The experimental peak in Fig. 3 shows all the characteristics described above for $H \| \langle 100 \rangle$ and, furthermore,

$$\Gamma_e/(\Gamma + \Delta) = 0.67 \ .$$

Thus, we conclude that the magnetic easy axis in γ'-Fe_4N lies along the $\langle 100 \rangle$ direction.

Knowing that the easy axis is along $\langle 100 \rangle$, it is possible to calculate the EFG at the Fe II sites in γ'-Fe_4N. From Eq. (2), using the S_Q values in Table I, we find that for the Fe II atoms

$$e^2qQ = -0.87 \text{ mm/sec} \ . \quad (4)$$

Using the known value of $Q = +0.2b$,[8] we calculate

$$eq = -7.0 \times 10^{14} \text{ esu/cm}^3 \ . \quad (5)$$

*Present address: TRW, IRC Philadelphia Division, Philadelphia, Pa.

[1]A. J. Nozik, J. C. Wood, Jr., and G. Haacke, Solid State Commun. 7, 1677 (1969).

[2]A. J. Nozik, J. C. Wood, Jr., and G. Haacke, Solid State Commun. 8, viii(E) (1970).

[3]E. Lehrer, Z. Elektrochem. 37, 460 (1930).

[4]P. H. Emmett, S. B. Hendricks, and S. Brunauer, J. Am. Chem. Soc. 52, 1456 (1930).

[5]V. G. Paranjpe, M. Cohen, M. B. Bever, and C. F. Floe, J. Metals 188, 261 (1950).

[6]R. H. Jack, Proc. Roy. Soc. (London) A195, 34 (1948).

[7]G. K. Wertheim, *Mössbauer Effect: Principles and Applications* (Academic, New York, 1964), p. 82.

[8]A. J. Nozik and M. Kaplan, Phys. Rev. 159, 273 (1967).

PHYSICAL REVIEW B VOLUME 4, NUMBER 7 1 OCTOBER 1971

Susceptibility of the *s-d* Model

K. D. Schotte*
Service de Physique Théorique CEA-BP No. 2, Gif-sur-Yvette 91, Saclay, France
and
U. Schotte
Institut für Theoretische Physik der Universität Düsseldorf, 4 Düsseldorf, Germany
(Received 19 April 1971)

The analogy between the thermodynamics of the *s-d* model and those of a one-dimensional classical Coulomb gas is exploited to calculate the impurity-spin susceptibility on the computer. The numerical method is a special Monte Carlo procedure first used by Metropolis *et al.* We find a Curie-Weiss form for the static susceptibility with a Néel temperature of about one-third of the Kondo temperature. We discuss the connection between our results and a recent scaling theory of Anderson, Yuval, and Hamann.

I. INTRODUCTION

In this paper we follow a recent development started by Anderson and Yuval.[1] This theory deals with the fluctuation behavior of the impurity spin. The essential result—and our starting point—is an expression for the partition function for the Kondo or *s-d* model[2,3] which turns out to be at the same time the grand partition function of a classical Coulomb gas. The charged particles correspond to spin-up and spin-down flips.

Although this result is not explicit with respect to the physical properties of the *s-d* model, one can study the thermodynamics and the correlation functions of the classical gas. Thereby one obtains information about the dynamics of the spin fluctuations and, for example, the susceptibility of the impurity spin.

The static magnetic susceptibility of dilute magnetic alloys has been the subject of intensive theoretical investigations; the results, however, have remained controversial.[4-6] While at high temperatures a Curie-like dependence characterizes the paramagnetic behavior of the impurities, at low temperatures a degradation to less than Curie dependence seems to be the general result of the calculations based on the *s-d* model for antiferromagnetic coupling. The current theories differ in the degree of this degradation.

The experimental situation too is not very clear.[7] Qualitatively the behavior described above has been observed on a wide range of dilute alloy systems. However, even for comparatively low concentrations, interaction effects seem to be present[8] and complicate the interpretation. Recent experiments where proper attention to this difficulty has been paid confirm the Curie-Weiss behavior above and near T_K (for *Cu*Mn with $\Theta = 10$ mK), while far below T_K a constant plus quadratic term have been measured (for *Au*V with $\Theta = 300$ K).[9]

The problem is how to explain the "disappearance" of the spin towards $T = 0$. Essentially two views have been suggested of the low-temperature be-

havior of the impurity spin embedded in the "sea" of quasifree electrons. Some authors[10,11] say the spin is screened by a cloud of antiferromagnetically coupled electron spins; i.e., the spin is compensated. The formation of this cloud starts at a critical temperature T_K. Below T_K the paramagnetic behavior of the impurity is more and more suppressed because of the surrounding spin cloud of opposite spin direction. A rather different picture[12–14] is the consequence of a spin-fluctuation approach. It suggests that the impurity spin fluctuates rapidly at low temperatures so that the spin is no longer "seen" in experiments which average over a sufficiently long time. These two pictures seem to contradict each other. However, one can show that for a special case both approaches lead to the same result.[15] The special case is the Anderson model with $U=0$ and $E_0=0$, which we will call the resonant-level (RL) model in the following.

This RL model is of special interest in Anderson's classical-gas approach and the subsequent development of a scaling theory.[16] Before outlining the results and predictions of this theory as far as it is relevant in this context, let us restate the basic concepts.

The approximation used to derive the partition function of the s-d model is quite different from those applied before. A good way to get a feeling for it is to replace all conduction-electron operators in the s-d Hamiltonian by density-wave operators in an adaptation of the Tomonaga model.[14] This means that in the neighborhood of the Fermi surface all hole-hole, particle-particle, and particle-hole processes are taken into account. This approximation stresses that the interesting physical effects are due to what happens close to the Fermi surface. Starting with this simplified Hamiltonian, the derivation of the partition function is straightforward. The use of the Tomonaga model is equivalent to the "asymptotic time approximation" applied by Anderson and Yuval[3] and by Hamann in his path-integral theory.[17]

From the view point of the physics of the classical gas, this approximation suggests the importance of long-range interactions. The classical gas describes also the resonant-level model as a limiting case. This has led Anderson *et al.*[16] to develop a scaling theory comparing the RL model with the antiferromagnetic s-d model.

We have studied the properties of the Coulomb gas with the help of the computer. We report here the method and results of the numerical calculation of the low-field spin susceptibility. This is equivalent to calculating the square of the dipole moment of the Coulomb gas. By doing this for gas parameters corresponding to the s-d as well as to the RL model, in a way we test Anderson's scaling theory and the validity of the basic approximations.

In Sec. II we restate our theoretical starting point and discuss the connection between corresponding thermodynamic quantities of the classical gas and the s-d model. We will do the same for the RL model in order to compare the susceptibility as calculated by the computer to the correct one. In Sec. III we will describe our numerical method which is an adaptation of a method of biased random walk by Metropolis *et al.*[18] Finally, the results and their relevance to the scaling theory are discussed.

II. THEORY

The partition function of the s-d model can be obtained by expanding

$$Z = \mathrm{Spur}(e^{-\beta(H_0+H_i)}) \tag{1}$$

with respect to H_i. Here the Hamiltonian is decomposed as

$$H_0 = \sum_{k,\sigma} \epsilon_{k\sigma} n_{k\sigma} + J_Z S_Z \frac{1}{2N} \sum_{k,k'} (c^\dagger_{k+} c_{k'+} - c^\dagger_{k-} c_{k'-}) + g\mu_B H S_z \tag{2}$$

and

$$H_i = J_+ S_+ \frac{1}{N} \sum_{kk'} c^\dagger_{k-} c_{k'+} + \text{c.c.}\ , \tag{3}$$

with the usual notations, except that we distinguish between the different components of the exchange integral. As has been shown,[2,3] the partition function for $S=\frac{1}{2}$ can be written as

$$Z = Z_0 Z_1\ , \tag{4}$$

$$Z_0 = \mathrm{Spur} e^{-\beta H_0}\ , \tag{5}$$

$$Z_1 = \sum_{\substack{n=0,2,\ldots \\ \sigma=\pm}}^{\infty} \left(\frac{J_\pm}{2}\right)^n \int_0^\beta d\tau_n \cdots \int_0^{\tau_2} d\tau_1 P_{n\sigma}(\tau_1,\ldots,\tau_n)\ . \tag{6}$$

Z_1 has the structure of the grand partition function of a classical gas. The Boltzmann factor $P_{n\sigma}$ is

$$P_{n\sigma} = \exp\left(-\beta_g \sum_{\nu>\nu'}^{n} (-1)^{\nu-\nu'+1} v(\tau_\nu - \tau_{\nu'}) - \sigma\mu^* H D_n \right), \tag{7}$$

where

$$\beta_g = 2(1 - J_z \tau) \tag{8}$$

represents the gas temperature; the interaction potential is

$$v(x) = \begin{cases} \ln\left(\dfrac{\sin\pi x/\beta}{\sin\pi\tau/\beta}\right) & \text{for } \tau < x < \beta - \tau \\ 0 & \text{elsewhere}\ . \end{cases} \tag{9}$$

The cutoff τ is of the order of magnitude of the density of states of the conduction electrons at the Fermi surface. We have chosen to avoid the sin-

gularities for short distances by introducing a soft core beyond the cutoff. We will come back to this point below. D_n has the form of the dipole moment

$$D_n = \sum_{\nu=1}^{n} (-1)^{\nu} \tau_\nu - \beta/2 \,. \tag{10}$$

μ^* contains the g factors of the electrons g_e and the impurity spin g:

$$\mu^* = \mu_B (g - g_e J_Z \rho/2) \,. \tag{11}$$

The form of $P_{n\sigma}$ suggests that one deals with particles of positive and negative unit charges distributed alternatingly on a ring of length β. This comes about by performing the trace over S_+ and S_- in the expansion of Z. Thus, spin-up and spin-down flips alternate, and there are equal numbers of each sort. Obviously, one can as well talk about a spin-flip gas.

We will show now how the "electric" susceptibility of the charged gas is computed and how from this one obtains the magnetic susceptibility of the s-d model. Differentiating the free energy of the gas

$$F_g = -\beta_g^{-1} \ln Z_1 \tag{12}$$

with respect to the "electric" field $\mu^* H$ gives the average dipole moment, which, of course, vanishes for zero field. Differentiating twice, one obtains the susceptibility

$$\chi_g = \beta_g^{-1} \langle D^2 \rangle \,. \tag{13}$$

The free energy of the s-d model is

$$\begin{aligned} F_{sd} &= -\beta^{-1} \ln(Z_0 Z_1) \\ &= (\beta_g/\beta) F_g - \beta^{-1} \ln Z_0 \,, \end{aligned} \tag{14}$$

so that up to the Pauli term the susceptibility of the s-d model is

$$\chi_{sd} = (\beta_g/\beta) \chi_g = \beta^{-1} \langle D^2 \rangle \,. \tag{15}$$

We put $\mu^* = 1$ for simplicity. The average is to be understood in the usual classical thermodynamic sense. It means that one has to square the dipole moment D_n for n charges, multiply by the Boltzmann factor, and integrate over all position variables τ_ν. One has to do this for all n, to sum and to divide by Z_1 calculated similarly. This, of course, is an impossible task in practice. Instead we restrict the calculation to a single N. We choose N as the mean particle number in the volume β as given by differentiating the free energy with respect to the chemical potential. This means we use the well-known concept in thermodynamics that for large particle numbers all sorts of ensemble averages should give the same result. In order to derive N we point out that $\frac{1}{2} J_\pm$ must be proportional to the fugacity of the gas:

$$J_\pm \propto e^{\beta g \mu_\pm} \,, \tag{16}$$

where $\mu_\pm$ is the chemical potential. Then with

$$N = N_+ + N_- = 2N_\pm \tag{17}$$

for $J_+ = J_-$, we have

$$N_\pm = -\frac{\partial F_g}{\partial \mu_\pm} = -\beta_g J_\pm \frac{\partial F_g}{\partial J_\pm} \,, \tag{18}$$

and with (14)

$$N_\pm = -\beta J_\pm \frac{\partial F_{sd}}{\partial J_\pm} \,. \tag{19}$$

F_{sd} is unknown, but there is a trick for calculating its derivative. First, by rotational invariance,

$$\frac{\partial F_{sd}}{\partial J_\pm} = \frac{\partial F_{sd}}{\partial J_Z} \tag{20}$$

holds. Using Eq. (14) we have to consider that J_Z appears in Z_0 [see Eqs. (2) and (5)] and also in Z_1 via β_g. As to the Z_1 term, the derivative above with respect to J_Z leads to the average potential energy $\langle V \rangle$, that is, to the grand canonical thermodynamic average of the quantity

$$V_n = \sum_{\nu > \nu'}^{n} (-1)^{\nu - \nu' + 1} v(\tau_\nu - \tau_{\nu'}) \,. \tag{21}$$

For the Z_0 term one can expand

$$F_0 = -\beta^{-1} \ln Z_0 \tag{22}$$

as usual in time-dependent perturbation theory, and calculate it term by term (sum of "unlinked bubbles"). Here also one consistently uses the infinite-time approximation for the electron propagator. We restrict the calculation to lowest order in J_Z as it turns out that higher-order and temperature-dependent terms are negligible in the numerical approach. In these approximations,

$$F_0 = -\tfrac{1}{8} J_Z^2 \int_0^\beta d\tau_2 \int_0^{\tau_2} d\tau_1 \, \mathcal{G}_0^2(\tau_2 - \tau_1) \,, \tag{23}$$

$$\mathcal{G}_0(t) = \frac{\sin \pi \tau/\beta}{\sin \pi t/\beta} \quad \text{for } \tau < t < \beta - \tau \,. \tag{24}$$

Outside this interval,

$$\mathcal{G}_0(t) = 1 \quad \text{for soft core;}$$

if one had chosen to use a hard core beyond the cutoff, one would have

$$\mathcal{G}_0(t) = 0 \quad \text{elsewhere} \,.$$

The result for F_0 depends on this choice:

$$F_0 = -\tfrac{1}{4} J_Z^2 \tau \quad \text{for soft core,} \tag{25}$$

and

$$F_0 = -\tfrac{1}{8} J_Z^2 \tau \quad \text{for hard core.} \tag{26}$$

With (25), using (19), (20), and (17) we finally have, with $J = J_Z = J_\pm$,

$$N = \beta J^2 \tau + 4 J \tau \langle V \rangle \,. \tag{27}$$

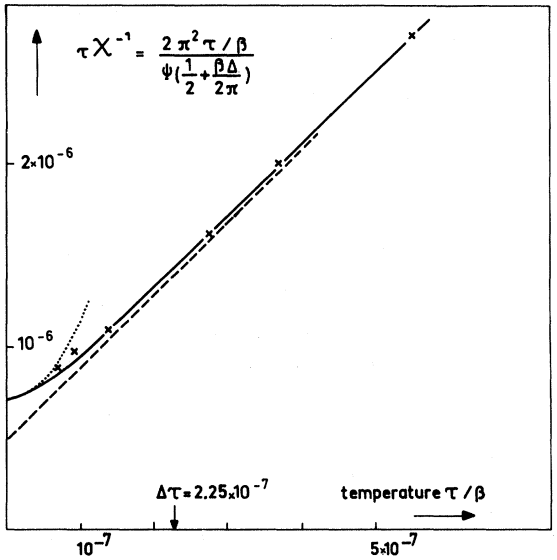

FIG. 1. Inverse susceptibility of the RL model (solid curve), asymptotic behavior for $T \ll 1$ (dotted curve) and for $T \gg 1$ (broken line), and computer results (crosses). The high-temperature asymptotic curve has the same slope as the free-spin Curie law and therefore indicates the vertical scale.

This relation connects the particle number of the classical gas with the temperature of the s-d problem. $\langle V \rangle$ poses the same averaging problem as $\langle D^2 \rangle$. If, however, we calculate $\langle V_N \rangle$ in the canonical ensemble of N particles, then (27) is a self-consistent equation for N. Once N is determined, one can calculate

$$\chi_{sd} = \beta^{-1} \langle D_N^2 \rangle \,. \tag{28}$$

This procedure sounds more complicated than it is, because we found a well-obeyed proportionality between N and $\langle V_N \rangle$. This means that the volume β and the number of particles N it contains are proportional to each other, so that even for rather small N the particle density is constant.

We also want to apply the numerical approach to the RL model in order to check the quality of the numerical procedure and also of the basic approximation by comparing numerical results to the exact ones. For the RL Hamiltonian,

$$H_{\mathrm{RL}} = \sum_k \epsilon_k n_k + \sum_k V_{kd}\, c_k^\dagger c_d + \mathrm{c.c.} \,, \tag{29}$$

the corresponding gas temperature is

$$\beta_g = 1 \,, \tag{30}$$

and the mixing potential $V_{kd} \approx V$ is proportional to the fugacity of the gas. The free energy can be calculated exactly and, therefore, also the relation between N and β. The derivation of this relation and of the susceptibility is given in the Appendix. One finds

$$N = -\beta(2\Delta/\pi)\ln(\Delta\tau) \tag{31}$$

and

$$\chi_{\mathrm{RL}} = \frac{\beta}{2\pi^2}\, \psi'\left(\frac{1}{2} + \frac{\beta\Delta}{2\pi}\right), \tag{32}$$

where $\Delta = \pi V^2 \tau$ and ψ' is the derivative of the digamma function. In Fig. 1 we have plotted χ_{RL}^{-1}. Asymptotically it behaves Curie-Weiss-like for high temperatures:

$$\chi_{\mathrm{RL}}^{-1} \approx 4(T + \Delta/1.85) \,, \tag{32'}$$

and for low temperatures as

$$\chi_{\mathrm{RL}}^{-1} \approx \pi\Delta + \frac{\pi^3 T^2}{3\Delta} \,. \tag{32''}$$

Therefore, the entropy vanishes as $T \to 0$ as it should be. According to the scaling theory, [16] an impurity spin described by H_{sd} should behave qualitatively in the same way as a resonant level described by H_{RL}. Therefore, we expect χ_{sd}^{-1} to obey (32) qualitatively, that is, to have the same analytic form.

Now, before entering the numerical problem, what is to be expected from the behavior of the classical gas and, therefore, of its dipole moment? The particles tend to form pairs or dipoles. For the ferromagnetic case ($\beta_g > 2$), the system is predicted to be completely ordered, so that for all N (or β) D_N is finite and approximately $+\beta/2$ or $-\beta/2$. Therefore, in the limit of large volume β,

$$\langle D^2 \rangle \propto \beta^2 \,, \tag{33}$$

and with (28)

$$\chi_{sd} \propto \beta \,. \tag{34}$$

This means that χ_{sd}^{-1} should show a straight Curie behavior. For $\beta_g < 2$, that is, for the antiferromagnetic case and for the RL model, the system is in a disordered phase. However, it shows domain structure where groups of ordered pairs are separated by single + or − charges. For such a system and for a volume large compared to the mean domain length, the average of the square of the dipole moment should become proportional to the particle number or the volume:

$$\langle D^2 \rangle \propto \beta \,, \tag{35}$$

and, therefore,

$$\chi_{sd} = \mathrm{const} \quad \text{for } \beta \to \infty \,.$$

For smaller β, when the domain length is comparable to the volume, the system appears to be more or less ordered, so that

$$\chi_{sd} \propto \beta \,,$$

as above in the ferromagnetic case.

Evidently, in this theory the degradation of the impurity spin or the change from Curie to Pauli behavior of the susceptibility is the consequence of

$\langle D^2 \rangle$ reaching the thermodynamic limit. If one wants to characterize the regime where this behavior changes by introducing a critical N_K or volume β_K, one expects the latter to be of the order of the inverse Kondo temperature,

$$\beta_K = \tau^{-1} e^{-(J\tau)^{-1}} . \tag{36}$$

Similarly, for the RL model this change of behavior can be characterized by

$$\beta_K \Delta = 1 , \tag{37}$$

or

$$N_K = -(2/\pi)\ln(\Delta\tau) .$$

III. MONTE CARLO METHOD

In order to calculate the thermodynamic averages $\langle D_N^2 \rangle$ and $\langle V_N \rangle$, we use a particular Monte Carlo method devised by Metropolis *et al.*[18] It is frequently used in statistical mechanics to evaluate averaged quantities of the form

$$\langle f \rangle = \int_{\Omega_N} dx\, P(x) f(x) / \int_{\Omega_N} dx\, P(x) , \tag{38}$$

where $P(\tau)$ is an unnormalized probability density as our Boltzmann factor. The method allows one to evaluate averages of this form as

$$\langle f \rangle \approx \frac{1}{M} \sum_{\nu=0}^{M} f(x^\nu) . \tag{39}$$

This is accomplished by starting with an initial configuration $x^0 = \{\tau_1^0, \ldots, \tau_N^0\}$ of N particles. The position of a single particle is then changed randomly to give a new configuration x^1. In our case we have to take care that the charges do not pass each other. Then by comparing $P(x^0)$ and $P(x^1)$, one settles the question of whether the new configuration is a "good" one and if $f(x^1)$ is to be added in (39). In practice one calculates the change of the potential ΔV produced by the change of position of the moved particle. If $\Delta V \leq 0$ or $P(x^1) \geq P(x^0)$, the x^1 is the second configuration and $f(x^1)$ is added in (39). If $\Delta V > 0$, one calculates $\exp(-\beta_g \Delta V)$ and compares this with a random number γ; $0 \leq \gamma < 1$, provided by the computer. If $\exp(-\beta_g \Delta V) > \gamma$, the new configuration is still chosen as the second one, otherwise the initial one is used again and the old $f(x^0)$ is added again in (39).

This procedure is repeated on the second configuration in order to choose the third, etc. As we deal with two kinds of charges which tend to stick together, we also move pairs after having moved single particles. This prevents "freezing" in certain configurations. In the limit $M \to \infty$, after long random walks, a configuration x^ν occurs with frequency $P(x^\nu)$ independent of the initial configuration chosen and independent of the special way of generating new ones.

In a problem like ours the biggest task is to optimize the program. As the potential energy is to be calculated for such an immense number of times, one has to see that ln sin is calculated economically. For this purpose, we devised a linear interpolation scheme between tabulated ln sin values. They get closer spaced in the region of larger variation of ln sin with a scale where use is made of the pseudo-logarithmic notation of floating-point numbers (hexadecimal base, on the computer IBM No. 360/91). Then, 8×10^5 steps of the sort described above take 10 min for $N = 96$. For smaller N, the calculation gets faster; for instance, 2.4×10^6 steps can be done in 10 min for $N = 16$. For each N we had the computer do 8000 steps per particle. One could see that after a few hundred steps the initial configuration was "forgotten." The average $\langle D^2 \rangle$ was calculated between the 1000th and 8000th run per particle.

IV. RESULTS AND DISCUSSION

The numerical results are plotted in Figs. 1–3. Figure 1 shows the inverse susceptibility of the RL model. The solid curve is the exact χ_{RL}^{-1} with $\Delta\tau = 2.25 \times 10^{-7}$. The dots are calculated with the Monte Carlo method for particle numbers between 4 and 48 and Eq. (31) has been used for converting particle numbers into temperature. The reason for this very small $\Delta\tau$ is that we wanted to see the change from Curie to Pauli behavior. Here we have $N_c = 10$. Bigger $\Delta\tau$ involves smaller N_c, but then all reasonable particle numbers, that is, $N > N_c$, would fall into the "Pauli region."

The agreement between the measured and exact χ^{-1} values is quite satisfactory in three respects: (i) One gains confidence in the theory, especially the basic approximation, (ii) the Monte Carlo method is adequate, and (iii) the method works quite well already for small particle numbers N like 6, 8, etc, ...

Figure 1 shows in addition the asymptotic behavior of χ_{RL}^{-1} according to Eqs. (32′) and (32″). In Fig. 2 the computer results for χ_{sd}^{-1} for antiferromagnetic coupling ($\beta_g < 2$) are presented. Calculations were done for $J_1\tau = 0.175$, $J_2\tau = 0.200$, and $J_3\tau = 0.225$. These parameters were chosen for the following reason. If one calculates the critical N_K, inserting (36) into (27) one finds approximately $N_{K1} = 14$, $N_{K2} = 8$, and $N_{K3} = 6$; from (27) they do not come out as even numbers, which, however, they must be. For computer time reasons we did not go beyond $N = 100$. Therefore, for small N_K like $N_K = 6$ one hopes to see the turnoff to Pauli behavior, because then for all $N > 6$ one moves below the Kondo temperature.

For the values chosen for J_2 and J_3 one should cover the region near and above T_K. The solid curves shown were obtained by the method of least-squares fit, that is, we tried to fit the computer results to a straight line as well as a curve of the form of (32),

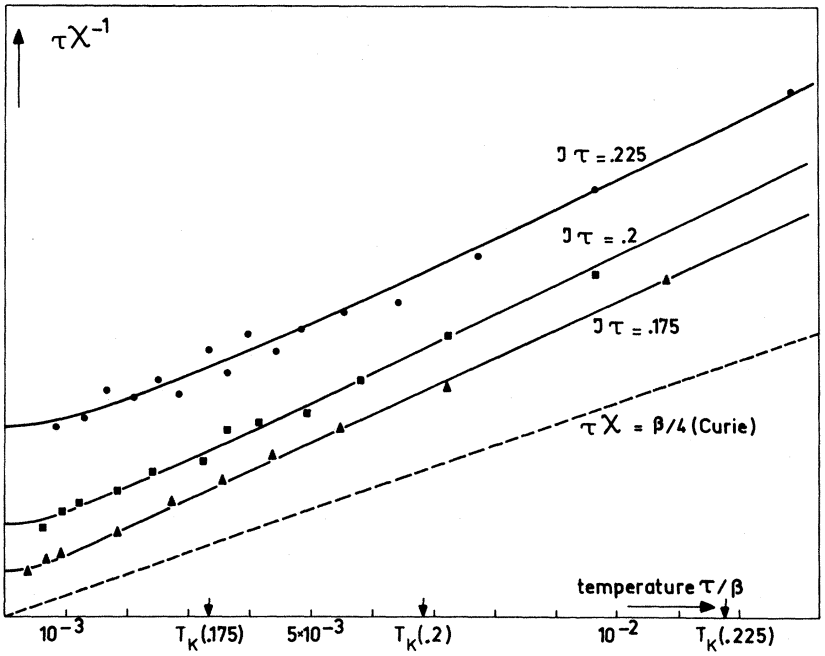

FIG. 2. Inverse susceptibilities of the s-d model; computer results for $J\tau = 0.225$; $J\tau = 0.200$; $J\tau = 0.175$. The free-spin Curie curve (solid line) provides the vertical scale.

$$\chi_{sd}^{-1} = \frac{2\pi^2\delta}{\beta} \Big/ \psi'\left(\frac{1}{2} + \frac{\Delta\beta}{2\pi}\right). \tag{40}$$

For J_1, (40) gave a better fit than a simple straight line; for J_2 and J_3 the straight fit and (40) are of the same quality. The parameters δ and Δ are supposed to indicate the modification of the effective magnetic moment and the Néel temperature—from (32′) it should be about $\Delta/2$—respectively. In Table I we listed Δ, δ, and T_k for comparison for the three cases of J we studied.

The relatively large scatter of the dots for J_3 has probably to do with the tendency of the particles to build up domains of dipoles which move about in the volume and fluctuate in size. Therefore, the convergence of the Monte Carlo method is poorer below than above T_K, where for smaller volume the system looks more or less ordered. Very much below T_K (or for very large N) these fluctuations are expected to average out so that the convergence im-

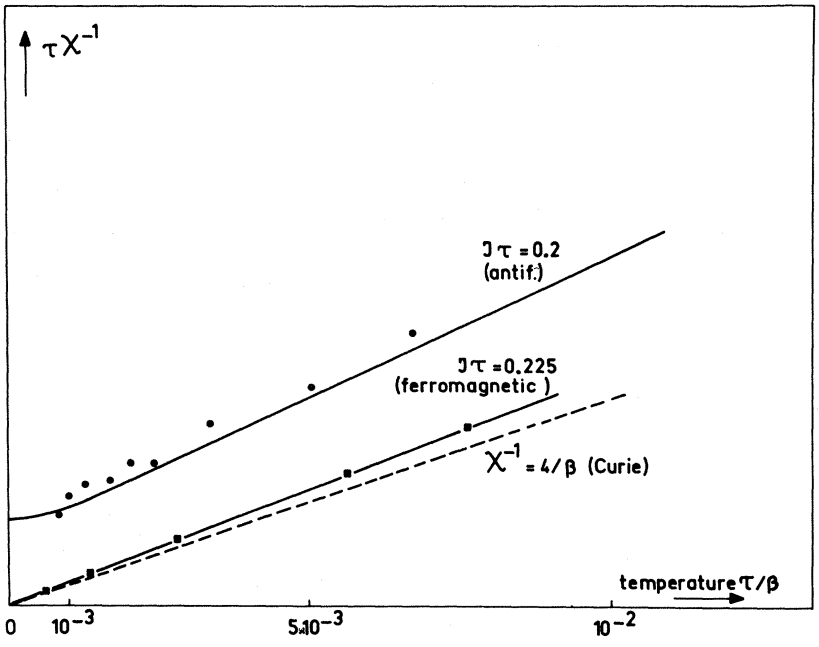

FIG. 3. Upper curve: machine calculations based on a hard-core cutoff compared to soft-core results for $J\tau = 0.200$ (solid line); lower curve: machine results for ferromagnetic s-d coupling for $\mu^* = 1$; solid line; free-spin Curie law.

proves. Obviously, $N=100$ is not yet large enough.

Our results, of course, do not confirm the scaling theory unambiguously. Especially the region where χ_{sd}^{-1} is predicted to behave as

$$\chi_{sd}^{-1}=a+bT^2$$

is very close to the vertical axis—closer than we could go.

To study the region near $T=0$ more closely, one could think of the trick of choosing a very large $J\tau$ with a small N_K for an "enlarged picture" of the Pauli region. This, however, is impossible, because the classical gas gets too dense, that is, N/β gets too large. If the mean distance of the particles becomes comparable to the cutoff τ, the asymptotic time approximation will certainly break down because the short-time (or short-distance) behavior becomes crucial. Here, for $J_1\tau$ we have for the mean-particle distance $\beta/N=12\tau$, which we felt is as far as one can go.

The results show convincingly that χ_{sd} is finite at $T=0$, which is here the consequence of the dipole moment saturating in the thermodynamic limit, $N\to\infty$. If one extrapolates the curves one finds Néel temperatures of about $\frac{1}{3}T_K$.

We also checked into the importance of how one treats the cutoff region of the interparticle potential. All calculations for Fig. 2 were done using a soft core as in Eq. (9). We repeated the calculations for $J\tau=0.200$ introducing a hard core, that is, $v(x)=\infty$ (or, on the computer, a very large number), for $x<\tau$ and $x>\tau-\beta$.

Instead of (27) one has then to use

$$N=\tfrac{1}{2}\beta J^2\tau+4J\tau\langle V_N\rangle \tag{41}$$

for a particle-number–versus–temperature scale. The results are plotted in Fig. 3; the solid line is from Fig. 2 for $J\tau=0.200$. The computer's dots are a little higher than the "soft-core curve," therefore, Δ changes a little, but the results are the same in principle.

This confirms Anderson's argument[16] that though the use of the cutoff is extremely important, the details of its structure are not. So the result establishes self-consistently the validity of the "asymptotic time approximation," or of the Tomonaga model, as long as one studies a dilute classical gas, as indicated above.

In Fig. 3 we also plotted computer results for the case of a ferromagnetic exchange integral $\beta_g>2$. A rather extreme value of $|J\tau|=0.225$ ($\beta_g=2.45$) was chosen, because for $\beta_g\geq 2$ ordering of the system is predicted, and we wanted to see whether for a rather high density an instability in the order can be noticed. It turns out that the system remains completely ordered, and one finds a straight and an unambiguous Curie line for χ_{ferro}^{-1}. The figure drawn is misleading in that we put $\mu^*=1$ throughout. If one takes account of (11), the curve for χ_{ferro}^{-1} will be flatter than the free-spin Curie curve,

$$\chi_{\text{ferro}}=\beta/3.8\ ,$$

as compared to $\chi_{\text{Curie}}=\frac{1}{4}\beta$ ($g=g_e=2$).

TABLE I. Parameters of the susceptibility (40) and the Kondo temperatures (36) for three different values of antiferromagnetic exchange integral J. δ^{-1} is the square of the effective magnetic moment. μ^* from (11) has not been taken account of; it changes δ^{-1} by a factor of $g^2(1-J\tau)$, if we put $g_e=g$.

	$J^{(1)}\tau=0.225$	$J^{(2)}\tau=0.2$	$J^{(3)}\tau=0.175$
δ^{-1}	0.73	0.72	0.75
$\tau\Delta$	8.1×10^{-3}	3.9×10^{-3}	2.2×10^{-3}
τT_K	11.8×10^{-3}	6.86×10^{-3}	3.33×10^{-3}

V. CONCLUSION

Two qualitative conclusions have emerged from the calculation: (i) that the static magnetic spin susceptibility is finite at $T=0$ and behaves Curie-Weiss-like for high temperatures with a Néel point of about $\frac{1}{3}T_K$; (ii) that using the asymptotic time approximation, the susceptibility of the resonant-level model is reproduced very well and the results for the s-d model are practically independent of the treatment of the short-time behavior. Therefore, this approximation must be supposed to reproduce the physical properties of the s-d model correctly.

Our calculations confirm the scaling theory with respect to the behavior of the classical gas: It is completely ordered for ferromagnetic s-d coupling and has a domain structure of rather wild behavior for low temperature in the antiferromagnetic case. The susceptibility (or mean-square dipole moment), however, behaves quite undramatically. Especially in the region below the Kondo temperature, the accuracy of the Monte Carlo method seems limited but sufficient for qualitative purposes.

ACKNOWLEDGMENTS

The authors are grateful to Professor G. Eilenberger who has given them the opportunity to use the computer facility of the Kernforschungsanlage Jülich in the earlier stage of the calculations. One of the authors (D. S.) expresses his thanks to the Service de Physique Théorique of the C. E. N. Saclay for their hospitality. The Saclay computer has been used for most of the calculations; special thanks are due to Dr. Levesque and Dr. Renardy for advice with the numerical methods.

APPENDIX: PARTICLE NUMBER AND SUSCEPTIBILITY FOR THE RL HAMILTONIAN

Particle Number

As mentioned above, the RL model is a special

case for the classical-gas approach. All its properties can be calculated directly from the RL Hamiltonian.

With respect to the classical gas, the mixing potential is connected with the fugacity by

$$V_{\pm} \propto e^{\beta_g \mu_{\pm}} , \tag{A1}$$

where

$$V_{+} \equiv V_{kd} , \quad V_{-} \equiv V_{kd}^{*} = V_{dk} . \tag{A2}$$

Therefore, we have from (18)

$$N_{\pm} = -\frac{\partial F_g}{\partial \mu_{\pm}} = -\beta_g V_{\pm} \frac{\partial F_g}{\partial V_{\pm}} . \tag{A3}$$

The free energies of the classical gas and the RL model are connected as in (12) and (14):

$$F_{\mathrm{RL}} = -\beta^{-1} (\ln Z_0 + \ln Z_1) , \tag{A4}$$

where Z_0 refers to the kinetic energy alone, and

$$F_g = -\beta_g^{-1} \ln Z_1 , \tag{A5}$$

so that

$$N_{\pm} = -\beta V_{\pm} \frac{\partial F_{\mathrm{RL}}}{\partial V_{\pm}} . \tag{A6}$$

If we write the RL Hamiltonian as

$$\begin{aligned} H_{\mathrm{RL}} &= E_{\mathrm{kin}} + \tilde{V} , \\ \tilde{V} &= V_{+} \textstyle\sum_k c_k^{\dagger} c_d + V_{-} \sum_k c_d^{\dagger} c_k , \end{aligned} \tag{A7}$$

one has

$$\begin{aligned} N = N_{+} + N_{-} &= -\beta \left(V_{+} \frac{\partial F_A}{\partial V_{+}} + V_{-} \frac{\partial F_A}{\partial V_{-}} \right) \\ &= \beta \,\mathrm{Spur}[\, \tilde{V} e^{-\beta (E_{\mathrm{kin}} + \tilde{V})}] [\mathrm{Spur} e^{-\beta (E_{\mathrm{kin}} + \tilde{V})}]^{-1} , \end{aligned} \tag{A8}$$

so that finally

$$N = -\beta \langle V \textstyle\sum_k (c_k^{\dagger} c_d + c_d^{\dagger} c_k) \rangle , \tag{A9}$$

where we put $V_{+} = V_{-} = V$.

This expectation value is evaluated in the usual way with the help of the Green's function

$$G_{kd}(\omega^{+}) = \frac{1}{\omega - \epsilon_k} \quad \frac{V}{\omega + i\Delta} . \tag{A10}$$

Here, and in the following, use is made of the approximation

$$V^2 \textstyle\sum_k (\omega - \epsilon_k + i\eta)^{-1} \approx -i\Delta = -i V^2 \pi \tau . \tag{A11}$$

Then the usual technique gives

$$N = -\frac{\beta \Delta}{\pi} \int_{-D}^{D} \frac{2\omega}{\omega^2 + \Delta^2} f(\omega) d\omega , \tag{A12}$$

where a band cutoff $D = \tau^{-1}$ has to be introduced because of the approximation (A11), and $f(\omega)$ is the Fermi function.

The easiest way to evaluate this integral of the form

$$J = \int_{-D}^{D} \alpha(\omega) f(\omega) d\omega$$

is by decomposing it as

$$J = \int_{-D}^{D} \alpha(\omega) \Theta(-\omega) d\omega + \int_{-D}^{D} \alpha(\omega) [f(\omega) - \Theta(-\omega)] d\omega , \tag{A13}$$

where Θ is the step function. The first integral can easily be done, the second has now a convergent integrand so that one can evaluate it by contour integration methods with $D \to \infty$. The result is

$$N = -\frac{2}{\pi} \Delta\beta \ln(\Delta\tau) - 4 \frac{\beta\Delta}{2\pi} \left[\Psi\left(\frac{1}{2} + \frac{\beta\Delta}{2\pi} \right) - \ln \frac{\beta\Delta}{2\pi} \right] . \tag{A14}$$

We checked that the second term is always smaller than 0.316, so for the numerical produce it is sufficient to use

$$N = -\frac{2}{\pi} \Delta\beta \ln(\Delta\tau) . \tag{31}$$

Susceptibility

As has been shown before,[2] the $c_d^{\dagger}$ and c_d operators act like Pauli operators or like S^{+} and S^{-}. Therefore, the term which corresponds to the Zeeman term in the s-d Hamiltonian,

$$\mu_B H S_Z = \mu_B H \tfrac{1}{2} (S^{+} S^{-} - S^{-} S^{+}) , \tag{A15}$$

has the form

$$\mu_B H \tfrac{1}{2} (c_d^{\dagger} c_d - c_d c_d^{\dagger}) = \mu_B H (n_d - \tfrac{1}{2}) \tag{A16}$$

for the RL model.

The susceptibility

$$\chi_{\mathrm{RL}} = \left. \frac{\partial \langle n_d - \frac{1}{2} \rangle}{\partial H} \right|_{H=0} \tag{A17}$$

is evaluated using the Green's function G_{dd} for the expectation value,

$$G_{dd}(\omega^{+}) = (\omega - \mu_B H + i\Delta)^{-1} . \tag{A18}$$

Then

$$\begin{aligned} \langle n_d - \tfrac{1}{2} \rangle &= -\tfrac{1}{2} \mu_B \\ &\quad \times \int_{-\infty}^{\infty} d\omega f(\omega) \,\mathrm{Im}[(\omega - \mu_B H + i\Delta)^{-1} - (\omega + i\Delta)^{-1}] \\ &= \frac{\mu_B}{\pi} \,\mathrm{Im} \Psi\left(\frac{1}{2} + \frac{i \mu_B H \beta}{2\pi} + \frac{\Delta\beta}{2\pi} \right) , \end{aligned} \tag{A19}$$

so that finally

$$\chi_{\mathrm{RL}} = \frac{\mu_B^2 \beta}{2\pi} \Psi'\left(\frac{1}{2} + \frac{\Delta\beta}{2\pi} \right) . \tag{32}$$

*On leave of absence from the Institut für Theoretische Physik der Universität Düsseldorf, Düsseldorf, Germany.
[1]P. W. Anderson and G. Yuval, Phys. Rev. Letters 23, 89 (1959).
[2]K. D. Schotte, Z. Physik 230, 99 (1970).
[3]G. Yuval and P. W. Anderson, Phys. Rev. B 1, 1522 (1970).
[4]D. R. Hamann, Phys. Rev. 158, 570 (1967).
[5]J. Zittartz, Z. Physik 217, 155 (1968).
[6]W. Brenig, J. A. Gonzalez, W. Götze, and P. Wölfle, Z. Physik 235, 52 (1970).
[7]J. E. Van Dam and G. J. Van den Berg, Phys. Status Solidi 3, 11 (1970).
[8]For a summing up of this problem, see W. M. Star, thesis (University of Leiden, 1971) (unpublished).
[9]E. C. Hirschkoff, O. G. Symko, and J. C. Wheatley, Phys. Letters 33A, 19 (1970); J. E. Van Dam and P. C. M. Gubbens, *ibid.* 34A, 185 (1971).
[10]Y. Nagaoka, Progr. Theoret. Phys. (Kyoto) 37, 13 (1967).
[11]K. Yosida and H. Ishii, Progr. Theoret. Phys. (Kyoto) 38, 61 (1967).
[12]H. Suhl, Phys. Rev. Letters 19, 442 (1967).
[13]N. Rivier and P. J. Zuckermann, Phys. Rev. Letters 21, 904 (1968).
[14]K. D. Schotte, Z. Physik 235, 155 (1970).
[15]K. D. Schotte (unpublished).
[16]P. W. Anderson, G. Yuval, and D. R. Hamann, Phys. Rev. B 1, 4464 (1970).
[17]D. R. Hamann, Phys. Rev. B 2, 1373 (1970).
[18]N. Metropolis, A. W. Rosenbluth, M. N. Rosenbluth, A. H. Teller, and E. Teller, J. Chem. Phys. 21, 1087 (1953); for a detailed discussion, see the review article by W. W. Wood, in *Physics of Simple Liquids*, edited by H. N. V. Temperley, J. S. Rowlinson, and G. S. Rushbrooke (North-Holland, Amsterdam, 1968).

PHYSICAL REVIEW B VOLUME 4, NUMBER 7 1 OCTOBER 1971

Influence of True Crystallographic Structure on Spin Waves in Ferromagnetic $CrBr_3$†

Jean Sivardière* and Richard Silberglitt‡
Brookhaven National Laboratory, Upton, New York 11973
(Received 6 May 1971)

Previous spin-wave-renormalization calculations, even those including correlation effects, have been unable to explain correctly both the temperature and field dependence of the magnetization of ferromagnetic $CrBr_3$. However, these calculations were based on a simplified model lattice, which has been shown by recent neutron-diffraction experiments to be only qualitatively correct. In the following, we present a first-order renormalized spin-wave theory which takes into account the true crystallographic structure of $CrBr_3$. This theory explains simultaneously the temperature and field dependence of the magnetization and the magnon dispersion curves. Although the simplified model contains only two exchange parameters, while the exact model contains five, we find that once the true crystallographic structure is introduced, a two-parameter model can explain $M(T, H)$ and the low-energy part of the magnon spectrum. We conclude that the previous discrepancies between theory and experiment were not a result of any failure of spin-wave theory or of the presence of too few exchange parameters, but were due simply to a misrepresentation of the crystal structure.

I. INTRODUCTION

$CrBr_3$ is a rhombohedral insulating ferromagnet with $T_c = 32.5\,^\circ$K. Because of its weak layered structure, this system is particularly useful for testing the validity of spin-wave renormalization calculations.[1]

Nuclear-magnetic-resonance (NMR) measurements[2-5] have furnished a very accurate determination of the magnetization $M(T, H)$ versus temperature and field. These results were explained by means of various spin-wave theories. Because of the complicated crystallographic structure of $CrBr_3$, a simplified model was introduced by Gossard, Jaccarino, and Remeika,[2] involving only two exchange parameters: J_L between nearest neighbors in adjacent planes and J_T between nearest neighbors in the same plane. From their measurement of $M(T, 0)$ versus T, Davis and Narath[3] calculated $J_T = 0.497 \pm 0.013\,^\circ$K and $J_L = 8.25 \mp 0.10\,^\circ$K using a self-consistent first-order renormalized spin-wave theory. However, as has been recently shown,[5] while this set of values of the two exchange parameters gives a very good fit to the magnetization $M(T, 0)$ versus T in zero field, it does not explain the magnetization $M(T_0, H)$ versus H at $T_0 = 18\,^\circ$K. In fact, it is shown in Ref. 5 that even a more sophisticated spin-wave theory correct to all orders in the magnon-magnon interaction is unable to explain simultaneously the $M(T, 0)$-vs-T and $M(T_0, H)$-vs-H data. The authors attributed this inability to the inadequacy of the two-parameter model, citing as evidence recent inelastic neutron scattering measurements[6] of the magnon spectrum. Indeed, a five-parameter model (which takes into account the true crystallographic structure) was introduced