

Theories of the static and dynamic Kondo susceptibility

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III. THEORY

THEORIES OF THE STATIC AND DYNAMIC KONDO SUSCEPTIBILITY

BY

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Let me remind you first of the *elementary results* concerning the impurity susceptibility of magnetic ions like *Mn*, *Fe* alloyed into normal metals like copper or gold. I will consider the one impurity problem only. The low energy dynamics of such ions is described by the spin variables S_x , S_y , S_z formed by the spins of the unfilled electron shells. The problem is: how does the spin move in the metal? To ask the question precisely we imagine the most direct probe of our magnetic impurity: the change of the impurity spin polarization $\delta \langle S_z \rangle$ if we change the magnitude of the external magnetic field by δB . In linear approximation $\delta \langle S_z \rangle = \chi(\omega) \delta B$, where the constant of proportionality depends on the frequency ω we suppose our field to vary with as a function of time. $\chi(\omega)$, the longitudinal dynamical impurity susceptibility, is the subject of the following discussion. $\chi(\omega)$ determines for instance *NMR* and Mössbauer relaxation spectra, the cross relaxation rate for *EPR*, the spin-flip cross section for neutron scattering in dilute magnetic alloys. The imaginary part of the susceptibility, $\chi''(\omega)$, is called the absorptive part of $\chi(\omega)$, since the energy the spin absorbs from the external field is given by $\omega \chi''(\omega) (\delta B)^2$. $\chi''(\omega)$ gives the excitation spectrum of the impurity: if $\chi''(\omega)$ is large states with energy ω can be excited easily, if $\chi''(\omega)$ is small excitations with energy ω cannot be made. $\chi''(\omega)$ determines $\chi(\omega)$ by a Kramers-Kronig-relation; in particular, the static susceptibility is given by

$$\chi^0 = \chi(\omega=0) = \frac{1}{\pi} \int d\omega \frac{\chi''(\omega)}{\omega}. \quad (1)$$

χ^0 is a thermodynamical quantity, namely the change of the spin polarization with changing field

$$\chi^0 = d\langle S_z \rangle / dB. \quad (2)$$

The interaction of the impurity with the metal we assume to be given by the trivial coupling of the impurity spin with the conduction electron spin density at the impurity site: $H_{int} = J \vec{S} \vec{s}(r=0)$. This interaction model is oversimplified for most examples, but before one has not understood the properties of this model completely, it does not seem reasonable to try more sophisticated hamiltonians. The exchange coupling constant J times the conduction electron density of states ρ is of the order $\rho J \sim 0.1$. The coupling is antiferromagnetic for the transition metals, it may be ferromagnetic ($J < 0$) or antiferromagnetic ($J > 0$) for rare earth ions depending on the specific alloy.

The ground state of a free magnetic ions is degenerate. It does not cost energy to polarize the system and hence the zero field zero temperature susceptibility χ^0 is infinite. At nonzero temperature T , χ^0 decreases according to a Curie law $\chi^0 = C/4T$. The conduction electrons get polarized in the external field too and enhance or shield off the effective field acting on the impurity. This yields to an increase or decrease of the Curie constant C depending on the sign of J . In leading order one finds for the g -shift (in appropriate units)

$$\chi^0 = \frac{1}{4T} (1 - \rho J + (\rho J)^2 K_1); \quad B \ll T. \quad (3)$$

In an external field B one has to polarize against the Zeeman energy and therefore $d\chi^0/dB < 0$. The main effect of the interaction is to make the impurity spin a non-conserved quantity. Spinpolarizations deviating from the equilibrium value die out within a characteristic time interval T_1 , the longitudinal spin relaxation time. These changes of $\langle S_z \rangle$ are due to spin flips accompanied by spin density wave emission. In leading order one finds for the relaxation rate the Korringa formula

$$1/T_1 = \pi (\rho J/2)^2 (1 + (\rho J) K_2) / \chi^0; \quad B \ll T. \quad (4)$$

Since χ^0 diverges for vanishing temperature the relaxation rate slows down to zero if $T \rightarrow 0$. For increasing field one finds an increasing relaxation rate.

The range of validity of leading order calculations like the ones yielding (3), (4), has to be determined by calculating the correction terms K_1, K_2 ; a task first carried out 1964 in other connections by Kondo [1]. Yosida and Okiji [2] obtained for the next order susceptibility correction in equ. (3) $K_1 = \log T/D$, where D is a band width parameter of the order of 10^4 deg. Hence the Curie constant is reduced with decreasing temperature and the leading order formula is valid for $\log T \gg \log T_K$, where $T_K = D \exp(-1/\rho J)$ denotes the Kondo temperature. T_K depends sensitively on the exchange coupling constant and can be any value on the accessible temperature scale. Similarly, Walker, Orbach and Spencer [3] have obtained the logarithmic correction factor $K_2 = -2 \log(T/D)$ for the relaxation rate. For ferromagnetic

coupling $1/T_1$ is smaller than the Korringa value, for antiferromagnetic coupling it is larger. Due to the logarithmic divergent K_1, K_2 formulae (3), (4) lose any meaning for vanishing temperature; χ^0 as well as $1/T_1$ may become negative then. The reason for the Kondo divergence appearing e.g. in K_2 can be explained as follows. The leading contribution to $1/T_1$ is due to spin flip accompanied by the emission of one spin density wave, the correction K_2 is due to the spin flip processes accompanied by the emission of two spin density waves. Since the spin wave energy approaches zero for long wave lengths the phase space for the second process is larger than the one for the first process, and so the second process dominates provided the relevant energies are small enough. So Kondo predicted anomalies to occur for T approaching T_K .

Progress in eliminating unphysical divergencies like K_1, K_2 has been made within approximation schemes proposed by Nagaoka and Suhl [4]. Zittartz [5] has shown the equivalence of both approximations and so we discuss the second one only. Suhl considers the scattering of a conduction electron by the impurity and makes two closely connected assumptions. First he assumes the impurity in the metal to have a degenerate groundstate which then can be described in the language of spin dynamics. There are many scattering channels: electron-impurity scattering with only momentum changes occurring, electron-impurity scattering with spin flip involved, scattering processes with additional creation of one, two, etc. spin density waves. The first process is described by the spin non flip scattering amplitude t , the second one by the spin flip amplitude τ . τ is the heart of Suhl's theory and the possibility of its definition depends on the validity of the first assumption mentioned. The second assumption consists of considering all processes with spin wave excitations involved as negligible. Then one could derive closed nonlinear dispersion equations connecting t and τ which are solvable and yield the scattering amplitudes as function of energy ω and temperature T . Further approximations are necessary to connect the susceptibility with the scattering amplitudes t and τ . Brenig, Wölfle and I [6] have found for the relaxation rate essentially the Korringa formula (4), except that the Born approximation ρJ for the spin flip scattering amplitude has to be replaced by the correct one at zero frequency

$$1/T_1 = \pi T |\tau(\omega = 0)|^2. \quad (5)$$

The important consequence of this formula is, that $1/T_1$ can be very much larger than the Korringa expression (4). In particular for temperatures T close to T_K , τ becomes of order one [7] and hence the relaxation rate is as large as the Kondo temperature $1/T_1 \sim T_K \gg (1/T_1)_{\text{Korringa}}$. The static susceptibility has also been calculated by Brenig *et al.* and Ting [8] and for low temperatures for impurity spin 1/2 it was found to be

$$\chi^0 \propto 1 / (T \log(T_K/T)) ; \quad T \ll T_K. \quad (6)$$

χ^0 approaches infinity for vanishing temperature, but the Curie divergence is weakened slightly by logarithmic corrections. Some doubts have been expressed concerning our derivations of results (5), (6), but attempts to find improvements [9] have not been successful. Because of the spin relaxation the impurity in the metal is an unstable particle as far as the spin degree of freedom is concerned. Asymptotic scattering states of the impurity with definitive spin cannot be defined strictly. Approximately, the first assumption of the Suhl theory is valid only if the electron impurity collision time t_c is smaller than the spin life time $1/T_1$. The collision time t_c is given by the cross section $\text{Im } t(\omega)$; the latter quantity exhibits a resonance of width T_K [10] and hence $t_c \sim 1/T_K$ for $T \sim T_K$. Thus the Suhl theory becomes inconsistent for temperatures approaching the Kondo temperature. For $T > T_K$ the results give a reasonable qualitative description of magnetic impurities; for $T \rightarrow 0$ the theory lacks any justification.

Because of impurity interaction effects it is difficult to perform *experiments*. Good data for χ^0 have been obtained for *Cu:Fe* by Tholence and Tournier and by Steiner *et al.* and for *Cu:Mn* by Wheatley *et al.* [11]. Furthermore there are computer experiments by Schotte and Schotte [12] done for the *s-d*-model. For the temperature range covered these experiments yield

$$\chi^0 \propto 1/(T + \theta) \quad (7)$$

with a θ of the order of T_K . There are no experiments known, which are in contradiction to such a Curie-Weiss law. Hence the zero temperature susceptibility is finite, there is no degenerate impurity ground state. If we are willing to acknowledge experiments at all, we have to conclude that the Suhl-Nagaoka theory is qualitatively wrong for low enough temperatures. The first task of the theory is to explain the finite zero temperature impurity susceptibility.

Post Suhl attempts to find the Kondo susceptibility have first been made by Anderson and Yuval [13]. They developed a scaling theory for the free energy which—besides many other things—allows them to conclude, that χ^0 should be finite varying like $C_1 - C_2 T^2$ for low temperatures and approaching formula (7) for high temperatures. Some other versions to motivate the Anderson-Yuval results have been tried [14], but till now it was not possible to derive quantitative results connecting e.g. C_1 , C_2 , θ . Wilson [15] has used his renormalization group technique, which was so successful in explaining critical phenomena, to attack the Kondo hamiltonian. Successively eliminating lower and lower electron excitation energies by means of a computer he has derived the zero temperature susceptibility as a function of the exchange coupling $1/\chi_0 \sim 10 \sqrt{\rho J} T_K$. He also obtained the asymptotic limit of the specific heat (C_K/T) ($T = 0$). Presumably he will be able to calculate the thermodynamical functions in a parameter regime interesting for the experimentalist and in principle he should also get the dynamics thus providing us an exact solution of the Kondo hamiltonian. At the moment it cannot be judged,

how much his work will contribute to our physical understanding of the Kondo problem. Recently P. Schlottmann and I have proposed [16] an approximate treatment of the Kondo susceptibility yielding χ^0 , T_1 and $\langle S_z \rangle$ as functions of field, temperature and exchange coupling. I am now going to spell out our approach in some detail.

Let us remember first that besides the Kramers-Kronig relation (1) there is the fluctuation-dissipation theorem connecting the absorption spectrum with the fluctuation of the spin. For spin 1/2 impurities this theorem reads

$$1/\chi^0 = 4T + \frac{2}{\pi} \int d\omega \left\{ \omega \left[\coth\left(\frac{\omega}{2T}\right) - \frac{2T}{\omega} \right] \right\} \left\{ \frac{\chi''(\omega)}{\omega \chi^0} \right\}, \quad B = 0. \quad (8)$$

Second, we notice that we expect $\chi(\omega)$ to have a resonant structure and hence it is more useful to discuss the parameters characterizing the resonance rather than $\chi(\omega)$ itself. This is done by introducing a function $N(\omega)$, such that

$$\chi(\omega)/\chi^0 = \frac{N(\omega)/\chi^0}{\omega + N(\omega)/\chi^0}. \quad (9)$$

The absorptive part of $N(\omega)$, $N''(\omega)$, is called the *relaxation spectrum* or noise spectrum. In lowest order one gets the Korringa formula $N''_k(\omega) = \pi(\rho J/2)^2$. There is no structure in the relaxation spectrum, and such trivial $N_k(\omega) = i\pi(\rho J/2)^2$ yields the correct Lorentian resonance for $\chi(\omega)$ in (9) corresponding to the first Bloch equation. Obviously, the properties of $N(\omega)$ are simpler than those of $\chi(\omega)$ and hence we propose to consider $N''(\omega)$ as the primary quantity approximations should be tried for. Having made an approximation for the relaxation spectrum the exact relation (8) yields a transcendental equation of the form $1/\chi^0 = F(1/\chi^0)$ to determine the static susceptibility χ^0 . For the Korringa approximation N_k equ. (8) yields for vanishing temperature a finite though exotic large value χ^0 independent of the sign of J . Hence one concludes: χ^0 can approach infinity only if the relaxation spectrum at zero temperature is smaller than the Korringa value, and χ^0 can be finite of reasonable size only if the relaxation spectrum is much larger than the approximation $N''_k(\omega)$.

To find a reasonable *first approximation* for $N''(\omega)$ we follow Anderson and Yuval and split the total hamiltonian into two parts

$$H = \underbrace{H_{el} + J'' S_z s_z}_{H''} + \underbrace{J^\perp (S^+ s^- + S^- s^+)}_{H^\perp} \quad (10)$$

The first part H'' contains the conduction electron energy and the longitudinal coupling. Here the impurity spin is not a dynamical variable. The eigenstates are scattering waves in the potential $\pm \rho J$ depending on the value of S_z . The ground state of H'' is degenerate and is given by the physical spin states: spin up with its

Friedel polarization cloud and spin down with its polarization cloud. The second part H^\perp induces transitions between the physical spin states thus being responsible for the nontrivial dynamics. In leading order with respect to H^\perp the relaxation spectrum is given by $(\rho J^\perp)^2$ times the overlap of the Slater determinant representing physical spin up states with energy ω and spin down states with energy zero. These overlaps have been found by DeDominicis and Nozières [17] in connection with the X-ray threshold problem to be $(D^2/\omega^2)^{\rho J''}$. Actually, this nonanalytic $\rho J''$ -dependence is the same—and has the same physical origin—as that known from [18] the infra-red divergence problem in quantum electrodynamics. For non zero temperature we have used results from Schotte [19] to obtain a smearing out of the singularity

$$N''(\omega) \sim \pi (\rho J^\perp / 2)^2 [D^2 / (\omega^2 + (2\pi T)^2)]^{\rho J''}. \quad (11)$$

Now it is crucial to notice the basic difference in the behaviour of for positive and negative sign of J'' . For antiferromagnetic coupling $N''(\omega)$ is much larger than $N_k''(\omega)$ and approaches infinity for $\omega, T \rightarrow 0$. For ferromagnetic coupling $N''(\omega)$ is lower than $N_k''(\omega)$ and tends to zero for vanishing ω and T . This nontrivial dependence on J of the overlap of Slater determinants formed with scattering states is a basic fact which cannot be reduced to simpler physical arguments. Result (11)

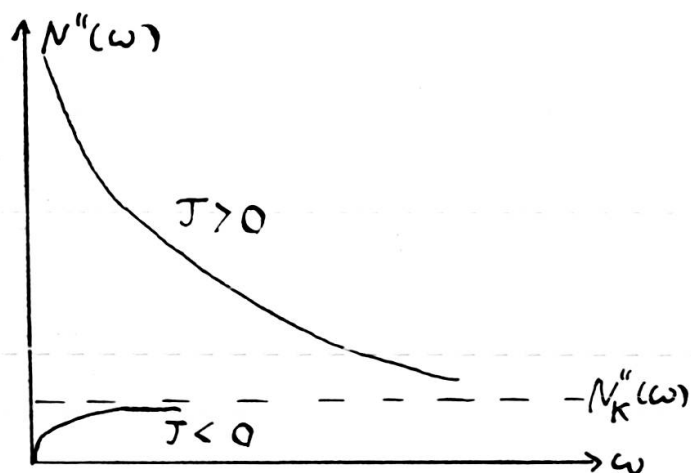


FIG. 1.

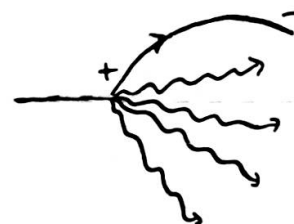


FIG. 2.

can also be interpreted as follows [18]: a spin deviation from equilibrium relaxes by spin flip accompanied by the emission of one, two, three, etc. spin density waves. No process with only a finite number of spin waves involved is important. Only the total sum gives a convergent result. It has to be admitted, that we know $N''(\omega)$ only asymptotically for $\omega, T \rightarrow 0$. We will need it later for all frequencies and use expression (11) throughout (long time approximation). It is not known at present whether or not this approximation implies more than just some numerical errors.

Substituting (11) into (9) and (8) we get an equation for χ^0 and hence for $\chi(\omega)$. This *first approximation* for χ^0 can be discussed in the complete J'' vs J^\perp plane. Neglecting band edge effects the equation for χ^0 at zero temperature can even be solved in terms of known functions. We found an area bounded by a parabola

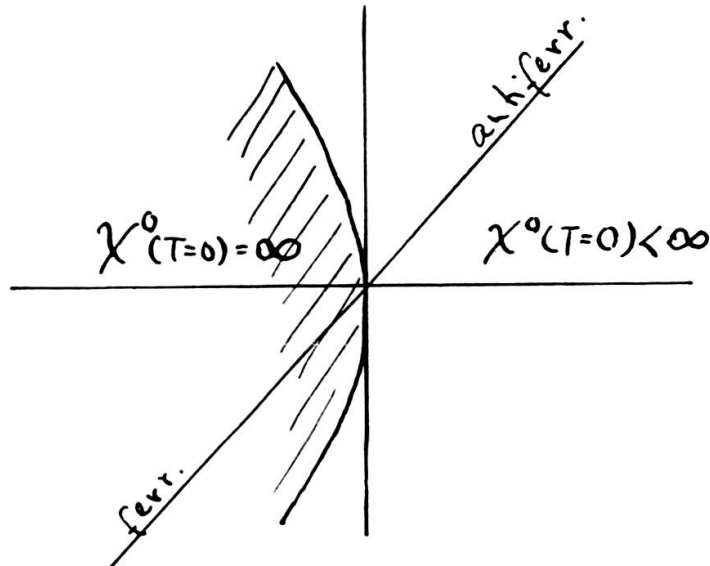


FIG. 3.

containing the case of ferromagnetic isotropic coupling in which the zero temperature susceptibility is infinite. In the complementary area containing the case of isotropic antiferromagnetic coupling χ^0 is finite. To test the theory we compare our results with the computer data by the Schottes [12] for three representative coupling constants, figure 4. Notice that we do not have any parameter to fit. The agreement of our curves with the data is an annoyingly good one. Notice the minimum of our curves; this nonsense we attribute to the divergence of approximation (11). Our treatment is flexible enough to allow an improved *second approximation*. The diver-

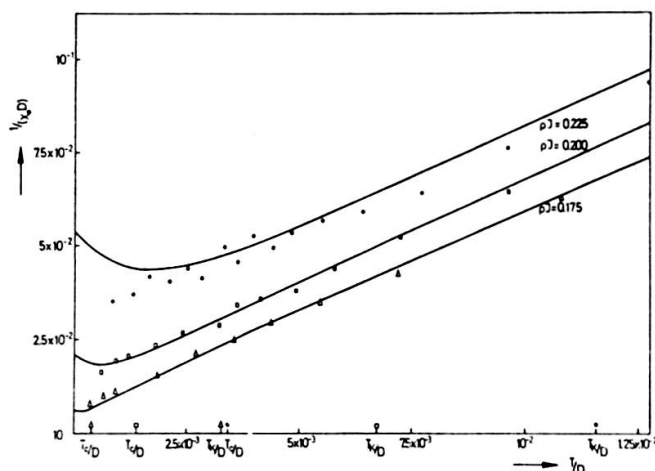


FIG. 4.

gence of $N''(\omega)$ in the antiferromagnetic case has to be doubted a priori, at least it is not consistent with the arguments used to introduce the concept of a relaxation spectrum. In the leading order calculation, figure 2, the spin-flip propagator has been replaced by the one of the noninteracting system showing a infinitely sharp resonance. It seems more reasonable to replace this propagator by one having a line width which we calculate then in leading second order in H^\perp . Another argument in favour of this approximation is the following. Approximately the relaxation spectrum $N''(\omega)$ is the same as the spin wave absorption cross section $\tau(\omega)$. This is very plausible since it is the spin wave absorption which makes the spin to relax. According to equ. (11) we have resonance absorption and our first approximation violates the unitarity bound for $\tau(\omega)$. To repair this error we remember the theory of resonance fluorescens in atomic physics. The same hamiltonian H^\perp coupling the spin wave into the impurity also yields a natural linewidth of the impurity excitations. Taking this into account by introducing a second order self energy for the transverse spin propagator, we find a $\tau(\omega)$ and hence a $N''(\omega)$ being finite for all ω, T . In figure 5 the corresponding result for χ^0 is compared with the curve for the first approximation ($\rho J = 0.200$). No drastic changes have been made for intermediate or high temperatures, but at low temperatures the fictitious minimum has disappeared almost completely.

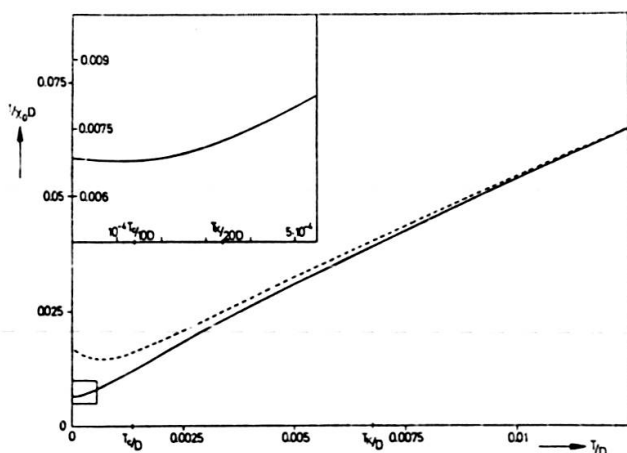


FIG. 5.

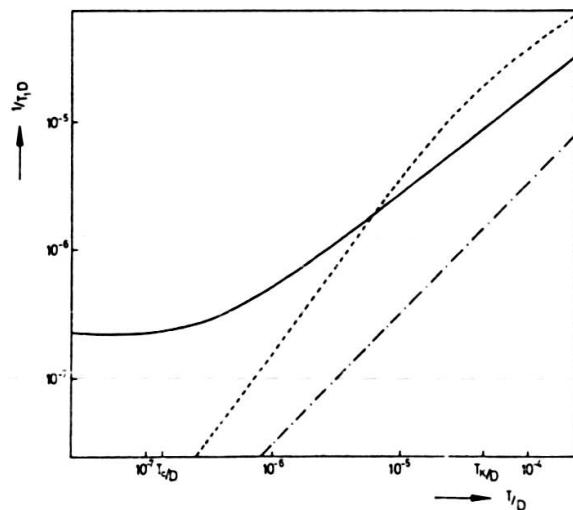


FIG. 6.

The results for $B = 0$ we obtained this way are: a Curie-Weiss like susceptibility $1/\chi^0 = 4.6(T + \theta)$ for $T > \theta$, a flattening of $1/\chi_0$ vs T for lower temperatures and a finite zero temperature $1/\chi_0 = 3.8 \theta$. $\theta = 0.55 D (\rho J/2)^{1/2} \rho J$ depends sensitively on the exchange coupling. The scaling of θ with J has to be considered with some scepticism, but figure 4 is at least an argument in favour of this result. Our $1/\chi_0$ vs T curve has a finite negative slope at zero temperature; however, an accuracy of 0.2%

would be necessary to detect this contradiction to the Anderson-Yuval result. Certainly our errors are larger than 0.2%. Hence, attributing fair but small error bars to our formulae the results are in agreement with all we know today about χ^0 . The relaxation rate $1/T_1$ decreases with decreasing temperature, but for temperatures of order θ it approaches the constant value $1/T_1 (T = 0) \sim 1.5 \theta$ in disagreement with the Korringa formula (3) or the Suhl theory result (5). To make a quantitative statement we have used the susceptibility data for *Cu:Mn* [11] to find $\rho J = 0.100$, $D = 10^4 \text{ deg} = 8.32 \cdot 10^{14} \text{ sec}^{-1}$ for this alloy. In figure 6 you see the relaxation rate as function of temperature. For reason of comparison we have also plotted formula (3) (dashed dotted) and (5) (dotted).

To obtain *results for* $B \neq 0$ only one more trick has to be invented. The left hand side of formula (8) changes to $(1 - 4 \langle S_z \rangle^2) / \chi^0$; the spin polarization enters the theory as a new thermodynamic quantity. To determine $\langle S_z \rangle$ we use equ. (1) as a differential equation to be solved simultaneously with equ. (8). As an representative example of the outcoming polarization curves for fixed temperature as function of field are shown in figure 7 (the dashed dotted curve is the polarization for a free

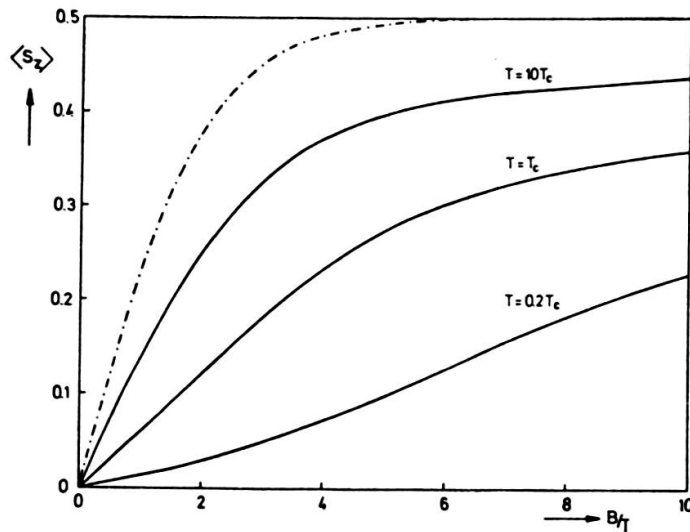


FIG. 7.

spin 1/2). The spin polarization is much smaller than the one for a free spin. Zeeman energies much higher than T are necessary to get saturation. For low enough temperatures $d\chi^0 / dB$ can be positive. The relaxation rate decreases with increasing field, since the anomalous large $1/T_1$ gets quenched.

A *summary* of the results of the approximation by Schlottmann and myself can be given as follows:

| ferrom. coupling ($J < 0$) | Antiferrom. coupling ($J < 0$) |
|--|--|
| destructive interference of spin polarization clouds | constructive interference of spin polarization clouds |
| depression of the relaxation spectrum $N''(\omega)$ | enhancement of the relaxation spectrum $N''(\omega)$ |
| degenerate ground state with $\delta(\omega)$ —excitation spectrum for $T \rightarrow 0$ | nondegenerate ground state with continuous excitation spectrum for $T \rightarrow 0$ |
| $1/\chi^0(T \rightarrow 0) = 0$ | $1/\chi^0(T \rightarrow 0) \sim 4\Theta > 0$ |
| $1/T^1(T \rightarrow 0) = 0$ | $1/T^1(T \rightarrow 0) \sim \Theta > 0$ |
| $B \neq 0$ yields spin orientation | $B \neq 0$ yields breaking of the impurity complex |

In other words, a magnetic impurity with ferromagnetic exchange coupling behaves qualitatively as understood in terms of the elementary results given at the beginning of this talk. An unrealistic experimental accuracy would be necessary to detect the nontrivial many body effects. An impurity with antiferromagnetic coupling, on the other hand, gradually transforms into a nonmagnetic complex if one decreases the temperature; its low temperature properties are very different from the ones of a quasi free spin.

I think that the Kondo problem should not be ignored if one is discussing dilute magnetic alloys and I hope that our results concerning the spin dynamics are non-trivial enough to challenge experimental verification.

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DISCUSSION

STEINER: Will the results change for a spin other than $\frac{1}{2}$?

GÖTZE: I would say not drastically.

STEWART: At high temperatures, does your theory agree with perturbation theory ?

GÖTZE: Sure.

