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g Tensors and Tensor Interactions—Their Effect on Conduction-Electron Spin Resonance*

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Conduction-electron g tensors depending on electron momentum can (through the cyclotron motion of the electron) cause transitions between spin Zeeman levels which broaden and shift the conduction-electron spin resonance. Tensor quasiparticle interactions are capable of similar effects. The relaxation time and exchange parameter controlling motional and exchange narrowing of the conduction-electron spin resonance are precisely defined.

Lubzens, Shanabarger, and Schultz[1] have recently reported detailed measurements of the frequency dependence, temperature dependence, and dependence on resistivity ratio of the g value and linewidth of conduction-electron spin resonance (CESR) in aluminum. The initial report[2] of CESR in aluminum led Dupree and Holland[3] to suggest that g anisotropy might be important, and theories of motional and exchange narrowing of g anisotropy have been formulated by de Botton[4] and Fredkin and Freedman.[5] The results of Lubzens, Shanabarger, and Schultz[1] fit very well the predictions of these latter theories.[4,5]

In the present work, the tensor nature of the g value is explicitly considered (for the first time) and such considerations lead to new processes affecting the linewidth and g shift of the CESR. In the light of this development, a reinterpretation of the results of Lubzens, Shanabarger, and Schultz[1] would be useful, as it is believed that these new results may strongly affect the value of the parameter B (defined below).

Also, the motional-narrowing relaxation rate is given in terms of a weighted average of the impurity scattering cross section,[6] the weighting factor being different from that determining the resistivity relaxation rate. Similarly, the exchange parameter B controlling exchange narrowing is precisely defined.

Finally, we follow up a remark of de Botton[4] that the influence of spin-orbit coupling on the quasiparticle interaction can produce effects qualitatively similar to those produced by g anisotropy. A new type of tensor quasiparticle interaction is introduced and is shown to give rise to contributions to the effective g tensor. Nothing is presently known about the magnitude of such tensor interactions, but it is known that the magnitude of the g anisotropy required to account for the aluminum results is surprisingly large.[1] A first-principles estimate of the magnitude of the tensor interaction would thus be of value.

We shall study in detail an isotropic electronic Fermi liquid. For a Zeeman Hamiltonian of the form $H_Z = -g(\vec{p})\mu_B\vec{s}\cdot\vec{H}$ to be invariant with respect to simultaneous rotations of $\vec{s}$, $\vec{H}$, and $\vec{p}$, we must have $g(\vec{p})$ independent of the direction $\vec{p}$. Anisotropy in g can only be introduced by the use of a g tensor, and we therefore assume a Zeeman Hamiltonian

$$H_Z = -\mu_B\vec{s}\cdot\overleftrightarrow{g}\cdot\vec{H}, \quad \overleftrightarrow{g} = g_0\overleftrightarrow{1} + \tfrac{1}{2}g_2\overleftrightarrow{T}, \tag{1}$$

$$\overleftrightarrow{T} = 3\vec{p}\vec{p} - \overleftrightarrow{1}. \tag{2}$$

The quantity $\overleftrightarrow{1}$ is the unit dyadic, and $\vec{p}$ denotes a unit vector in the direction of the electron momentum.

The tensor part of the Zeeman interaction can be written in the form $-g_2\mu_B\vec{s}\cdot\vec{H}_{eff}$, where $\vec{H}_{eff}$ is an effective magnetic field given by $\vec{H}_{eff} = \frac{1}{2}[3\vec{p}(\vec{p}\cdot\vec{H}) - \vec{H}]$. The effective field has a component in the direction of the momentum of the electron, as well as a component along the external field. Since the electron momentum precesses around the external field at the cyclotron frequency, ω_c, the effective magnetic field has a time-dependent component which can cause transitions between the spin energy levels. As a result of collisions of the electron with impurities or phonons, however, the cyclotron frequency is not sharply defined, and $\vec{H}_{\rm eff}$ possesses a spectrum of frequencies lying in the range $\omega_c - \tau^{-1} < \omega < \omega_c + \tau^{-1}$, τ^{-1} being the collision frequency. So long as the electron spin-resonance frequency lies within this range, $\vec{H}_{\rm eff}$ can limit the lifetime of the spin by causing transitions between its energy levels. The precessing component of $\vec{H}_{\rm eff}$ also gives rise to a g shift, as will be seen below. Thus the tensor nature of the g value gives rise to new effects which are not present in previous theories[4,5] assuming a scalar g value.

Following Silin,[7] and making use of the Zeeman interaction as defined by (1), the spin-dependent part of the quasiparticle interaction is written $\delta\epsilon_{\rm op}(\vec{p},\vec{r},\vec{t}) = \vec{\sigma}_{\rm op}\cdot\vec{\epsilon}_2(\vec{p},\vec{r},\vec{t})$, where the components of $\vec{\sigma}_{\rm op}$ are Pauli matrices,

$$\vec{\epsilon}_2(\vec{p},\vec{r},\vec{t}) = -\tfrac{1}{2}\mu_B \overleftrightarrow{g}\cdot\vec{H} + \sum_{\vec{p}'}\psi(\vec{p},\vec{p}')\vec{\sigma}(\vec{p}',\vec{r},\vec{t}), \quad (3)$$

and $\frac{1}{2}\vec{\sigma}(\vec{p},\vec{r},\vec{t})$ is the spin density in phase space at time t. As a result of the assumed isotropy, ψ depends only on the angle θ between $\vec{p}$ and $\vec{p}'$ and can be written

$$\rho\psi(\theta) = \sum_l (2l+1)B_l P_l(\cos\theta), \quad (4)$$

where ρ is the density of states.

Now let $\vec{\sigma} = \vec{\sigma}_0 + \delta\vec{\sigma}$, where $\frac{1}{2}\vec{\sigma}_0$ is the spin density induced by the static external field $\vec{H}_0$, and write

$$\delta\vec{\sigma} = -(\partial f_0/\partial\epsilon)\vec{G}(\vec{p},\vec{r},\vec{t}), \quad (5)$$

where $f_0 = 2[\exp(\beta\epsilon_p)+1]^{-1}$. We shall look for the homogenous normal modes only (i.e., assume that $\vec{G}$ is independent of $\vec{r}$). The components G_α ($\alpha = 0, \pm 1$) of $\vec{G}$ are defined by $G_0 = G_z$, and $G_{\pm 1} = (1/\sqrt{2})(G_x \pm iG_y)$; the external field $\vec{H}_0$ is assumed along the z axis. $G_\alpha(\vec{p})$ can be expanded in spherical harmonics, i.e.,

$$G_\alpha(\vec{p}) = \sum_{lm} G_{\alpha lm} Y_{lm}(\theta,\phi). \quad (6)$$

The equation of motion for $G_{\alpha lm}$ is [from Eq. (5′) of Silin]

$$\frac{dG_{\alpha lm}}{dt} = (i\alpha\bar{\omega}_0 - im\omega_c - 1/\tau_l)(1+B_l)G_{\alpha lm} + \int d\Omega\, Y_{lm}{}^*[\vec{\omega}_2\times\vec{G}']_\alpha, \quad (7)$$

where $\hbar\bar{\omega}_0 = -[g_0\mu_B H_0/(1+B_0)]$ (note $g_0 < 0$ for electrons), $\hbar\vec{\omega}_2 = -\frac{1}{2}[g_2/(1+B_2)]\mu_B\overleftrightarrow{T}\cdot\vec{H}_0$, and $\vec{G}' = \vec{G} + \delta\vec{\epsilon}_2$. Collisions of electrons with impurities are described by the τ_l's in (7) following Wilson and Fredkin,[8] the τ_l's being defined as in the electrical-resistivity problem (e.g., see Peierls[9]), i.e.,

$$\tau_l{}^{-1} = \int [1 - P_l(\cos\theta)]w(\theta)\,d\Omega, \quad l \geq 1. \quad (8)$$

The resistivity is controlled by τ_1.[9] $\tau_0{}^{-1}$ as defined by (8) is zero, but is assumed to have a small nonzero value arising from spin-orbit coupling.

The tensor g value couples the different components, G_α, of $\vec{G}$ in Eq. (7), and this makes the solution of (7) more complex than in cases previously considered.[4,5,7,8,10] Therefore we introduce a different method motivated by the approach of Wangsness and Bloch, and Redfield[11] to the general motional-narrowing problem. The starting point is a transformation to an interaction representation in which $\tilde{G}_r$ is defined by

$$G_r(t) = \tilde{G}_r(t)\exp(i\omega_r t), \qquad i\omega_r = (i\alpha\bar{\omega}_0 - im\omega_c - \tau_l{}^{-1})(1+B_l), \quad (9)$$

where $r = (\alpha, l, m)$. Matrices

$$A_{rr'}(t) = \exp(-i\omega_r t)A_{rr'}(0)\exp(i\omega_{r'}t) \quad (10)$$

are defined by the equation

$$\exp(-i\omega_r t)\int d\Omega\, Y_{lm}{}^*[\vec{\omega}_2\times\vec{G}']_\alpha = i\sum_{r'}A_{rr'}(t)\tilde{G}_{r'}(t). \quad (11)$$

Equation (7) now becomes, in this interaction representation,

$$d\tilde{G}_r(t)/dt = i\sum_{r'}A_{rr'}(t)\tilde{G}_{r'}(t). \quad (12)$$

We are interested in the frequency and decay rate of the CESR mode which, in the limit $\vec{\omega}_2 = 0$, is an oscillation of G_{-100}; we call this the $r=0$ mode [i.e., $(\alpha, l, m) = (-1,0,0)$ implies $r=0$]. Therefore, Eq. (12) is solved by iteration to second order in $A_{rr'}$ subject to the initial condition that, at $t=0$, $\tilde{G}_r = \tilde{G}_0(0)\delta_{r,0}$. This yields the expression

$$\omega_R - \frac{i}{T_2} = \omega_0 - i\frac{(1+B_0)}{\tau_0} + \sum_{r\neq 0}\frac{A_{0r}(0)A_{r0}(0)}{\omega_r - \omega_0} \quad (13)$$

for the frequency, ω_R, and transverse relaxation time, T_2, of the CESR. This solution is valid when $|A_{r0}| \ll |\omega_r - \omega_0|$, where $r \neq 0$; this inequality implies, for example, that our results have no significance in the limit $B_l \to 0$ first, and $\tau_l \to \infty$ later. The result (13) is valid, however, for cases[1] of current interest experimentally. The values of r contributing to (13) are $r=(0,2,1)$ and $r=(-1,2,0)$ and the explicit evaluation of (13) leads to the results

$$\frac{\omega_R-\omega_0}{\omega_0}=\frac{(1+B_2)B\langle\omega_{2z}{}^2\rangle\tau_2{}'^2}{1+(B\bar{\omega}_0\tau_2{}')^2}+\frac{(1+B_2)(\omega_0-\omega_c{}')\frac{1}{2}[\langle\omega_{2x}{}^2\rangle+\langle\omega_{2y}{}^2\rangle]\tau_2{}'^2}{\bar{\omega}_0[1+(\omega_0-\omega_c{}')^2\tau_2{}'^2]}, \tag{14}$$

$$\frac{1}{T_2}=\frac{1}{\tau_0{}'}+\frac{(1+B_0)(1+B_2)\langle\omega_{2z}{}^2\rangle\tau_2{}'}{1+(B\bar{\omega}_0\tau_2{}')^2}+\frac{(1+B_0)(1+B_2)\frac{1}{2}[\langle\omega_{2x}{}^2\rangle+\langle\omega_{2y}{}^2\rangle]\tau_2{}'}{1+(\omega_0-\omega_c{}')^2\tau_2{}'^2}. \tag{15}$$

The notation $\langle\cdots\rangle=(4\pi)^{-1}\int(\cdots)d\Omega$ giving

$$\langle\omega_{2z}{}^2\rangle=\tfrac{4}{3}\langle\omega_{2x}{}^2\rangle=\tfrac{4}{3}\langle\omega_{2y}{}^2\rangle=\tfrac{1}{5}[g_2\mu_BH/(1+B_2)\hbar]^2.$$

Also $\tau_0{}'=\tau_0/(1+B_0)$, $\tau_2{}'=\tau_2/(1+B_2)$, $\omega_c{}'=\omega_c(1+B_2)$, and

$$B=B_0-B_2=(4\pi)^{-1}\rho\int[1-P_2(\cos\theta)]\psi(\theta)d\Omega. \tag{16}$$

A Zeeman Hamiltonian of the form

$$H_Z=-g(p)\mu_B\vec{s}\cdot\vec{H}\equiv-[g_0+\tfrac{1}{2}g_2T_{zz}(p)]\mu_B\vec{s}\cdot\vec{H}$$

would give the terms in (14) and (15) linear in $\langle\omega_{2z}{}^2\rangle$; these are thus formally analogous to results obtained previously.[4,5] The terms linear in $\frac{1}{2}[\langle\omega_{2x}{}^2\rangle+\langle\omega_{2y}{}^2\rangle]$ give the contribution of the new lifetime effect. Note that the latter terms can be obtained from the former by replacing B with $[(\omega_0-\omega_c{}')/\bar{\omega}_0]$, and $\langle\omega_{2z}{}^2\rangle$ with $\frac{1}{2}[\langle\omega_{2x}{}^2\rangle+\langle\omega_{2y}{}^2\rangle]$.

The value of B for Al obtained by Lubzens, Shanabarger, and Schultz[1] is unreliable as they were not aware of the possible influence of a g tensor in producing the lifetime effect described here.

In the case of a nonspherical Fermi surface, there will be a distribution of cyclotron frequencies depending on the external field direction; this will cause the g shift and linewidth to be anisotropic (anisotropy is observed in Cu and Ag[1]).

The experiments of Lubzens, Shanabarger, and Schultz[1] show that the motional-narrowing relaxation rate differs from the resistivity relaxation rate. Both rates are weighted averages of the impurity scattering cross section $w(\theta)$ [see Eq. (8)], but the motional-narrowing rate $\tau_2{}^{-1}$ uses a weight factor $[1-P_2(\cos\theta)]$ which weights most heavily collisions which significantly change the g value, whereas the resistivity rate $\tau_1{}^{-1}$ uses the weight factor $[1-P_1(\cos\theta)]$ which favors backward scattering. These weight factors are compared in Fig. 1. The weight factor $[1-P_2(\cos\theta)]$ also determines the exchange parameter B responsible for exchange narrowing; this is consistent with the fact that exchange between electrons with equal g values is unimportant in the exchange-narrowing process. The recent progress in determining impurity scattering cross sections[6] means that one might soon be able to calculate τ_2 from Eq. (8) for specific impurities, and this would be useful in interpreting CESR experiments.

The preceding discussion assumes an interaction between quasiparticles with momenta $\vec{p}$ and $\vec{p}'$ of the form $\psi(\vec{p},\vec{p}')\vec{\sigma}\cdot\vec{\sigma}'$ [see Eq. (3)]. For our isotropic model, however, interactions between quasiparticles of the form

$$\rho^{-1}D\vec{\sigma}\cdot[\overleftrightarrow{T}(\vec{p})+\overleftrightarrow{T}(\vec{p}')]\cdot\vec{\sigma}' \tag{17}$$

are allowed by symmetry (D is a dimensionless parameter which characterizes the strength of this interaction, which se call the tensor interaction). Following Silin[7] [but including the tensor interaction (17)] the energy of a quasiparticle in an external field $\vec{H}_0$ is written $\epsilon=\vec{\sigma}\cdot\vec{\epsilon}_{20}$ where an integral equation [Silin's Eq. (6)] is found to determine $\vec{\epsilon}_{20}$. The solution of this equation is

$$\vec{\epsilon}_{20}=-\tfrac{1}{2}\bar{g}_0{}'\mu_B\vec{H}_0-\tfrac{1}{4}\bar{g}_2{}'\mu_B\overleftrightarrow{T}\cdot\vec{H}_0, \tag{18}$$

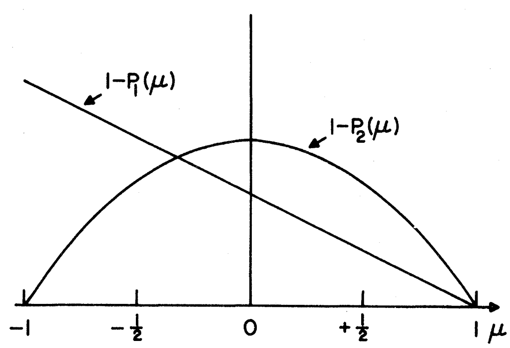

FIG. 1. Comparison of the weight factors $[1-P_1(\mu)]$ and $[1-P_2(\mu)]$ used to define the resistivity and motional-narrowing relaxation rates; $\mu=\cos\theta$ and θ is the scattering angle.

where

$$\bar{g}_0{}' = \frac{g_0}{1+B_0}\left[1 - \frac{Dg_2}{g_0(1+B_0)} + \frac{2D^2}{(1+B_0)(1+B_2)}\right],$$

$$\bar{g}_2{}' = \frac{g_2 - 2D\bar{g}_0{}'}{1+B_2},$$

correct to second order in D and g.

Since tensor quasiparticle interactions contribute to $\bar{g}_2{}'$ in (19), they can be expected to produce effects similar to those produced by a nonzero g_2 in Eq. (1). A complete analysis of these effects bears out these expectations.

Since the g tensors and tensor interactions considered in this paper limit the lifetime of a spin, a contribution to T_1^{-1} similar to the final term in (15) will result. No such contribution to T_1^{-1} will result in the case of the scalar g considered in Refs. 4 and 5. A measurement of T_1 would thus distinguish between models employing a scalar g and those employing a tensor g.

In summary, the above results [Eqs. (14) and (15)] provide a basis for a qualitative interpretation of motionally and exchange narrowed CESR similar to that observed in Cu, Ag, and Al.[1] To make the interpretation quantitatively accurate, however, the true Fermi-surface geometry must be considered (because of its effect on the cyclotron frequency, for example); this would be of particular interest in the cases of Cu and Ag which display anisotropic CESR.

Finally, we note that the question "which has the dominant effect on CESR, the momentum-dependent g tensor, or tensor quasiparticle interaction?" has yet to be answered.

I would like to thank S. Schultz for suggesting to me that a solution to the problem of the correct definition of the motional-narrowing relaxation time, and its difference from the resistivity relaxation time, would be useful; this provided my initial interest in this problem. I would also like to thank Mme. Lewiner (née de Botton) for sending me a copy of her thesis.[4]

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