# REMARKS ON Gd g-SHIFTS

by

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A. A. Gomes, A. P. Guimarães and L. Iannarella
*Centro Brasileiro de Pesquisas Físicas*
*Rio de Janeiro, Brazil*

## ABSTRACT

The s-d hybridization is given a simple formulation in order to discuss its relevance to the Gd g-shifts; it is apparent that the effective exchange parameters $J_{eff}$ can be negative even with negligible interband mixing.

A possible explanation for the signs of the g-shift and band magnetization in Gd metal is also suggested.

It has been suggested by Coles et al. (1970) that the negative $Gd^{3+}$

g-shifts observed in several metallic hosts as Pd, $LaRu_2$ and others (see the

review by Peter et al. (1967)) could be explained if one assumed that the s

and d bands had antiparallel magnetization, as it is the case in iron metal.

Within this hypothesis the exchange parameter $J^S$ is positive (i.e. the

Heisenberg exchange dominates the contribution from interband mixing)    and

also the d susceptibility is larger than the s susceptibility. The  g-shift

is negative since the s-d induced d magnetization (antiparallel to s)  domi-

nates the s magnetization (see below). Although Coles and co-workers     do

not actually say it explicitly,  it appears that they are taking $J^d = 0$, and

therefore the d band does not give any (direct) contribution to the g-shift.

We want to discuss the implications of s-d hybridization in the

analysis of gadolinium g-shifts. The general picture of the alloy  consists

of a localized and stable f-moment which interacts with the   band and   the

s band, these interactions being characterized by two distinct parameters $J^d$

and $J^S$. From Watson et al. (1965) we take that $J^d$ is positive, and $J^S$ =

$= J^S_H + J^S_I$ is a sum of the ordinary exchange and interband mixing    effects

(Anderson and  Clogston (1961)) and can in principle have either sign.  The

hybridization couples the s and d  magnetizations and tends to align    them

antiparallel; the final orientation of the s- and d-band magnetizations will

depend on the  magnitude of $U_{sd}$ and on the sign and magnitudes of $J^d$ and $J^S$.

We assume that the effect of the s-d interaction is linear; we write the  s

magnetization induced by the d- band, and the d magnetization induced    by

the s band as

$$\vec{m}^S(d) = -\alpha_{sd}\,\vec{m}^d$$

$$\vec{m}^d(s) = -\alpha_{ds}\,\vec{m}^s$$

The values of the parameters $\alpha$ will be discussed later on.

The total s- and d-band magnetizations are

$$\vec{m}^s = \frac{J^s \chi^s}{g_e \mu_B}\,<\vec{S}> - \alpha_{sd}\,\vec{m}^d$$

(1)

$$\vec{m}^d = \frac{J^d \chi^d}{g_e \mu_B}\,<\vec{S}> - \alpha_{ds}\,\vec{m}^s$$

where $<\vec{S}>$ is the spin of the rare-earth. The total band magnetization becomes

$$\vec{m} = \vec{m}^d + \vec{m}^s = \left\{ J^d \cdot \frac{1-\alpha_{sd}}{1-\alpha_{sd}\,\alpha_{ds}} \right\} \frac{\chi^d <\vec{S}>}{g_e \mu_B} + \left\{ J^s\,\frac{1-\alpha_{ds}}{1-\alpha_{sd}\,\alpha_{ds}} \right\} \frac{\chi^s <\vec{S}>}{g_e \mu_B}$$

(2)

Defining the two effective parameters

$$J^d_{eff} = J^d\,\frac{1-\alpha_{sd}}{1-\alpha_{sd}\,\alpha_{ds}} \qquad \text{and} \qquad J^s_{eff} = J^s\,\frac{1-\alpha_{ds}}{1-\alpha_{sd}\,\alpha_{ds}}$$

(3)

one has for the total g-shift:

$$\Delta g = J^s_{eff}\,\frac{\chi^s}{g_e \mu_B^2} + J^d_{eff}\,\frac{\chi^d}{g_e \mu_B^2}$$

(4)

An important consequence of the inclusion of hybridization is that the  ef-

fective exchange parameters $J_{eff}$ (Eq. 3) and the parameters $J$ computed in the usual way (see Watson et al. (1969)) may have opposite signs, depending on the values of the $\alpha$'s. One may even be able to explain the negative g-shifts without having to invoke large contributions from $J_I^s$; Coles et al. (1970) have actually argued that $J_I$ should be small in view of the large ($\sim 10$ eV) energy denominators.

To estimate the parameters $\alpha$ we could treat the s-d hybridization by the approximate Hamiltonian (e.g. Long and Turner (1970)):

$$\mathcal{H}_{sd} = U \vec{S}_s \cdot \vec{S}_d$$

Writing

$$- g_e \mu_B \vec{H}^{sd} \cdot \vec{S}^s = U \langle \vec{S}_d \rangle \cdot \vec{S}_s = U \frac{\langle \vec{m}_d \rangle}{\mu_B} \cdot \vec{S}_s \quad \text{and} \quad \vec{H}^{sd} = \frac{\langle \vec{m}^s(d) \rangle}{\chi^s}$$

we finally derive

$$\alpha_{sd} = \frac{U\chi^s}{g_e \mu_B^2} \quad \text{and} \quad \alpha_{ds} = \frac{U \chi^d}{g_e \mu_B^2}$$

Consequently $\alpha_{sd}/\alpha_{ds} = \chi^s/\chi^d$; since we expect $\chi^d \cong 10\chi^s$ and $\alpha_{sd} \lesssim 0.1$ we obtain $1-\alpha_{ds} < 0$. This would lead to $J^s_{eff}$ and $J^d_{eff}$ of opposite signs (from Eqs. (3)). We need an estimate of U before we can attribute with any certainty the signs of the effective parameters $J^s_{eff}$ and $J^d_{eff}$; the main point to be stressed is that $J^s_{eff}$ may be easily made negative, without the requirement of large interband mixing.

From equations (2) and (4) it is obvious that the total band magnetization and the g-shift cannot have opposite signs. There are, however, some results in the literature where there is a contradiction be-

tween the low temperature results and g-shift measurements. In gadolinium metal, for example, the band magnetization is positive and (assuming a free ion moment for $Gd^{3+}$) approximately equal to 0.5 $\mu_B$ (Nigh et al. (1963)) whereas $\Delta g \cong - 0.04$ (Peter et al. (1962), Chiba and Nakamura (1970)).

Since the magnetization results are obtained at low temperatures, and the g-shifts are measured in the paramagnetic region the experimental results could be reconciled if we assume that $\chi^d$ varies substantially with temperature, in such a way that the d-magnetization dominates at low temperatures, being smaller than the s-magnetization at high temperatures. We have then:

$$| J^d_{eff} \chi^d(T = 0\ ^oK)| > |J^s_{eff} \chi^s| > |J^d_{eff} \chi^d(T)|$$

and

$$J^s_{eff} < 0 .$$

Making some assumptions, namely taking $J^d_{eff} > 0$ and $J^s_{eff} < 0$ and taking, for instance, $J^d_{eff} N^d(\varepsilon_F) \cong 0.03$ at high temperatures, we find that $\chi^d$ should drop to 1/5 - 1/10 of its low temperature value. To study if this quite large drop in susceptibility was plausible, Gomes et al. (1972) suggest a model for the Gd band structure with a sharp peak near the Fermi level. The d susceptibility was computed by solving numerically the expression for $\chi(T)$ (e.g. Shimizu and Tanaka (1961)).

The susceptibilities were calculated in the temperature range 400 - - 600 $^oK$ and the trend was then extrapolated to T = ∞; in this way one could parallel the procedure used to obtain the g-shift without the perturbation due to demagnetization effects (e.g. Davidov and Shaltiel (1968)).

We have considered also an alternative possibility where bottleneck

mechanisms could inhibit the observation of the d-like contribution to the g-shift. However using estimates from Gossard et al. (1968) for the electron-lattice relaxation times and the localized spin dynamics (Giovannini 1967) it turns out that the bottleneck condition is not satisfied for the d-electrons, supporting thus our view that both s- and d- contributions appear in expression (4).

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