^{Ex} + (g_d^{Ex} / g_e) \delta_{ed}^{Ex} \delta \tilde{M}_e^{Ex} \quad (23a)$$

$$\frac{d}{dt} \tilde{M}_e^{Ex} = \frac{d}{dt} \left(\tilde{M}_e^A + \frac{g_e}{g_d^{Ex}} A \tilde{M}_d^{Ex} \right) = g_e [\tilde{M}_e^{Ex} x (h + \lambda^{Ex} \tilde{M}_d^{Ex})] - (\delta_{ed}^{Ex} + \delta_{eo} + D \nabla^2) \delta \tilde{M}_e^{Ex} + (g_e / g_d^{Ex}) \delta_{de}^{Ex} \delta \tilde{M}_d^{Ex} \quad (23b)$$

where the departures $\delta \tilde{M}_d^{Ex}$ and $\delta \tilde{M}_e^{Ex}$ are

$$\delta \tilde{M}_d^{Ex} = \tilde{M}_d^{Ex} - \chi_d^{o,Ex} (h + \lambda^{Ex} \tilde{M}_d^{Ex}); \quad \delta \tilde{M}_e^{Ex} = \tilde{M}_e^{Ex} - \chi_e^o (h + \lambda^{Ex} \tilde{M}_d^{Ex}), \quad (24)$$

and where the transformed susceptibility $\chi_d^{o,Ex}$ and the relaxation rates δ_{ed}^{Ex} and δ_{de}^{Ex} are given by

$$\chi_d^{o,Ex} = M_d^{z,Ex} (h_z + \lambda^{Ex} M_e^{z,Ex}) \quad (25)$$

$$\delta_{de}^{Ex} = \delta_{ed}^A (1 + A)^{-1} = \pi \rho_F J^2$$

$$\delta_{de}^{Ex} = \delta_{de}^A \chi_d^{o,A} (1 + \alpha)^2 / \chi_d^{o,Ex} (1 + A) = \pi \rho_F^2 J^2 \omega_d^{Ex} (1 + \lambda^{Ex} \chi_e^{z,Ex}) \quad (26)$$

and where, to obtain the final form of equations 25 and 26 we have substituted for δ_{ed}^A , δ_{de}^A and $\chi_d^{o,A}$ their actual values, equations 5 and 8.

Equation 23 is the transformed version of equation 4, i.e. Bloch equations in the Anderson model transformed by Schrieffer-Wolff transformation from the ψ_A to the ψ_{Ex} space. The transformation was performed with the help of transformation equation 22 *only*, without using any form (assumed or derived) of the so-called "detailed balance relation", the use of which might (as it actually did in Ref. 3)

blur the distinction between the controversial concept of the relaxation (towards the internal or the external field only). They are first, of exactly the same form as the spin transport equations derived in the exchange model [2-4] and secondly, the values of δ_{de}^{Ex} , δ_{ed}^{Ex} , $\chi_d^{o,Ex}$ (equations 25 and 26) are not redefined arbitrarily (as was the case in Ref. 3), but, in fact, do agree with their low temperature values as calculated [2] in the exchange model. We thus see that the derived [8] Bloch equations, equation 4, are compatible (for general g -factors, $g_d \neq g_e$) with the exchange model results. We should point out that the fact that the calculated values of the relaxation rates δ_{de} and δ_{ed} are their low temperature values [2, 4] is not surprising. The present work is (as well as all other similar calculations) essentially the low temperature calculation; the strong Coulomb interaction $Un_{d\uparrow}n_{d\downarrow}$ term has been treated in the *RPA* approximation and the Hartree-Fock levels $E_{d\sigma} = E_{d\sigma}^0 + U \langle n_{d,-\sigma} \rangle$ assumed to lie outside the reach by the thermal fluctuations. It is due to the absence of the thermal fluctuations that the transverse contributions to the cross-transfer rates have been neglected. However, the present calculation does account for all but the thermal fluctuation dynamical processes. Therefore, we argue that as long as $U \gg \Gamma$ (Γ/U is the expansion parameter in our theory), the present form of the Bloch equations 4a and 4b is preserved for finite temperatures provided that δ_{de} , δ_{ed} and susceptibility χ_d^o are replaced by their high temperature values.

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DISCUSSION

ORBACH: I think the beauty of this calculation is that it shows that the Anderson model and the exchange model basically treat different things. The definition of the magnetization is different. In the exchange model you define a moment on the d site. You take into account the polarization correction of the conduction electrons by using the exchange parameter λ . The Anderson model

goes a step further by allowing delocalization of the d electrons through admixture, in particular you have admixture of the d electrons to the unoccupied conduction electron states above the Fermi surface. This always gives rise to a g shift, but not to relaxation because these are virtual transitions. So the electrons instantaneously follow the localized spin. The factor arises because the localized spins loose something to the conduction electrons at a rate proportional to g_d , but it is returned at a rate proportional to the g_e of the conduction electrons.

I would also like to add that this is not just a nice calculation in the sense that it shows that exchange is equal to the Anderson model in terms of *ESR*. More importantly it opens the way to do covalency corrections properly for resonance in dilute alloys. Most of us who have done covalency calculations in dilute alloys have used the exchange model and stuck admixture on to it, praying that we get it right. But this is a systematic way of taking into account admixture into the dynamics.

