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The magnetic susceptibility of heavy-fermion compounds including spin–orbit and crystal field effects

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Abstract. In this paper we consider a more realistic approach to the periodic Anderson model by including spin–orbit and crystal field effects using the slave-boson technique. Looking firstly at the degenerate case with appropriate spin orbit coupled bands, we show that while for $T = 0$ we obtain the same results as the $1/N$ approach there are differences when we extend to finite T or when we go beyond the mean-field theory. We consider also crystal field splitting within the $1/N$ approach and show that this gives an anisotropic susceptibility in qualitative agreement with experiment. The inclusion of spin–orbit coupling again does not affect the results at $T = 0$.

1. Introduction

The experimental properties of the normal state of the mixed valent and heavy-fermion compounds depend in general on two features. Firstly there are the universal properties arising from a Kondo-like effect. We observe a crossover from an enhanced Pauli-like susceptibility near $T = 0$ to a Curie–Weiss-like susceptibility at high temperatures. The linear coefficient of specific heat at $T = 0$, γ, is enhanced by approximately the same factor as the susceptibility, although the Wilson ratio appears to be less than one (Stewart 1984). The resistivity varies as AT^2 close to $T = 0$, where $A \sim \gamma^2$, goes through a maximum at temperatures somewhat lower than the Kondo temperature, T_K, and decreases as $\log T$ at high temperatures. Other features are non-universal and are due to the crystal field splitting which is important in many of the compounds. The susceptibility only becomes truly Curie–Weiss-like for relatively higher temperatures which are determined by the crystal field splittings and the resistivity may have extra structure with a second peak at a temperature roughly corresponding to the crystal field splitting. The magnetic susceptibility and the transport properties are strongly anisotropic in the non-cubic Ce compounds (Bhattacharjee *et al* 1989, Gignoux *et al* 1988, Jaccard *et al* 1987, 1988 and Ōnuki *et al* 1984).

The periodic Anderson model has been widely used as a starting point for theories of the heavy-fermion compounds. The slave-boson method has proved a useful and systematic approach for dealing with the problems of strong interactions in this model (see for example Coleman 1987, Millis and Lee 1987). The effects of spin–orbit coupling

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and crystal field effects have largely been ignored in previous theories where it has been assumed for simplicity that the hybridization matrix element, V, is independent of spin and wavevector and that both the f and conduction electrons have 'spin-N' (we shall refer to this as the '$1/N$' model). The main aim of this paper is to consider a more realistic approach to this Hamiltonian by including these effects. It has already been suggested by Zou and Anderson (1986) that spin–orbit coupling is an important effect. Their principal result was that the effective magnetic moment is strongly reduced due to spin–orbit effects thus explaining why the Wilson ratio, calculated using the full magnetic moment, is less than one. This was subsequently refuted by several people and the situation remains unresolved (Zhang and Lee 1987, Aeppli and Varma 1987 and Cox 1987). Nonetheless, it is clear that the inclusion of spin–orbit coupling gives a qualitatively different physical picture. Only one specific linear combination of the six local f states can hybridize with a conduction state of given spin and momentum. The remaining four f states are unhybridized. This can be compared with the $1/N$ model where we have two six-fold degenerate bands. It cannot be assumed that the results from the two models will be equivalent. The fluctuations around the mean-field solution will also be affected and in general we will not generate an expansion in $1/N$. Including crystal field effects is also clearly important for a realistic description of many of these compounds and gives rise to qualitatively different features. Crystal field splitting is most easily included in the '$1/N$' model where at low temperatures N is effectively 2. The physical picture is again modified when the correct spin–orbit coupling is included.

The layout of this paper is as follows. We start by considering the slave-boson approach to the periodic Anderson model where the f level is N-fold degenerate, $(N = 2j + 1)$, and the correct k- and spin-dependence of V is taken into account. In section 2 we consider the slave-boson mean-field solutions for this model and generate quasiparticle bands (these are essentially the same as those obtained by Zou and Anderson 1986). The mean-field solution is then used to calculate the magnetic susceptibility both at T=0 and at finite temperature. Within the slave-boson mean-field approximation the Van Vleck contribution to the susceptibility needs to be included for consistency and at $T = 0$ we recover the result from the $1/N$ model. We consider the arguments as to the validity of this result. As shown previously (Evans *et al* 1989) we can calculate the susceptibility up to temperatures well above the Kondo temperature. The results differ from those for the $1/N$ model in that we no longer find a pronounced Kondo peak at low temperatures.

In section 3 we use the same model to look at the Gaussian corrections to the mean-field solution. We use these to calculate the correction to the Wilson ratio and to look at the energy and frequency dependent susceptibility. The correction to the Wilson ratio has a term $\sim 1/N$ exactly as found in the $1/N$ model. There are, however, additional terms depending on μ_{eff}, the effective moment of the Pauli-like contribution to χ. $\operatorname{Im}\chi(\omega, k)$ is shown to be significantly different from that calculated with the $1/N$ model and has a much greater inelastic contribution.

We consider next the inclusion of crystal field splitting. We look first (section 4) at the inclusion of crystal field splitting within the $1/N$ model, i.e. we neglect the k- and spin-dependence of V, but lift the f-level degeneracy to give three doublets. We use the mean-field solution use to calculate the susceptibility as a function of temperature. This is strongly anisotropic and depends sensitively on the details of the crystal field levels. The effect of crystal field splitting on the Gaussian fluctuations is briefly mentioned.

The generalization to a model including both spin–orbit coupling and crystal field splitting is discussed in section 5. The mean-field solution is more complicated and in general we need to solve a quartic equation to find the quasiparticle bands. We can however calculate γ and χ_z at $T = 0$ without solving for the bands, giving precisely the results found previously.

Finally, in section 6 we compare our results for the anisotropy of χ with experiment. This gives us some sort of test of the accuracy of the $1/N$ expansion with $N = 2$. The quantitative agreement is far better than we might reasonably expect and several qualitative features are correctly predicted.

2. The magnetic susceptibility including spin–orbit coupling

We consider here the periodic Anderson model with a realistic hybridization matrix element for a six-fold degenerate f level. The hybridization term is of the form $\langle V_{m\sigma}(k) = V_0 Y_{3m-\sigma}(k)\langle 3lm-\sigma, \frac{1}{2}\sigma|\frac{5}{2}m\rangle$, where $Y_{3m-\sigma}(\boldsymbol{k})$ is the spherical harmonic for $l = 3$ and $\langle 3lm-\sigma, \frac{1}{2}\sigma|\frac{5}{2}m\rangle = (4\pi)^{1/2}[(7-4\sigma m/14)]^{1/2}$ is the Clebsch–Gordan coefficient for spin–orbit coupled states with $j = \frac{5}{2}$ and $l = 3$.

The slave-boson method allows us to write an effective Hamiltonian which is equivalent to this model in the limit $U \to \infty$ (see, for example, Millis and Lee 1987 and Rasul and Desgranges 1986 for details of the method). If we then take the 'mean-field' approximation we are left with an effective hybridization Hamiltonian in which the hybridization, $V_{m\sigma}(k)$, and f level energies, E_{0m}, are renormalized. The mean-field effective Hamiltonian including spin–orbit coupling can then be written

$$H = \sum_{k\sigma} \epsilon_k c^\dagger_{k\sigma} c_{k\sigma} + \sum_{im} \epsilon_{fm} f^{i\dagger}_m f^i_m + \sum_{k\sigma im} (\tilde{V}_{m\sigma}(k)\exp(\mathrm{i}\boldsymbol{R}_i\cdot\boldsymbol{k}) c^\dagger_{k\sigma} f^i_m + \mathrm{HC}) + \mathrm{i}\lambda(\rho^2-1) \tag{2.1}$$

where $\tilde{V}_{m\sigma}(k) = \rho V_{m\sigma}(k)$ and $\epsilon_{fm} = E_{0m} + \mathrm{i}\lambda$. ρ and $\mathrm{i}\lambda$ are the mean-field parameters which are determined by minimising the free energy. This gives $\rho^2 = 1 - n_f$ where n_f is the mean f valence. The effective hybridization becomes small in the Kondo limit ($n_f \to 1$). ϵ_{fm} gives the energy scale for the system and is small in the same limit. In this section we shall consider the degenerate case and drop the subscript m from ϵ_{fm}.

It is easily seen that only two orthogonal linear combinations of f levels hybridize with the conduction electrons and by defining

$$f^i_\sigma = \frac{1}{3^{1/2}V_0}\sum_m V_{m\sigma}(k) f^i_m \tag{2.2}$$

we can rewrite the Hamiltonian as

$$H = \sum_{k\sigma} \epsilon_k c^\dagger_{k\sigma} c_{k\sigma} + \sum_{i\eta} \epsilon_f f^{i\dagger}_\eta f^i_\eta + \sum_{k\sigma i} (\tilde{V} \mathrm{e}^{\mathrm{i}\boldsymbol{R}_i\cdot\boldsymbol{k}} c^\dagger_{k\sigma} f^i_\sigma + \mathrm{HC}) + \mathrm{i}\lambda(\rho^2-1) \tag{2.3}$$

where $V = 3^{1/2}V_0$. The sum over η is a sum over the six orthogonal linear combinations of f^i_m which include f^i_σ. It is clear that we obtain two quasiparticle bands and four f levels localized at ϵ_f.

The mean-field equations can be written in the usual way and we obtain

$$1 - \rho^2 = n_f = -T\sum_{\boldsymbol{k},\eta} G^f_\eta(\boldsymbol{k}) \tag{2.4}$$

$$\epsilon_f = E_0 + TV^2 \sum_{\boldsymbol{k},\sigma} G_0 G^f_\sigma(\boldsymbol{k}) \tag{2.5}$$

where $G^f_\eta(\boldsymbol{k})$ is the Fourier transform of $\langle T_\tau f^{i\dagger}_\eta(\tau) f^i_\eta(0)\rangle$ and $G_0 = (\omega - \epsilon_k)^{-1}$. Here $\boldsymbol{k}$ represents the four vector (k, ω).

These can be solved at finite temperature. It has previously been shown that the mean-field solution for the lattice differs from that for the single impurity in that we need to include the temperature dependence of the Fermi level, μ. This can have a significant effect on the value of the artificial phase transition, T_c, (Evans *et al* 1989). The Fermi level changes by an amount $\sim W(1 - n_f)$ from $T = 0$ to $T = T_c$ and the value of the bare f-level energy E_0 as measured from the Fermi level is shifted by the same amount. The mean-field equations are very sensitive to the value of E_0 and, for example, with the parameters used previously, i.e. $V/W = 0.1$ and $E_0/W = -0.08$, we obtain $T_c \approx 10T_K$. The result is, however, dependent on the bare parameters chosen. For n_f at $T = 0$ very close to one (the Kondo limit) the effect becomes negligible and we get back to the one impurity result, $T_c \sim T_K$. However, in the 'scaling' limit, $W \to \infty$, $T_c \to \infty$ also. The results at finite temperature, therefore, become non-universal and we expect the temperature dependence of the susceptibility to depend on T_c. In fact 'universal' behaviour is regained as T_c becomes large and we shall perform calculations in this region. We note that a calculation of the transition temperature for the lattice has recently appeared (Harigaya 1990). Similar parameters to those above were used but the temperature dependence of the Fermi level was neglected leading to a small transition temperature being found.

We want now to consider the calculation of the magnetic susceptibility within this model. The free energy is given by

$$F = T\sum_{k,i\omega}\sum_{\sigma,\alpha=\pm} \ln[i\omega - E_{\alpha\sigma}(k)] + T\sum_{i\omega,\eta\neq\sigma} \ln(i\omega - \epsilon_{f\eta}) - n_f(\epsilon_f - E_0) \tag{2.6}$$

where $E_{\pm\sigma}(k)$ are the quasiparticle energies for the two hybridized bands

$$E_{\pm\sigma}(k) = \tfrac{1}{2}\{\epsilon_k + \epsilon_{f\sigma} \pm [(\epsilon_k - \epsilon_{f\sigma})^2 + 4\tilde{V}^2]^{1/2}\} \tag{2.7}$$

and all energies are defined relative to the Fermi energy. To calculate the susceptibility we transform to real frequencies and take the second derivative with respect to h. For this we need to expand $E_{\alpha\sigma}(k)$ and $\epsilon_{f\eta}$ as perturbation series in h. We write wavefunctions for ψ_σ, describing the quasiparticle bands, and ψ_η $(\eta \neq \sigma)$ for the local levels. ψ_σ are essentially the same as those derived by Zou and Anderson (1986). We then need to take linear combinations of these, ψ_μ and ψ_ν, such that $\langle\psi_\mu|\mu_z|\psi_\nu\rangle = 0$. This allows us to write

$$E_{-\mu}(h) = E_-(0) + h\langle\psi_\mu|\mu_z|\psi_\mu\rangle + h^2\sum_{\eta\neq\mu}\frac{|\langle\psi_\mu|\mu_z|\psi_\eta\rangle|^2}{E_- - \epsilon_f} \tag{2.8}$$

with analagous expressions for $E_{+\mu}$ and $\epsilon_{f\eta}$. The first term is $O(h)$ and gives rise to Pauli or band like terms and the second, $O(h^2)$, gives a Van Vleck contribution. We now have

$$\chi = \frac{\partial^2 F}{\partial h^2} = \sum_{\alpha k}\int d\epsilon\, f(\epsilon)\left\{\delta'(\epsilon - E_\alpha)\left(\frac{\partial E_{\mu\alpha}}{\partial h}\right)^2 + \delta(\epsilon - E_\alpha)\frac{\partial^2 E_{\mu\alpha}}{\partial h^2}\right\}$$
$$+ \sum_{\eta\neq\sigma}\int d\epsilon\, f(\epsilon)\left\{\delta'(\epsilon - \epsilon_{f\eta})\left(\frac{\partial \epsilon_{f\eta}}{\partial h}\right)^2 + \delta(\epsilon - \epsilon_{f\eta})\frac{\partial^2 \epsilon_{f\eta}}{\partial h^2}\right\}. \tag{2.9}$$

Evaluating the first matrix element in (2.8) gives

$$\langle\psi_\mu|\mu_z|\psi_\mu\rangle = \frac{g\mu_B\tilde{V}^2}{\tilde{V}^2+(E_- - \epsilon_f)^2}\left[\left(\frac{\sum_m m|V_{m\sigma}|^2}{V^2}\right)^2 + \left(\frac{\sum_m mV_{m\sigma}V^*_{m\sigma}}{V^2}\right)^2\right]^{1/2}. \quad (2.10)$$

Using this, the contribution to the susceptibility at $T = 0$ from the Pauli-like terms is $\chi = 2\mu_{\rm eff}^2\rho_0 m^*/m$ where $m^*/m = \tilde{V}^2/\epsilon_f^2$ is the mass enhancement which is large, and the effective moment $\mu_{\rm eff}$ is much smaller than the full magnetic moment, μ_0: $\mu_{\rm eff}^2 \approx 0.18\mu_0^2$. It is claimed by Anderson and Zou (1987) that it is sufficient to consider these terms only and that this gives an explanation of the experimental observation that the Wilson ratio, which is evaluated using the full magnetic moment, is somewhat less than one.

The calculation neglects the contribution from the second term, the 'Van Vleck' terms which can be seen to have an effective moment $\mu_0^2 - \mu_{\rm eff}^2$. Including these the susceptibility is given by

$$\chi = 4\rho_0\mu_0^2\sum_\alpha \int_{y_\alpha^-}^{y_\alpha^+} \frac{\tilde{V}^2}{(\epsilon_f - E_\alpha)^3} f(E_\alpha)\,dE_\alpha - \mu_{\rm eff}^2\left(\frac{2\rho_0\tilde{V}^2}{(\epsilon_f - y_-^+)^2}f(y_-^+) - \frac{2\rho_0\tilde{V}^2}{(\epsilon_f - y_+^-)^2}f(y_+^-)\right)$$
$$- (\mu_0^2 - \mu_{\rm eff}^2)\left(\frac{2\rho_0\tilde{V}^2}{(\epsilon_f - y_-^+)^2}f(\epsilon_f) - \frac{2\rho_0\tilde{V}^2}{(\epsilon_f - y_+^-)^2}f(\epsilon_f)\right) \quad (2.11)$$

where $y_\alpha^+ = E_\alpha(W - \mu)$ and $y_\alpha^- = E_\alpha(-W - \mu)$. The Fermi level, μ, can be approximated by $\mu = W(1 - n_f)$. At $T = 0$ we regain a susceptibility with the full magnetic moment and a Wilson ratio of one as previously found by several people (Zhang and Lee 1987, Aeppli and Varma 1987, Cox 1987). The $1/N$ model and the spin–orbit coupled model thus give the same result at $T = 0$. Extending the calculation to finite temperatures, however, we find a somewhat different behaviour. At low T the susceptibility is virtually independent of temperature with a very weak maximum at $T \sim 0.2T_K$. This is followed almost immediately by a $1/T$ behaviour. This can be compared with the T dependence in the $1/N$ model where there is a pronounced Kondo maximum again at $T \sim 0.2T_K$ (Evans *et al* 1989). In both cases the magnetic moment in the high-temperature regime is reduced by a factor $\sim 1 - 1/N$ due to the mean-field approximation. It is puzzling that the more realistic model produces poorer results.

If we include only the Pauli contribution then the moment in the Curie–Weiss regime is $\sim \mu_{\rm eff}^2$. According to Anderson and Zou (1987) we also need to include the unrenormalized Van Vleck terms but these will give us a Curie–Weiss law only for $T > \Delta_0 = \rho_0 V^2/\pi$.

For exactly 2 electrons per site the lower band is just full and there is no contribution from the Pauli-like terms. In this case the Van Vleck terms remain and the susceptibility is still large. As suggested by Anderson and Zou (1987) we may hope then to deduce experimentally whether or not the Van Vleck terms need to be included in the calculation. There is experimental evidence that for SmB_6 the Fermi level lies in the gap so we appear to be able to resolve the situation by looking at the magnetic susceptibility of this compound. The situation is not, however, this simple. The addition of a single additional electron into our system pushes the Fermi level to ϵ_f and the contribution to the susceptibility from the Van Vleck terms also becomes zero. The system is therefore very sensitive to small perturbations. It appears that the experimental results are not sufficient to chose between the two theories.

3. The fluctuation terms

We have seen in the previous section the effect that including realistic spin–orbit coupling has on the lowest order approximation, the 'mean-field' solution. We now wish to look at the effect this has at the next level of approximation, the Gaussian fluctuations. In the mean-field theory we have written $\rho(\boldsymbol{k}) = \rho + \tilde{\rho}(\boldsymbol{k})$ and $\mathrm{i}\lambda(\boldsymbol{k}) = \mathrm{i}\lambda + \mathrm{i}\tilde{\lambda}(\boldsymbol{k})$ where ρ and $\mathrm{i}\lambda$ are constant, and then assumed that $\tilde{\rho}(\boldsymbol{k})$ and $\mathrm{i}\tilde{\lambda}(\boldsymbol{k})$ are small and can be neglected. We want now to include terms quadratic in $\tilde{\rho}$ and $\tilde{\lambda}$. The free energy can be written

$$F = -T \ln Z = F_{\mathrm{mf}} + T \sum_{\boldsymbol{k}} \mathrm{Tr} \ln \det \mathbf{S}^{-1}(\boldsymbol{k}) \tag{3.1}$$

where F_{mf} is the contribution from the mean-field terms and $\mathbf{S}$ is a 2 by 2 matrix coming from the fluctuations. The elements of $\mathbf{S}$ can be written

$$S_{11}(\boldsymbol{k}) = S_{\rho\rho}(\boldsymbol{k}) = \epsilon_{\mathrm{f}} - E_0 - TV^2 \sum_{\boldsymbol{k}_1 \sigma} G^f_\sigma(\boldsymbol{k}_1) G_0(\boldsymbol{k}_1 + \boldsymbol{k}) + \frac{T\tilde{V}^2 V^2}{2} \sum_{\boldsymbol{k}_1 \sigma} G^f_\sigma(\boldsymbol{k}_1) G^f_\sigma(\boldsymbol{k}_1 + \boldsymbol{k}) (G_0(\boldsymbol{k}_1) + G_0(\boldsymbol{k}_1 + \boldsymbol{k}))^2 \tag{3.2}$$

$$S_{12}(\boldsymbol{k}) = S_{\lambda\rho}(\boldsymbol{k}) = 2\rho + TV^2 \rho \sum_{\boldsymbol{k}_1 \sigma} G^f_\sigma(\boldsymbol{k}_1) G^f_\sigma(\boldsymbol{k}_1 + \boldsymbol{k}) (G_0(\boldsymbol{k}_1) + G_0(\boldsymbol{k}_1 + \boldsymbol{k})) + TV^2 \rho \sum_{\boldsymbol{k}_1 \sigma \eta \neq \sigma} G^f_\sigma(\boldsymbol{k}_1) G^f_\eta(\boldsymbol{k}_1 + \boldsymbol{k}) (G_0(\boldsymbol{k}_1) + G_0(\boldsymbol{k}_1 + \boldsymbol{k})) \tag{3.3}$$

$$S_{22}(\boldsymbol{k}) = S_{\lambda\lambda}(\boldsymbol{k}) = \frac{T\rho}{2} \sum_{\boldsymbol{k}_1 \sigma} G^f_\sigma(\boldsymbol{k}_1) G^f_\sigma(\boldsymbol{k}_1 + \boldsymbol{k}) + \frac{T\rho}{2} \sum_{\boldsymbol{k}_1 \sigma \eta \neq \sigma} G^f_\sigma(\boldsymbol{k}_1) G^f_\eta(\boldsymbol{k}_1 + \boldsymbol{k}) + \frac{T\rho}{2} \sum_{\boldsymbol{k}_1 \eta \neq \sigma} G^f_\eta(\boldsymbol{k}_1) G^f_\eta(\boldsymbol{k}_1 + \boldsymbol{k}). \tag{3.4}$$

These can be compared with the matrix elements for the $1/N$ model (Rasul and Desgranges 1986).

We can now calculate the correction to the Wilson ratio. The contributions to χ and γ consist of a variety of terms. First, there are terms which come from the corrections to n_{f} and ϵ_{f}. These are clearly the same for both χ and γ. Secondly there are terms coming from the temperature and field dependence of $S(\boldsymbol{k})$. Within the $1/N$ model these also give the same contribution to χ and γ. The situation is somewhat more complicated in the present case. We need to evaluate $\partial^2/\partial h^2$ applied to the elements of $\mathbf{S}$. At $T = 0$ there are two types of term. There are intraband terms from the lower band and interband terms which are most important between the lower quasiparticle band and the local levels. For the intraband terms we can take $\partial^2/\partial h^2$ inside the sum over k and write

$$\frac{\partial^2}{\partial h^2} = \left(\frac{\partial E_-}{\partial h}\right)^2 \frac{\partial^2}{\partial E_-^2} + \frac{\partial E_-^2}{\partial h^2} \frac{\partial}{\partial E_-}. \tag{3.5}$$

We can then perform a partial integration for the second term to obtain $\partial^2/\partial h^2 = \mu_0^2 \partial^2/\partial E_-^2$. The contribution to γ from these terms is entirely equivalent. In the

interband terms, however, both E_- and $\epsilon_{f\eta}$ depend on h and we obtain additional terms. There are differences between the temperature and field dependence and we obtain a contribution to the Wilson ratio which depends on $\mu_{\rm eff}^2$. It is difficult, however, to estimate the magnitude of this term without performing the full calculation.

Finally we have a term that contributes to the temperature dependence but not the field dependence of the free energy coming from the T dependence implicit in the frequency sum in the expression for $F_{\rm G}$. We can rewrite the contribution to the free energy from the fluctuation terms as

$$F_{\rm G} = \frac{1}{2}\sum_{k}\int \frac{{\rm d}\omega}{2\pi} n_{\rm B}(\omega)\tan^{-1}\left(\frac{{\rm Im}\det \mathbf{S}(k)}{{\rm Re}\det \mathbf{S}(k)}\right) \tag{3.6}$$

where $n_{\rm B}$ is the Bose function. To calculate the term in T^2 needed for the specific heat we only need to know the leading order terms in ω for $\mathbf{S}$. If we first neglect the terms coming from the localized levels, we find that the correction to the Wilson ratio is $\sim 1/2$ in the Kondo limit, the result we would have for an f degeneracy of 2 in the $1/N$ model (Millis and Lee 1987). We now need to see how the localized levels affect this result. For $\omega \to 0$ the imaginary part of $\mathbf{S}$ comes purely from intraband terms and the localized levels play no part. For the real part, however, the interband terms are of the same form as the intraband terms in the $T, \omega, k \to 0$ limit. This leads to a contribution to the Wilson ratio $\sim 1/N$, exactly as is found for the $1/N$ model.

We can also calculate the energy and frequency dependent magnetic susceptibility. It is interesting to note that for small ω the interband terms again do not contribute to ${\rm Im}\,\chi(k)$ and we therefore find that the effective moment for this is $\propto \mu_{\rm eff}^2$. At higher frequencies the full magnetic moment will be recovered. We find, therefore, a much greater weight for the inelastic contribution compared with the quasielastic contribution than is found in the $1/N$ model. In contrast the real part of the susceptibility as ω goes to zero depends on the full magnetic moment. The Fermi liquid relation between the two becomes

$$\lim_{\omega\to 0}\frac{{\rm Im}\,\chi(k,\omega)}{\omega} = \frac{\pi k_{\rm F}}{2k}\frac{\mu_{\rm eff}^2}{2\mu_0^4}\,{\rm Re}\,\chi(0)^2 \tag{3.7}$$

The $1/N$ result is recovered by replacing $\mu_{\rm eff}$ by μ_0 and the factor 2 by N. It appears that the imaginary part of the susceptibility, important for modeling neutron scattering, is significantly different in this model compared with the $1/N$ approach.

4. Crystal field splitting in the $1/N$ model

Having considered the effect of spin–orbit coupling in the case where the f level is degenerate, we now look at the effects of crystal field splitting. For simplicity we consider first the $1/N$ model where we lift the f-level degeneracy but neglect the k- and spin dependence of V, i.e. we put $V_{m\sigma}(k) \to V$ and $c_{k\sigma} \to c_{km}$ in (2.1). We consider principally the mean-field solution.

In non-cubic compounds the effect of the crystalline electric field is to split the N-fold degenerate level into doublets. The term in the Hamiltonian corresponding to the localized f level will be of the form $\sum_{iM}\epsilon_{{\rm f}M} f_M^{i\dagger}(k) f_M^i(k)$ where we have used M to denote the linear combination of m states which give eigenstates of the crystal field. For the three values of $|M|$ we can then write $\epsilon_{{\rm f}M_1} = \epsilon_{{\rm f}-M_1} = \epsilon_{\rm f0}$ for the

ground state, $\epsilon_{fM_2} = \epsilon_{f-M_2} = \epsilon_{f0} + \Delta_1 = \epsilon_{f1}$ for the first crystal field level and $\epsilon_{fM_3} = \epsilon_{f-M_3} = \epsilon_{f0} + \Delta_2 = \epsilon_{f2}$ for the second level. Within the slave-boson mean-field approximation we will have six quasiparticle bands with strong peaks in the densities of states at ϵ_{f0}, ϵ_{f1} and ϵ_{f2} respectively. The f electron doublets are all shifted by the same amount $i\lambda$ and there is no rescaling of the crystal field splitting. It is interesting to ask whether the same crystal field splitting is preserved to higher order in the expansion. If we calculate the f electron self-energy, Σ_m^f, to leading order in the fluctuations we find that this is dependent on m and hence the higher order effects do in fact alter the splitting. We can compare with a different method, the variational method, applied to the one impurity problem. Here the crystal field splitting is also preserved when we turn on the hybridization. It has been shown, however, that including an anisotropic mixing interaction (i.e. $V_0 \rightarrow V_m$) gives rise to a splitting of the f levels of the correct order of magnitude to account for the splitting observed (Levy and Zhang 1989). If we included this effect in the present model we would then find an f-level splitting in the Fermi liquid state which is very different to that of the magnetic ions.

The mean-field equations can be written as before. We now have

$$n_f = -T \sum_{kM} G_M^f(k) \tag{4.1}$$

and

$$\epsilon_f = E_0 + TV^2 \sum_k G_0(k) G_M^f(k) \tag{4.2}$$

where $G_M^f(k)$ is the Fourier transform of $\langle T_\tau f_M^{i\dagger}(\tau) f_M^i(0)\rangle$. Within this '$1/N$' type model there is no term giving scattering between subbands with different values of M and to be consistent we ought to compute the Fermi level separately for each subband. This feature has previously been noted for the one-impurity problem (Nozières and Blandin 1980) where it is a real effect. In the present case it is an artifact of the $1/N$-like approach and represents a shortcoming of the model. Although not self-consistent it appears more realistic to assume a common Fermi level for the three hybridized subbands.

We can now calculate the susceptibility both along the c axis, χ_z, and perpendicular to it, $\chi_{x,y}$. The susceptibility consists of a mixture of 'intraband' or 'Pauli' terms and 'interband' or 'Van-Vleck' like terms. In general we can write

$$\chi_\mu = 2\rho_0 \sum_{\alpha,n=0,2} \alpha_n^\mu \int_{y_{\alpha n}^-}^{y_{\alpha n}^+} \frac{\tilde{V}^2}{(E_{\alpha n} - \epsilon_{fn})^2} f'(E_{\alpha n})\, dE_{\alpha n}$$
$$+ 2\rho_0 \beta_n^\mu \int_{-W-\mu}^{W-\mu} \frac{f(E_{\alpha n}(k)) - f(E_{\alpha n+1}(k))}{E_{\alpha n}(k) - E_{\alpha n+1}(k)} A_{\alpha n}(k) A_{\alpha n+1}(k)\, d\epsilon_k \tag{4.3}$$

where the sum over n runs over the three crystal field levels and we have defined the index $n+1$ to be 0 for $n = 2$. $A_{\alpha n} = \tilde{V}^2/(\tilde{V}^2 + (E_{\alpha n} - \epsilon_{fn})^2)$ and $E_{\alpha n}$ is given by (2.7) but with $\epsilon_f \rightarrow \epsilon_{fn}$. The coefficients α_n^μ and β_n^μ depend on the crystal field configuration. For the configuration

$$|0\rangle = |\pm\tfrac{1}{2}\rangle \qquad |1\rangle = |\pm\tfrac{3}{2}\rangle \qquad |3\rangle = |\pm\tfrac{5}{2}\rangle \tag{4.4}$$

we have

$$\alpha_0^z = \tfrac{1}{4} \qquad \alpha_1^z = \tfrac{9}{4} \qquad \alpha_2^z = \tfrac{25}{4}$$
$$\beta_0^z = \beta_1^z = \beta_2^z = 0 \tag{4.5}$$

and

$$\begin{aligned} &\alpha_0^x = \tfrac{9}{4} \qquad \alpha_1^x = \alpha_2^x = 0 \\ &\beta_0^x = 4 \qquad \beta_1^x = \tfrac{5}{2} \qquad \beta_2^x = 0. \end{aligned} \tag{4.6}$$

We can consider also the configuration

$$\begin{aligned} |0\rangle &= a|\pm\tfrac{5}{2}\rangle - b|\mp\tfrac{3}{2}\rangle \\ |1\rangle &= |\pm\tfrac{1}{2}\rangle \\ |2\rangle &= a|\pm\tfrac{3}{2}\rangle + b|\mp\tfrac{5}{2}\rangle. \end{aligned} \tag{4.7}$$

As has been shown by Zhang and Levy (1989) we now get

$$\begin{aligned} &\alpha_0^z = \tfrac{1}{4}(5a^2 - 3b^2)^2 \qquad \alpha_1^z = \tfrac{1}{4} \qquad \alpha_2^z = \tfrac{1}{4}(3a^2 - 5b^2)^2 \\ &\beta_0^z = \beta_1^z = 0 \qquad \beta_2^z = -2(4ab)^2 \end{aligned} \tag{4.8}$$

and

$$\begin{aligned} &\alpha_0^x = \alpha_2^x = 5ab \qquad \alpha_1^x = \tfrac{9}{4} \\ &\beta_0^x = -4b^2 \qquad \beta_1^x = -4a^2 \qquad \beta_2^x = -\tfrac{5}{2}(a^2 - b^2)^2. \end{aligned} \tag{4.9}$$

For a large crystal field splitting we can approximate $\chi_\mu = 2\rho_0 g^2 \mu_B^2 \alpha_K \tilde{V}^2/\epsilon_{fK}^2$ where K is the ground state. In fact we expect the contribution from the upper crystal field levels to be significant, especially from the interband terms which fall off only as $1/\Delta$.

We can compare with the expression for γ

$$\gamma = 2\rho_0 \frac{k_B^2 \pi^2}{3}\left(\frac{\tilde{V}^2}{\epsilon_{f0}^2} + \frac{\tilde{V}^2}{\epsilon_{f1}^2} + \frac{\tilde{V}^2}{\epsilon_{f2}^2}\right). \tag{4.10}$$

If $\Delta_1, \Delta_2 \gg \epsilon_f$, a Wilson ratio of one is obtained provided the effective moment used in evaluating it, is that of the lowest energy doublet.

The susceptibility can also be calculated as a function of T. We note that χ is approximately a universal function of $\Delta_1/T_K, \Delta_2/T_K$ as is found from exact calculations for the one impurity model (Schlottman 1984). The results depend strongly on the values of $\Delta_1/T_K, \Delta_2/T_K$ and on the configuration used. The results are compared with experiment in the last section.

Finally in this section we mention the effect of crystal field splitting on the Gaussian fluctuations. The correction to the Wilson ratio can be calculated and is given by $\Delta R \sim \frac{1}{2} + O(T_K/\Delta_1, T_K/\Delta_2)$. The energy and frequency dependent susceptibility can also be calculated. We note that the relationship between $\mathrm{Im}\,\chi/\omega$ and $\mathrm{Re}\,\chi$ is more complicated in this case and there is no simple Korringa relation.

5. Crystal field splitting and spin–orbit coupling

In section 2 we saw that for a degenerate f level the physical picture is different depending on whether or not we include the spin–orbit coupling and that the results for

the two models are not necessarily the same. In view of this we consider here the case where both crystal field splitting and spin–orbit coupling are included in the model. Looking at the mean-field solution this gives a qualitatively different picture with four quasiparticle bands. Calculating the bands involves solving a quartic equation and we whall not attempt to do that here. We shall show, however, that it is possible to calculate γ and $\chi_z(0)$ without solving for the full band structure.

For simplicity we consider the case where the crystal field levels are pure m states. It is convenient to define linear combinations of f_m and f_{-m} which hybridize with the conduction electrons of a given spin

$$f_{n\sigma} = \frac{1}{(|V_{m\sigma}|^2 + |V_{-m\sigma}|^2)^{1/2}}(V_{m\sigma} f_m + V_{-m\sigma} f_{-m}) \tag{5.1}$$

Substituting (5.1) in (2.1) we obtain

$$H = \sum_{k\sigma} \epsilon_k c^\dagger_{k\sigma} c_{k\sigma} + \sum_{in\sigma} \epsilon_{fn} f^{i\dagger}_{n\sigma} f^i_{n\sigma} + \sum_{k\sigma in} (\tilde{V} h_m(k) c^\dagger_{k\sigma} f^i_{n\sigma} \exp(\mathrm{i}\boldsymbol{R}_i \cdot \boldsymbol{k}) + \mathrm{HC}) - \mathrm{i}\lambda n_f \tag{5.2}$$

where $h^2_{1/2}(k) = \frac{3}{4}(1 - 2x^2 + 5x^4)$, $h^2_{3/2}(k) = \frac{3}{8}(1 + 14x^2 - 15x^4)$ and $h^2_{5/2}(k) = \frac{15}{8}(1 - 2x^2 + x^4)$ with $x = \cos\theta$.

For large crystal field splitting it is reasonable to assume that it is only the ground state doublet which is involved in forming the lowest quasiparticle band. γ and χ_z can be calculated at $T = 0$ with this approximation and a Wilson ratio of one is found exactly as above. There is no change in χ due to the terms $h_m(x)$ and the reduction of the Wilson ration proposed by Zou and Anderson (1986) is not found.

We now wish to consider what happens when we allow for the presence of the higher lying doublets. The following relation between the f and c Green functions is easily shown:

$$G^f_{n\sigma}(k,\omega) = \frac{1}{\omega - \epsilon_{fn}} + \frac{\tilde{V}^2 h^2_m(k)}{(\omega - \epsilon_{fn})^2} G^c_\sigma(k,\omega)\,. \tag{5.3}$$

Ignoring the small contribution from the conduction electrons γ is found from $\mathrm{Im}\, G^f_{n\sigma}(0)$. Assuming the Fermi level lies in the lowest band, $E_{-0}(k)$, this gives $\mathrm{Im}\, G^f_{n\sigma} = \tilde{V}^2 h^2_m (\omega - \epsilon_{fn})^{-2}\, \mathrm{Im}\, G^c_\sigma = \tilde{V}^2 h^2_m (\omega - \epsilon_{fn})^{-2} a_c \delta(\omega - E_{-0})$ where a_c is the conduction electron weight in the lower band given by $a_c = \partial E_{-0}/\partial \epsilon_k$. γ is again given by (4.10).

A similar calculation for χ_z can also be performed. The perturbation due to the magnetic field is written $H_1 = \sum_{mi} hm f^{i\dagger}_m f^i_m$. The magnetic moment which is induced is $\bar{m} = \sum_m m n_{fm}$ and the susceptibility is given by $\partial\bar{m}/\partial h$. In terms of $f_{n\sigma}$ we can write the perturbation due to the magnetic field as

$$H_1 = \sum_{n\sigma} h|m|[(|V_{m\sigma}|^2 - |V_{-m\sigma}|^2) f^{i\dagger}_{n\sigma} f^i_{n\sigma} + 2V_{m\sigma} V^*_{m-\sigma} f^{i\dagger}_{n\sigma} f^i_{n-\sigma}] \tag{5.4}$$

and

$$\bar{m} = \sum_{n\sigma} |m| \sum_{k,\omega} (|V_{m\sigma}|^2 - |V_{-m\sigma}|^2) G^f_{n\sigma}(\boldsymbol{k}) + 2V_{m\sigma} V^*_{m-\sigma} G^f_{n\sigma-\sigma}(\boldsymbol{k}) \tag{5.5}$$

where $G^f_{n\sigma-\sigma}$ is the Fourier transform of $\langle T_\tau f^{i\dagger}_{n\sigma}(\tau) f^i_{n-\sigma}(0)$.

Evaluating this we find two types of contribution. The first is straightforward involving only $\partial\epsilon_{fm}/\partial h$ and gives

$$\chi_z = 2\rho_0 \sum_m m^2 \frac{\tilde{V}^2}{\epsilon_{fm}^2}. \tag{5.6}$$

The second type of terms depend on $\partial E_{-0}/\partial h$ for which we need more information about the quasiparticle band. If, however, we go back to the case where $\Delta = 0$ we find this term vanishes when the Fermi level lies in the lower band and gives a contribution $\propto \mu_{\text{eff}}^2$ when the Fermi level lies in the gap (exactly the result we had before). We do not expect this term to make a big contribution to the susceptibility even when $\Delta \neq 0$ and we obtain exactly the result we had from the $1/N$ model. As in the degenerate case the inclusion of the spin–orbit coupling does not affect the result for $T = 0$. At finite temperatures, however, it is possible that slightly different results would be obtained.

The calculation of χ_x involves interband terms even at $T = 0$ and cannot therefore be calculated without the full bandstructure.

6. Comparison with experimental results

In this section we present our results and compare with experimental data. We note that this provides some kind of test of the $1/N$ method with $N = 2$.

We start by considering results for $T = 0$. In figure 1 we show the ratio χ_z/χ_x as a function of Δ_1/T_K, where $T_K = \gamma^{-1}$, for different crystal field levels. We have taken $\Delta_2 = 2\Delta_1$. The broken curve gives the results for crystal field levels given by (4.4). χ_z/χ_x decreases rapidly from one for the degenerate case to the limiting value of $\frac{1}{9}$. The other curves are for the crystal field scheme given by (4.7) with different values of a and b. We see that for a nominal ground state $|\pm\frac{5}{2}\rangle$, $\chi_z/\chi_x > 1$ and can become large especially for a close to 1. For the other case, i.e. nominal ground state $|\pm\frac{3}{2}\rangle$ the situation is more complicated and χ_z/χ_x can be greater than or less than 1. The value of the ratio is a sensitive function of both a and Δ_1. For $a^2 \approx 0.62$ χ_z/χ_x varies very little with Δ_1. The results can be compared with a similar calculation based on the Coqblin–Schrieffer model (Hanzawa 1986). The results are seen to be qualitatively similar.

We want now to consider the temperature dependence for several specific compounds. In order to see the crystal field effects we need $T_c > \Delta_1, \Delta_2$. Convenient parameters are $V/W = 0.01$ and $E_0/W = -0.0028$. These give $T_c > 100\,T_K$. The effective moment in the Curie–Weiss regime is reduced due to the mean-field approximation by a factor between 2 and $1/(1-6^{-1})$ depending on the relative values of T_c and Δ.

We look firstly at the compound $CePt_2Si_2$ which has crystal field configuration given by (4.4) and $T_K \sim 50$ K, $\Delta_1 \sim 80$ K $\sim 1.6\,T_K$ and $\Delta_2 \sim 230$ K $\sim 4.6T_K$. For $T = 0$ we find $\chi_z/\chi_x = 0.2$. This can be compared with the experimental value of 0.57 (Gignoux *et al* 1988). We note that $\chi_z > \chi_x$ in both cases but the values differ significantly. The theoretical value can be increased by taking smaller values of Δ_1 and Δ_2. At finite T we find that the shapes of the curves for χ_z and χ_x are qualitatively different. $1/\chi_x$ has relatively little structure. There is a very weak maximum at very low temperatures $\sim 0.1T_K$ followed almost immediately by a linear dependence on T.

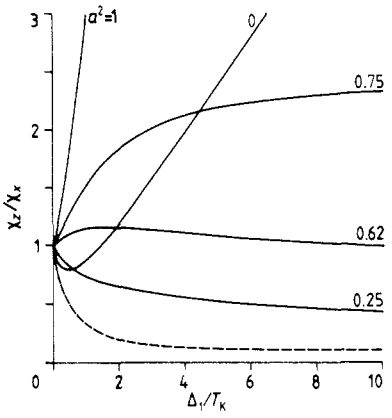

Figure 1. χ_z/χ_x at $T = 0$ for different crystal field schemes as a function of Δ_1/T_K. The broken curve corresponds to the crystal field scheme given by (4.4) and the full curves to (4.7) with different values of a^2.

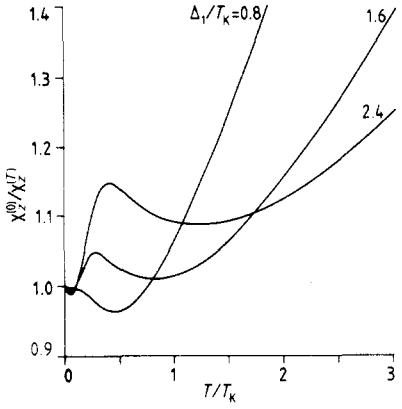

Figure 2. $\chi_z(0)/\chi_z(T)$ as a function of temperature for crystal field levels (4.4) for three different values of Δ_1/T_K.

$1/\chi_z$, in contrast, has a very distinctive structure. In figure 2 we show $\chi_z(0)/\chi_z(T)$ against T for three values of Δ_1/T_K with Δ_2/Δ_1 given by the experimental parameters listed above. We find a very weak minimum for $T \sim 0.1T_K$ after which there is a tendency for χ_z^{-1} to rise linearly with T with a slope corresponding to the magnetic moment of the ground-state doublet. This is followed by a maximum and then by a pronounced minimum after which the curve again rises linearly with T this time with the full magnetic moment. The position of the pronounced minimum scales approximately with Δ_1 and this feature is due to crystal field rather than Kondo effects. This feature in χ_z^{-1} is in qualitative agreement with experiment where a minimum due to crystal field effects is also seen (Ayache *et al* 1987). Experimentally, however, both χ_x and χ_z have a similar qualitative behaviour which is puzzling in view of our theoretical result. To clarify the situation we have performed a simple single ion crystal field calculation which is expected to give the correct qualitative

features associated with the crystal field while clearly missing the low temperature Kondo behaviour. In this calculation also the behaviour of χ_x and χ_z is very different and is consistent with the results described above. We note that the experimental results at low temperature indicated the presence of magnetic impurities which then had to be subtracted out. The apparent discrepancy between experiment and theory may then be due to experimental uncertainty in this region.

Table 1. Experimental parameters (see text for references).

Compound	a	b	T_K/K	Δ_1/T_K	Δ_2/T_K
$CeCu_2Si_2$	0.83	0.56	10	14	36
$CeRu_2Si_2$	0.96	0.28	19	12	53
$CeAl_3$	0.24	0.97	3	20	30
$CeSi_{1.86}$	0.45	0.89	40	8	7

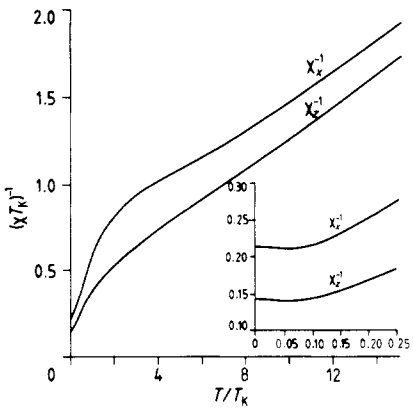

Figure 3. $(\chi_z T_K)^{-1}$ and $(\chi_x T_K)^{-1}$ as a function of temperature for $CeCu_2Si_2$ (parameters as in table 1). The inset shows the behaviour at very low temperatures.

We consider now those compounds which have crystal field levels given by (4.7). The experimental parameters for a number of compounds are given in table 1. In figure 3 we show the temperature dependence of $1/\chi_z$ and $1/\chi_x$ for $CeCu_2Si_2$ which has nominal ground state $|\pm\frac{5}{2}\rangle$. At $T = 0$, $\chi_z/\chi_x = 1.5$ which is between the two experimental values of 1.1 and 2 (Ōnuki *et al* 1984 and Assmus *et al* 1984). At very low temperatures there is a region relatively independent of T with a very weak minimum. The curves then rise monotonically showing a smooth crossover from the small magnetic moment characteristic of the ground state doublet to the full magnetic moment. This behaviour is fairly typical of all compounds with this level scheme and is in qualitative agreement with experiment. For $CeRu_2Si_2$, which also has nominal ground state $|\pm\frac{5}{2}\rangle$ (Lacerda *et al* 1989), we find a much larger anisotropy at $T = 0$ with $\chi_z/\chi_x = 11$ comparing reasonably well with the experimental value of 15 (Haen *et al* 1987). For the compounds with nominal ground state $|\pm\frac{3}{2}\rangle$ the situation is more complicated and it is possible for the two curves to cross. For $CeAl_3$ we find $\chi_z/\chi_x = 3.2$ at $T = 0$ in good agreement with the experimental value of

3.6 (Jaccard *et al* 1988). The two curves cross at $T \sim 3T_K$. This feature is also observed experimentally but at somewhat higher temperatures $\sim 12T_K$. For $CeSi_{1.86}$ the crossover is in the opposite direction. At $T = 0$ we have $\chi_z/\chi_x = 0.87$ compared with the experimental value 0.56 (Pierre *et al* 1990). The curves cross at $T \sim 0.5T_K$, which is again somewhat smaller than the experimental value of $2T_K$. The results for $T = 0$ are summarized in table 2.

Table 2. Comparison between theory and experiment for the ratio χ_z/χ_x at $T = 0$.

	χ_z/χ_x	
Compound	Experiment	Theory
$CePt_2Si_2$	0.57	0.2
$CeCu_2Si_2$	1.1, 2.0	1.5
$CeRu_2Si_2$	15	11
$CeAl_3$	3.6	3.2
$CeSi_{1.86}$	0.56	0.87

7. Conclusion

We have considered here the inclusion of spin–orbit and crystal field splitting within the slave-boson approach to the periodic Anderson model. Looking first at the case where the f level is six-fold degenerate, we find that at the mean-field level the use of the correct spin–orbit coupled quasiparticle bands in place of the $1/N$ bands makes no difference to the Wilson ratio as has been previously discussed. At finite temperatures, however, the behaviour is somewhat different with a much less pronounced Kondo maximum in χ for low temperatures. Within the same model we can then go beyond the mean field level and consider the fluctuations. We use these to look at the correction to the Wilson ratio. A term $\sim 1/N$ is found exactly as in the $1/N$ model but there are additional terms which depend on the value of μ_{eff}, the effective moment for the band contribution to the susceptibility. Furthermore, $\text{Im}\,\chi(k,\omega)$ is significantly different with a much smaller value as $\omega \to 0$ and correspondingly more weight for the inelastic contribution. In contrast we note that the T^2 term in the resistivity can also be calculated and in this case the result is equivalent to the $1/N$ model. We conclude that care is needed in using the $1/N$ model and in general we cannot assume that the model is equivalent to one in which the correct spin–orbit coupled quasiparticle bands are used.

We consider next the effect of crystal field splitting and show that this is important. By including crystal field splitting in the $1/N$ model at the mean-field level we obtain a large anisotropy for the magnetic susceptibility which depends sensitively on the details of the crystal field scheme. The ratio of χ_z to χ_x at $T = 0$ can be compared with other theoretical approaches and with experiment giving us some idea as to how well the $1/N$ approach works when $N = 2$. The quantitive agreement for χ_z/χ_x at $T = 0$ is good for most of the compounds considered. Furthermore several qualitative features are well described. This agreement with experiment is much better than we could reasonably expect and gives some justification for the view that the mean-field solution gives a better description of the normal state than the largeness of the

expansion parameter would lead us to believe. We can compare with a limited extent the effect of the correct spin–orbit coupling in the model with crystal field splitting. For $T = 0$ both γ and χ_z are unchanged as for the degenerate case. The T^2 term in the resistivity can also be calculated and this too is equivalent in the two models and gives an important anisotropy (Evans *et al* 1990). In analogy with the degenerate case we may, however, expect some differences at finite temperatures and in the Gaussian fluctuations.

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