Spatial distribution of the magnetic moment in single ground-state systems at $T=0$: From the single impurity case to the finite-concentration case

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A computer simulation of the distribution of the magnetic moment in one, two, and three dimensions was carried out for singlet-singlet systems with a single impurity, both for short-range (nearest-neighbor) and long-range interactions, using a molecular-field-theory approach. The case of a magnetic host ($\mathcal{J}/\Delta > 1$) with a nonmagnetic impurity ($\mathcal{J}/\Delta < 1$), and of a magnetic impurity in a nonmagnetic host at $T=0$ were treated. From the case of the single impurity, and using an extension of the Edwards-Mathon-Wohlfarth model (used in transition-metal alloys) for the case of rare-earth alloys, a Ginzburg-Landau equation was derived from first principles. A solution was thus obtained for the spatial distribution of the magnetic moment, in good agreement with the computer simulation.

I. INTRODUCTION

There has been considerable interest, since the early 1960's, on systems that have a singlet crystal-field ground state. Particularly, systems with two singlet levels have been studied extensively by means of different techniques. These two-singlet-level systems have a singlet ground state, and the only excited state is a singlet at an energy Δ above the ground state. For such systems, all magnetic moments present are of induced type; i.e., they result from polarization or mixing effects of the states for zero magnetic field. The model Hamiltonian considered was the following:

$$H=\sum_i V_{ci}+\sum_{i\neq j}\mathcal{J}_{ij}\vec{J}_i\cdot\vec{J}_j-g\mu_B H_i\sum_i J_{iz} \quad , \tag{1}$$

which has a crystal-field term, an exchange term, and a Zeeman term corresponding to the presence of an applied magnetic field.

In the molecular-field picture, one can think of the molecular field as mixing some of the zero-field singlet excited state into the zero-field singlet ground state to give a polarized ground state with a magnetic moment. But also, the existence of such a polarized ground state is necessary in order to have a molecular field. Therefore, there is a kind of self-consistent condition that says that the ratio of exchange interaction to crystal-field splitting must be greater than some critical value in order to get magnetic ordering even at zero temperature. In 1963, Bleaney[1] looked at this threshold by calculating, in the molecular-field approximation, the susceptibility. The crystal-field-only inverse susceptibility is

$$1/\chi=(\Delta/2g^2\mu_B^2\alpha^2)(1/\tanh\Delta/2T) \quad ,$$

where $\alpha\equiv\langle 0|J_z 1\rangle$ is the matrix element of J_z between the two singlets. The presence of the exchange interaction corresponds to a shift in this expression; i.e., $1/\tanh(\Delta/2T)$ becomes $1/\tanh(\Delta/2T)-A$, where $A=4J(0)\alpha^2/\Delta$, and $\mathcal{J}(k)=\sum\mathcal{J}_{ij}\times\exp[i\vec{k}\cdot(\vec{r}_i-\vec{r}_j)]$ gives the expression of the Fourier transform of the exchange interaction.

For ferromagnetic exchange, the $1/\chi$ vs T curve shifts rigidly downwards until the critical value of the exchange, $A=1$, is reached. At this value, the susceptibility diverges at zero temperature, indicating the onset of magnetic ordering. For values of the exchange greater than the threshold, we have a sudden increase of the ordering temperature, which becomes a linear increase for larger values of the exchange.

Trammell[2] was one of the first to arouse interest in singlet ground-state magnetism in his work published in 1963. Later, Grover[3] derived the excitation spectrum for ferro- and antiferromagnets, and he applied it to Pr^{3+} and Eu^{3+} in cubic and hexagonal environments. The molecular-field theory can be used in calculations involving the full crystal-field level scheme. Such a theory was quite successful in accounting for the behavior of the antiferromagnet $Tb_zY_{1-z}Sb$, in a work of Cooper and Vogt[4] in 1970, in good agreement with experiments. There has been a great deal of work on fcc Pr metal by Bucher *et al.*[5] and Chu *et al.*[6] on the Pr_3Tl compound,[7] which agree over all with the magnetic behavior predicted by molecular-field theory.

A related method of approach to the single-singlet system is the pseudospin representation. It was introduced by Wang and Cooper[8,9] and by Pink.[10] In the paramagnetic regime, the molecular-field states are identical to the crystal-field-only states. In that case, the pseudospin Hamiltonian is formally identical to the Hamiltonian for the Ising problem in a transverse field, where the crystal-field formally takes the role of the applied field. One can then derive the

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equations of motion for the Fourier-transformed pseudospin generating operations $S^+(\vec{k})$ and $S^-(\vec{k})$. The simplest decoupling scheme for the equations of motion is the random-phase approximation (RPA), where at $T=0$ one replaces the S^z by its ground-state expectation value $\langle S^z\rangle$. The RPA takes account of the fact that the true ground state of the system differs from the molecular-field ground state, indicating the admixture of the molecular-field excited state into the true ground state: $\langle S^z\rangle$ measures that difference. The RPA can be generalized to finite temperatures.[11] Within the approximation, one then gets an expression which is self-consistent for the $\langle S^z\rangle$ and thus the magnetization, viz.,

$$\langle J\rangle/\alpha = -2\langle S^z\rangle\left[1-\left[\frac{1}{2\langle S^z\rangle A}\right]^2\right]^{1/2}, \qquad (2)$$

where $\langle J\rangle \equiv$ magnetization, $\alpha=\langle 1|J_z|0\rangle$, and $A=4\mathcal{J}(0)\alpha^2/\Delta$. At finite temperatures, for an exchange slightly larger than the critical value for ferromagnetism at $T=0$, the Curie temperature is equal to 0.1 of the crystal-field splitting. As T/Δ increases from zero, the $\vec{k}=0$ mode energy drops to zero. The rate of decrease is slow until T is near the Curie temperature, where the $\vec{q}=0$ mode frequency goes to zero very rapidly. For $T>T_C$, this mode frequency rises again. From the self-consistent determination of the collective excitations one can also find the temperature dependence of the magnetization. The main feature is the sharp falloff in magnetization near T_C. As T_C/Δ increases, the RPA magnetization shows double value behavior at high T in the ordered regime. This indicates that there is a first-order phase transition at T_C. Wang and Cooper showed that it occurs for $T_C/\Delta>0.1$. The discontinuity in magnetization is very significant for $T_C/\Delta\simeq 1$. As T_C/Δ still decreases further in the RPA, the size of the discontinuity decreases, while the discontinuity in magnetization persists. The magnetization is seen to approach molecular-field behavior.

Elliot and Wood[12] and Pfeuty and Elliott[13] studied in the singlet-singlet system the transition temperature T_C. By assuming a second-order transition and finding the temperature at which the susceptibility diverges, they find a critical value of $A=1.16$ at $T=0$ for a simple cubic lattice with nearest-neighbor exchange. Here, a second-order transition was assumed for $\langle S^z\rangle$ and not for $\langle J\rangle$; i.e., it is the case of an Ising transverse field system and not a physical singlet-singlet problem. Nevertheless, this critical value of A found by Elliott is exactly the same as that found by Wang and Cooper.[9]

Hsieh and Blume[14] and Hsieh[15,16] have studied the excitation spectrum, the susceptibility and the self-consistent magnetization at $T=0$ in a large variety of crystal-field schemes, using a method first introduced by Pink of a two-pseudospins-$\frac{1}{2}$ system; the calculations were carried out in the molecular-field approximation and within the RPA.

On the other hand, a certain amount of experimental evidence has corroborated the theoretical predictions for the singlet ground-state system. Most experiments were connected to understanding collective excitations, i.e., the magnetic exciton. The first neutron scattering experiments in singlet ground-state systems were those of Rainford and Houmann in 1971[17] on single crystal double hcp (dhcp) Pr, and of Holden *et al.*[18] on TbSb. The greatest amount of experimental evidence comes from the study of Pr_3Tl by Birgeneau, Als-Nielsen, and Bucher[19,20] and Birgeneau.[21]

As far as theoretical work in induced moments systems containing impurities is concerned, success has been somewhat restricted. Wang and Shiles[22,23] presented a theory of such a system, both in the paramagnetic phase and for the case of vacancy impurities. The level scheme they treated was that of a singlet ground state and a singlet excited state, for ions in a simple cubic lattice and with only nearest-neighbor exchange interaction. They calculated the energies of the impurity modes, the temperature-dependent self-consistent magnetization at the single impurity site and the local susceptibility. In order to deal with the case of finite concentrations of impurities, they introduce scaling factors c and $(1-c)$ for the impurities and host correspondingly when writing the usual molecular-field expressions. In 1974, Shiles, Taggart, and Tahir-Kheli[24] treated the singlet-singlet system and the singlet-triplet system using a mean-field model for what they call a "two-component system" rather than impure. They were able to show a difference between the magnetization calculated with their model and with simple mean-field theory; but their theory breaks down for large concentrations of impurities and away from the critical region. In 1975, Lebech, McEwen, and Lindgard[25] presented a molecular-field theory for the PrNd and PrTb alloys, which was also worked out independently by the present author,[26] who, considered the first excited states of both Tb and Nd in Pr surroundings, achieving a good fit with the experimental values of Lebech, McEwen, and Lindgard.[25] A generalization of the molecular-field approach was published by Lingard in 1977.[27]

In this paper, we intend to study the behavior of the magnetization in singlet-singlet systems containing impurities, starting from the case of the induced magnetization of a system with a single impurity. Within molecular-field theory, we derive a self-consistent expression for the magnetization at the impurity site or at a pure lattice site. This assumes the form of a Landau expansion in powers of the self-consistent magnetization. The cases of $T=0$ and finite T are considered. A computer simulation was then carried out from that expression of the self-

consistent magnetization to study how the said quantity spreads from the impurity site into the pure lattice sites, as far as the fourth neighbor. The calculation, the details of which will be given below, was carried out using an interation procedure to give self-consistent values for the magnetization at all sites. This was done for the following cases: a one-dimensional chain of atoms, a two-dimensional square lattice, and a three-dimensional crystal (simple cubic and bcc). In all cases we considered the situation of a nonmagnetic impurity in a magnetic host and a magnetic impurity in a nonmagnetic host. The case of a long-range exchange interaction (simulated by a $1/r^3$ exchange) was considered for a square lattice so as to compare those results with the usual nearest-neighbor exchange and thus give a better approximation for the case of the exchange in a real metal. Equivalent results are to be found with this exchange interaction in the case of a three-dimensional crystal. The variation of the magnetization from the impurity site allowed us to consider the extension of the Landau expansion to the continuum limit, for the values of the $\mathcal{J}/\Delta$ ratio which will be specified. This in turn leads us to a Ginzburg-Landau equation for the magnetization. One then finds a Schrödinger-like differential equation for the distribution of the magnetization in space. A theory for the finite concentration case is proposed, using a model similar to that used by Edwards, Mathon, and Wohlfarth[28] for transition-metal alloys, especially Pd-Ni alloys. The results given by the model are then compared to those given by the computer simulation.

II. MOLECULAR-FIELD THEORY OF SINGLET-SINGLET SYSTEMS

We discuss first the case of a pure singlet-singlet system in order to arrive to an expression for the self-consistent magnetization, and then the case will be specified for a single impurity at site $i=0$. We shall use the molecular-field Hamiltonian in the pseudospin formalism, $H_{\rm mf}$, viz.,

$$H_{\rm mf}(i)=\Delta_i S_i^z + S_i^x \sum_j J_{ij}\langle S_j^x\rangle \quad . \tag{3}$$

where Δ_i is the crystal-field splitting of the ith atom, $\mathcal{J}_{ij}$ the exchange interaction between two atoms at sites i and j, and here $\langle S_i^x\rangle$ being the order parameter will lead to the self-consistent expression of the magnetization. We can picture this Hamiltonian in a two-dimensional set of coordinates S_i^x, S_i^z. From this picture, calling $\Delta\equiv\Delta_i$ as it is still the same for all atoms of the pure crystal,

$$\cot\theta=\frac{\Delta}{\sum_j \mathcal{J}_{ij}\langle S_j^x\rangle} \tag{4}$$

and

$$H_{\rm mf}=\left[\Delta^2+\left(\sum_j \mathcal{J}_{ij}\langle S_j^x\rangle\right)^2\right]^{1/2} \quad . \tag{5}$$

Then one has the usual relationship for singlet-singlet systems

$$\langle S_i\rangle_r=\tfrac{1}{2}\tanh(H_{\rm mf}/2kT) \quad , \tag{6}$$

where r is either the x or z component. Using Eq. (5), this is

$$\langle S_i\rangle_r=\tfrac{1}{2}\tanh\left(\frac{[\Delta^2+(\sum_j \mathcal{J}_{ij}\langle S_j^x\rangle)^2]^{1/2}}{2kT}\right) \quad . \tag{7}$$

With this we can write an expression for $\langle S_i^x\rangle$, viz.,

$$\langle S_i^x\rangle=\tfrac{1}{2}\tanh\left(\frac{[\Delta^2+(\sum_j \mathcal{J}_{ij}\langle S_j^x\rangle)^2]^{1/2}}{2kT}\right)\sin\theta \quad . \tag{8}$$

If we take $\langle S_j^x\rangle$ to be small, we can then expand the argument of the tanh, and using the relationship $\tanh(A+B)=(\tanh A+\tanh B)/(1+\tanh A\tanh B)$ with $A=\Delta/2kT$ and

$$B=\frac{1}{2}\frac{(\sum_j \mathcal{J}_{ij}\langle S_j^x\rangle)^2}{(\Delta 2kT)} \quad .$$

As B is small compared to A, $\tanh B\simeq B$, so that $\tanh(A+B)\simeq(\tanh A+B)/(1+B\tanh A)$. Therefore, $\tanh(A+B)\simeq\tanh A+B(1-\tanh^2 A)+$ terms of order B^2. If we substitute this expression into Eq. (8), bearing in mind that in this approximation θ is small and $\sin\theta\simeq\tan\theta\simeq\theta$, and also recalling what A and B stand for, we obtain the self-consistent expression for the magnetization

$$\langle S_i^x\rangle=\sum_j \frac{\mathcal{J}_{ij}\langle S_j^x\rangle}{\Delta}\tfrac{1}{2}\tanh\left(\frac{\Delta}{2kT}\right) + \frac{1}{2}\frac{(\sum_j \mathcal{J}_j\langle S_j^x\rangle)^3}{\Delta^2(2kT)}\left[1-\tanh^2\left(\frac{\Delta}{2kT}\right)\right] \quad . \tag{9}$$

This has the form of an expansion in powers of $\langle S_j^x\rangle$, in which appear only the linear and cubic terms.

For a pure system, $\langle S_i^x\rangle=\langle S_j^x\rangle$ because all sites are equivalent. The ordering temperature is given by the first-order term in Eq. (9); i.e.,

$$\langle S_i^x\rangle=\frac{\sum_j \mathcal{J}_{ij}\langle S_i^x\rangle}{\Delta}\tfrac{1}{2}\tanh(\Delta/2kT) \quad ,$$

which readily gives us the expression for the ordering temperature as

$$\tfrac{1}{2}\tanh\left(\frac{\Delta}{2kT}\right)\sum_j \frac{\mathcal{J}_{ij}}{\Delta}=1 \quad . \tag{10}$$

In we now call $M_i=\langle S_i^x\rangle$, the magnetization at any site i of the pure crystal, we get the self-consistent

expression

$$M_i = M_i \sum_j \frac{\mathcal{J}_{ij}}{\Delta} \frac{1}{2} \tanh(\Delta/2kT) + \frac{1}{2} \frac{(\sum_j \mathcal{J}_{ij})^3 M_i^3}{\Delta^2 (2kT)} [1 - \tanh^2(\Delta/2kT)] \quad . \tag{11}$$

This self-consistent expression for M_i assumes the form of a Landau expansion in powers of the magnetization M_i where only the odd powers of M_i appear (up to the third power of M_i in this approximation).

Let us now consider a crystal with a single impurity. In this case, $\langle S_i^x \rangle \neq \langle S_j^x \rangle$ at the impurity site. If we include this impurity now in our system, we can rewrite the general expession for the magnetization as

$$\langle S_i^x \rangle = \frac{1}{2} \tanh\left(\frac{\Delta_i}{2kT}\right) \sum_j \frac{\mathcal{J}_{ij} \langle S_j^x \rangle}{\Delta_i} + \frac{1}{4} \sum_j \frac{(\mathcal{J}_{ij} \langle S_j^x \rangle)^3}{kT(\Delta_i)^2} \left[1 - \tanh^2\left(\frac{\Delta_i}{2kT}\right)\right] \quad . \tag{12}$$

Here, $\Delta_i = \Delta_0$ for $i = 0$ (impurity site), and $\Delta_i = \Delta_1$ for $i \neq 0$ (host crystal sites). Let us call M_0 the magnetization at the impurity site (site $i = 0$) and M_i, with $i \neq 0$, the magnetization at any pure crystal site, i.e., with $i = 1, \ldots, n$. We will calculate M_0 and see how this magnetization spreads away from the impurity site, as far as the fourth neighbor of that impurity. This will give us an indication of how fast or slow the variation is from the value at the impurity site (and whether a rise or a falloff, according to the case of a magnetic impurity in a nonmagnetic crystal or a nonmagnetic impurity in a magnetic crystal). These results will in turn tell us whether we have to adopt a discrete or a continuum type of solution.

In order to carry out this calculation, we programmed an iteration cycle which will be cut at the fourth neighbor to the impurity site. That is, we will calculate M_0, M_1, M_2, M_3, M_4 moments at the impurity site and first, second, third, and fourth neighbor, respectively; we first assume a value for M_0 (which can be $M_0 = 0.5$, the maximum value that the moment can take), and then, that value is to be used to calculate M_1, M_2, M_3, M_4, and finally a "calculated M_0." We set up a convenient convergence factor, and when the difference between the calculated M_0 and the "assumed M_0" is smaller than the convergence factor, the iteration will stop. We will thus obtain the values of all the moments in a self-consistent way.

The calculation was performed for the case of $T = 0$; although the finite T does not pose any principal problem because it would only involve an extra factor which contains the temperature dependence [see Eq. (6)], and we have preferred to carry out the calculation for $T = 0$ for simplicity. At $T = 0$, our expression for the self-consistent magnetization is the same as Hsieh's,[15,16] viz.,

$$M_i = \frac{H_i}{2(\Delta_i^2 + H_i^2)^{1/2}} \tag{13}$$

with

$$H_i = \sum_j M_j \mathcal{J}_{ij} \quad .$$

As the exchange interaction is a function of the two atoms involved, we have two different types of exchange: between the impurity atom and any of the host crystal atoms, and between any two host atoms. We are left with two parameters mainly in our calculation: $\mathcal{J}/\Delta$ ($\mathcal{J}$ exchange between the impurity atom and any host atom, Δ crystal-field splitting of the impurity), this being $\mathcal{J}/\Delta > 1$ for a magnetic impurity and < 1 for a nonmagnetic or vacancy impurity; and $\mathcal{J}'/\Delta'$ ($\mathcal{J}'$ exchange between any two host atoms and Δ' crystal-field splitting of the host atom), this also being $\mathcal{J}'/\Delta' > 1$ for a magnetic host and < 1 for a nonmagnetic host crystal. We will see that it is also important to specify in each case the ratios $\mathcal{J}/\mathcal{J}'$ and Δ/Δ', i.e., ratios between the two exchange interactions and between the two crystal-field splittings.

A. Linear chain of atoms

We consider here the case of nearest-neighbor interaction only (short range). The effective fields are thus

$$\begin{aligned} H_0 &= 2\mathcal{J}M_1 \quad , \\ H_1 &= \mathcal{J}M_0 + \mathcal{J}'M_2 \quad , \\ H_2 &= \mathcal{J}'(M_1 + M_3) \quad , \\ H_3 &= \mathcal{J}'(M_2 + M_4) \quad , \\ H_4 &= \mathcal{J}'M_3 \quad , \end{aligned} \tag{14}$$

and the expressions for the moments are always given by Eq. (13). The calculation was performed both for the case of a magnetic impurity in a nonmagnetic host and for a nonmagnetic impurity in a magnetic host. The following set of curves [Fig. 1(a)] shows the spread of the magnetization from the impurity site down to its fourth neighbor, for the case of a magnetic impurity in a nonmagnetic host. The curves show the spread of the magnetization for the values of $\mathcal{J}/\Delta$ (impurity) = 1.1, 1.5, 2, and 3, i.e., for increasingly magnetic impurity, as a function of the parameter $\mathcal{J}'/\Delta'$ (host). The magnetic impurity is seen to strongly polarize the first and second neighbors, even for $\mathcal{J}/\Delta$ (impurity) being just above the magnetic condition. For values of $\mathcal{J}'/\Delta'$ from 0.99 to 0.8, and even to 0.7 in the case of a strongly magnetic impurity, there is still an appreciable mo-

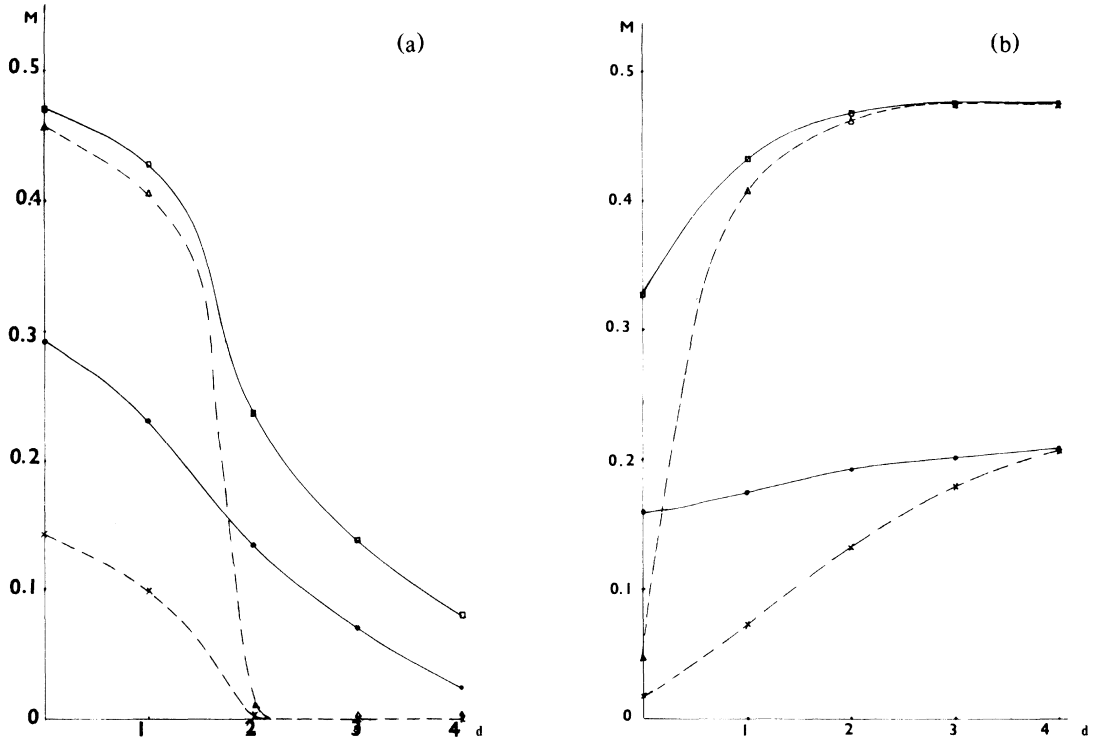

FIG. 1. Singlet-singlet systems, 1 impurity ($T=0$). One-dimensional case. Nearest-neighbor interaction. (a) Magnetic impurity in nonmagnetic host: ● $-\mathcal{J}/\Delta=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=1.1/0.99$; × $-\mathcal{J}/\Delta=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=1.1/0.01$; □ $-\mathcal{J}/\Delta=3$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=3/0.99$; and Δ $-\mathcal{J}/\Delta=3$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}=3/0.01$. (b) Nonmagnetic impurity in magnetic host: ● $-\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}'/\mathcal{J}=1.1/0.99$; × $-\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}'/\mathcal{J}=1.1/0.01$; □ $-\mathcal{J}'/\Delta'=3$, $\Delta/\Delta'=1$, $\mathcal{J}'/\mathcal{J}=3/0.99$; and Δ $-\mathcal{J}'/\Delta'=3$, $\Delta/\Delta'=1$, $\mathcal{J}'/\mathcal{J}=3/0.01$. In all figures, the abcissa d indicates the distance from the impurity (at 0) to the first, second, third, and fourth nearest neighbors (indicated as 1, 2, 3, and 4). M is in bohr magnetons/atom.

ment at all sites. But as $\mathcal{J}'/\Delta'$ decreases further, the decay in the magnetization at the second-, third- and fourth-neighbor sites is very rapid. For $\mathcal{J}'/\Delta'=0.5$ down to values of 0.01, there is only an appreciable moment at the impurity site and its first neighbor due to the magnetic impurity strongly polarizing only that site. This is consistent with our having assumed a short-range interaction. Because of this, the variation of the moments is only slow for $\mathcal{J}'/\Delta'$ between 0.9 and 0.99. Therefore a continuum solution for the moments is more suitable in this region, whereas a discrete solution is more appropriate for values of $\mathcal{J}'/\Delta'<0.9$. We will discuss the continuum solution fully in the last section of the paper.

The next set of graphs [Fig. 1(b)] shows the moment distribution from the impurity site down to its fourth neighbor, for the case of a nonmagnetic impurity in a magnetic host crystal. The curves show the spread of the magnetization for the values of $\mathcal{J}'/\Delta'=1.1$, 1.5, 2, and 3, i.e., for an increasingly magnetic host, as a function of $\mathcal{J}/\Delta$. We can see that the magnetization increases as one moves away from the impurity site towards host atoms. As the host becomes increasingly magnetic, the nonmagnetic impurity tends to have less and less influence on the bulk value (taken here as 0.5, i.e., the maximum value that the moment can take in a singlet-singlet system). While for $\mathcal{J}'/\Delta'=1.1$ the depolarizing effect of the impurity is felt by all neighbors, for values of $\mathcal{J}'/\Delta'$ corresponding to a strongly magnetic host the effect of the impurity is very localized. Indeed, only for $\mathcal{J}/\Delta$ near the magnetic condition is the effect felt by all other neighbors. As $\mathcal{J}/\Delta$ decreases even further from the value 1, i.e., the magnetic condition, the hole created in the lattice becomes more localized. We can see that the effect of the vacancy is then only felt by the first neighbor. This is consistent with our assumption of a short-range exchange interaction.

B. Case of a square lattice

We now study the case of a two-dimensional square lattice. The study will be done again for a single impurity, either magnetic or nonmagnetic, in a host that is, respectively, nonmagnetic and magnetic. The calculation of the moments was performed for

$T=0$; the finite-temperature case introduces further mathematical difficulties though the fundamental physics is retained. The computer calculation follows the same procedure as that used for the linear chain. One then obtains self-consistent values for all the moments. The equations that give us the effective fields acting on the impurity and host atoms are the following:

$$\begin{aligned} H_0 &= 4\mathcal{J}M_1 \ , \\ H_1 &= \mathcal{J}M_0 + \mathcal{J}'(2M_2 + M_3) \ , \\ H_2 &= 2\mathcal{J}'(M_1 + M_4) \ , \\ H_3 &= \mathcal{J}'(M_1 + 2M_4) \ , \\ H_4 &= \mathcal{J}'(M_2 + M_3) \ . \end{aligned} \tag{15}$$

The moments are given by Eq. (13), with $\Delta_i = \Delta$ for $i=0$ and $\Delta_i = \Delta'$ for $i=1,\ldots,4$. Equation (15) corresponds to the nearest-neighbor (NN) exchange interaction.

The calculation was carried out for two different physical situations: (i) the case of an insulator, simulated by a short-range exchange interaction between an atom and its NN shell, (ii) the case of a metal, this being simulated by a long-range exchange interaction proportional to $1/r^3$ between an atom and its neighbors, as far as the fourth neighbor. We thus obtain the moment distribution at the impurity site and at its first, second, third, and fourth neighbors.

For the case of a magnetic impurity in a nonmagnetic host, we obtain a falloff of the magnetic moment as one goes away from the impurity towards outer shells of atoms. In the case of a nonmagnetic impurity in a magnetic host, we see an increase of the moment from its value at the impurity site up to the maximum value it can reach, the bulk value being $M=\frac{1}{2}$. For all the situations described above, a discussion of the values of the moment distribution is given not only in terms of the $\mathcal{J}/\Delta$ and $\mathcal{J}'/\Delta'$ parameters but also the ratios $\mathcal{J}/\mathcal{J}'$ and Δ/Δ'. It will be shown that these two ratios must be taken into account in order to explain the different behaviors in some of the curves. Figure 2(a) shows the decrease of the moment from its value at the impurity site, as one goes away from the impurity towards the atoms of outer shells. For $\mathcal{J}'/\Delta'$ small (i.e., < 0.5), the magnetic impurity effect will be felt mainly by the nearest-neighbor atom, which is consistent with our having imposed a short-range interaction. In that region, the curves show a sharp decrease of the moments for atoms further away from the impurity. This has the effect of depolarizing the impurity, thereby reducing its moment and thus the moment at its nearest neighbor.

As $\mathcal{J}/\Delta$ is increased, the impurity moment increases and its variation becomes almost negligible as one reduces $\mathcal{J}'/\Delta'$. This shows that, as the impurity becomes more strongly magnetic, the surrounding "vacancies" have less and less of an effect on it. The moments at the outer atoms will rapidly fall off as the impurity will only polarize its nearest neighbor in an appreciable way. For large values of $\mathcal{J}'/\Delta'$, we can see that all moments still have an appreciable value; this is increasingly so as $\mathcal{J}'/\Delta'$ approaches the magnetic condition, i.e., the value 1. The outstanding feature is that a single impurity with a $\mathcal{J}/\Delta$ just above the magnetic condition produces moments at all sites for values of $\mathcal{J}'/\Delta'$ just below the critical ratio 1. In this region, the host atoms will not produce an important depolarization of the impurity atom, so that the effect of the magnetic impurity will be felt by all atoms to the fourth neighbor. Thus, in this region, we have to give a continuum solution to the problem, which will be given later when we discuss the Ginzburg-Landau equation for the moment distribution.

Let us now analyze the results of the special cases in which we take the different possible combinations of $\mathcal{J}/\mathcal{J}'$, Δ/Δ' which still allows us to have the same values for $\mathcal{J}/\Delta$ and for $\mathcal{J}'/\Delta'$ as those discussed in the previous case. This is shown in the next set of curves in Figs. 2(b) and 2(c).

For the case of $\Delta/\Delta'=2$, i.e., the impurity atom having a greater crystal-field splitting between the two singlet levels than that of the host atoms, we must also increase by the same factor the parameter $\mathcal{J}$ so that $\mathcal{J}/\Delta$ remains unchanged. The main effect is that of the enhancement of the impurity and first-neighbor moments, for all values of $\mathcal{J}'/\Delta'$. For values of the latter near 1, we can see an interesting effect on the nearest neighbor to the impurity in Fig. 2(b). As $\mathcal{J}'/\Delta'$ increases, M_1 grows rapidly and can even become greater than M_0, the moment at the impurity site. Looking at the diagram of the square lattice, we can see that the impurity atom has four nearest neighbors, labeled 1. The first-nearest neighbor has in turn, as nearest neighbors, the impurity atom, two atoms of the second shell labeled 2, and one atom of the third shell labeled 3. Therefore, H_0, the effective field acting on the impurity, can be larger than H_1, the effective field on the first-neighbor atom. Equation (13) gives the value of the magnetic moment at any site i in the case of M_0, the denominator will be larger than the denominator for the corresponding expression for M_1, as $\Delta'=2\Delta$ and $H_0 > H_1$. Thus, for sufficiently large values of M_2 and M_3, i.e., for values of $\mathcal{J}'/\Delta'$ near the magnetic condition, M_1 can be larger than M_0. This will apply to a very narrow range of values of $\mathcal{J}'/\Delta'$, since M_2 and M_3 decrease rather rapidly.

If we increase the ratio $\mathcal{J}/\mathcal{J}'$ keeping $\Delta/\Delta'=2$, i.e., the impurity is now strongly magnetic and the enhancement of M_0 and M_1 is even more dramatic: this can be clearly seen in Fig. 2(b). In this case, M_1 cannot be larger than M_0 for values of $\mathcal{J}'/\Delta'$ near 1 as the effective field H_0 is now very much greater

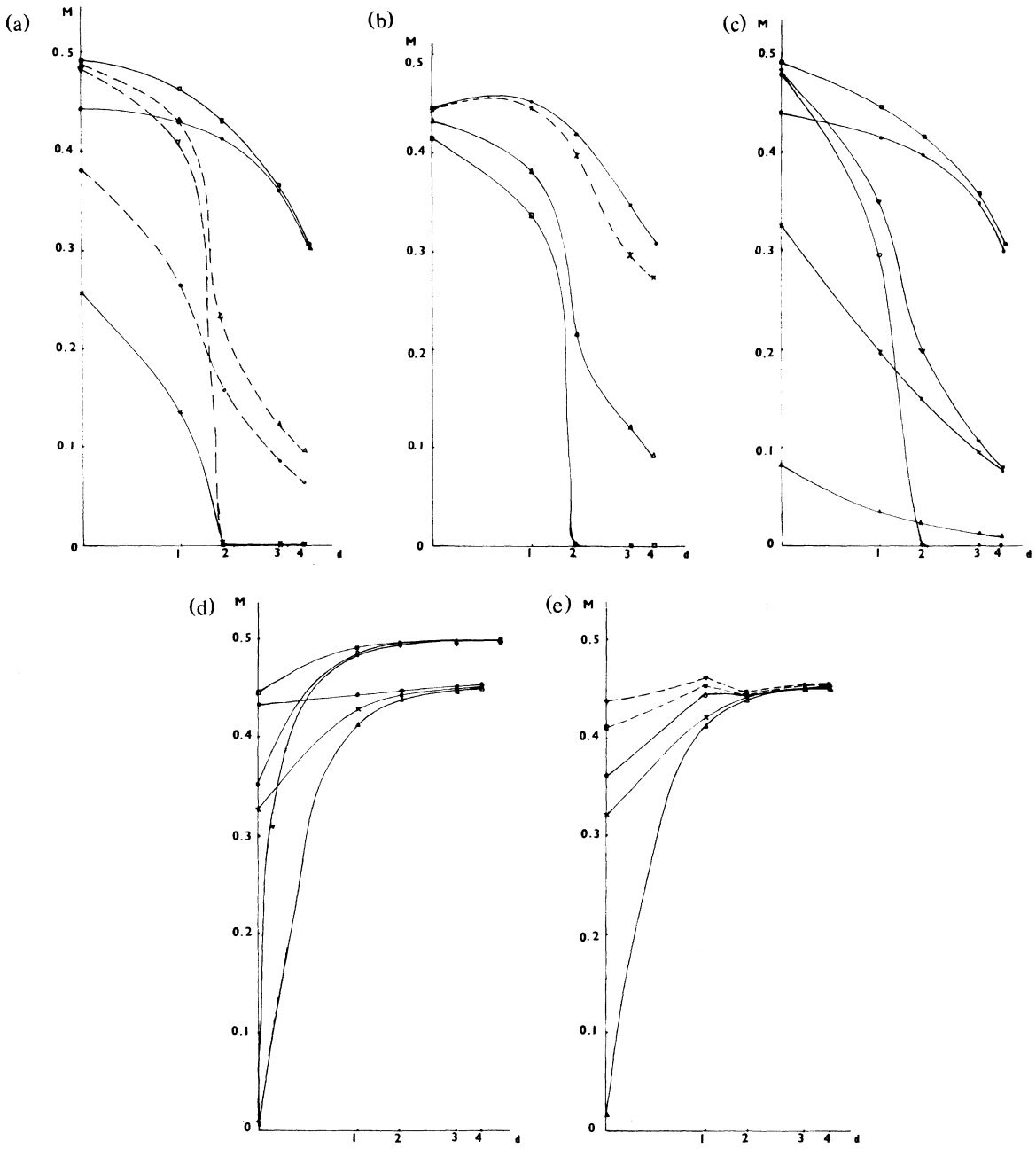

FIG. 2. Singlet-singlet systems, 1 impurity ($T=0$). Two-dimensional case. Nearest-neighbor interaction. (a) Magnetic impurity in nonmagnetic host: ●$-\mathcal{J}/\Delta=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=1.1/0.99$; ○$-\mathcal{J}/\Delta=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=1.1/0.5$; ×$-\mathcal{J}/\Delta=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=1.1/0.01$; Δ$-\mathcal{J}/\Delta=3$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=3/0.99$; □$-\mathcal{J}/\Delta=3$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=3/0.5$; and ▽$-\mathcal{J}/\Delta=3$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=3/0.01$. (b) Magnetic impurity in nonmagnetic host: ●$-\mathcal{J}/\Delta=1.1$, $\mathcal{J}/\mathcal{J}'=2.2/0.99$, $\Delta/\Delta'=2$; ×$-\mathcal{J}/\Delta=1.1$, $\mathcal{J}/\mathcal{J}'=2.2/0.9$, $\Delta/\Delta'=2$; Δ$-\mathcal{J}/\Delta=1.1$, $\mathcal{J}/\mathcal{J}'=2.2/0.5$, $\Delta/\Delta'=2$; and □$-\mathcal{J}/\Delta=1.1$, $\mathcal{J}/\mathcal{J}'=2.2/0.01$, $\Delta/\Delta'=2$. (c) Magnetic impurity in nonmagnetic host: ●$-\mathcal{J}/\Delta=1.1$, $\Delta/\Delta'=0.5/1$, $\mathcal{J}/\mathcal{J}'=0.55/0.99$; ×$-\mathcal{J}/\Delta=1.1$, $\Delta/\Delta'=0.5/1$, $\mathcal{J}/\mathcal{J}'=0.55/0.6$; Δ$-\mathcal{J}/\Delta=1.1$, $\Delta/\Delta'=0.5/1$, $\mathcal{J}/\mathcal{J}'=0.55/0.5$; □$-\mathcal{J}/\Delta=3$, $\Delta/\Delta'=0.5/1$, $\mathcal{J}/\mathcal{J}'=1.5/0.99$; ▽$-\mathcal{J}/\Delta=3$, $\Delta/\Delta'=0.5/1$, $\mathcal{J}/\mathcal{J}'=1.5/0.5$; and ○$-\mathcal{J}/\Delta=3$, $\Delta/\Delta'=0.5/1$, $\mathcal{J}/\mathcal{J}'=1.5/0.01$. (d) Nonmagnetic impurity in magnetic host: ●$-\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}'/\mathcal{J}=1.1/0.99$; ×$-\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}'/\mathcal{J}=1.1/0.5$; Δ$-\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}'/\mathcal{J}=1.1/0.01$; □$-\mathcal{J}'/\Delta'=3$, $\Delta/\Delta'=1$, $\mathcal{J}'/\mathcal{J}=3/0.99$; ○$-\mathcal{J}'/\Delta'=3$, $\Delta/\Delta'=1$, $\mathcal{J}'/\mathcal{J}=3/0.5$; and ▽$-\mathcal{J}'/\Delta'=3$, $\Delta/\Delta'=1$, $\mathcal{J}'/\mathcal{J}=3/0.01$. (e) Nonmagnetic impurity in magnetic host: ▽$-\mathcal{J}'/\Delta'=1.1$, $\Delta'/\Delta=2$, $\mathcal{J}'/\mathcal{J}=2.2/0.99$; □$-\mathcal{J}'/\Delta'=1.1$, $\Delta'/\Delta=2$, $\mathcal{J}'/\mathcal{J}=2.2/0.8$; ○$-\mathcal{J}'/\Delta'=1.1$, $\Delta'/\Delta=2$, $\mathcal{J}'/\mathcal{J}=2.2/0.6$; ×$-\mathcal{J}'/\Delta'=1.1$, $\Delta'/\Delta=2$, $\mathcal{J}'/\mathcal{J}=2.2/0.5$; and Δ$-\mathcal{J}'/\Delta'=1.1$; $\Delta'/\Delta=2$, $\mathcal{J}'/\mathcal{J}=2.2/0.01$.

than H_1, and although M_2 and M_3 have still appreciable values in that region, the value of M_1 cannot be greater than M_0. The fact that the impurity has such a strong moment will compensate the anomaly that was to be observed in Fig. 2(b).

If we now reduce $\mathcal{J}$ and Δ by $\frac{1}{2}$, thus leaving the ratio $\mathcal{J}/\Delta$ unchanged, we see another interesting effect for $\mathcal{J}/\Delta$ just above the magnetic condition, i.e., for a not too strong magnetic impurity in [Fig. 2(c)]. The curve shows the abrupt falloff of all the moments, even the impurity moment, below a certain value of $\mathcal{J}'/\Delta'$. Although $\mathcal{J}/\Delta > 1$, there exists a region for which $\mathcal{J} < \mathcal{J}'$, i.e., the region of large values of $\mathcal{J}'/\Delta'$. This results in a progressive depolarization of the impurity atom, whose moment decreases as $\mathcal{J}'/\Delta'$ decreases. We can establish the condition for all moments to go to zero by means of an expansion, for small values of the M_i's, of Eqs. (13) and (15); to first order in $\mathcal{J}/\Delta$ or $\mathcal{J}'/\Delta'$ we get

$$\begin{aligned} M_4 &= \frac{\mathcal{J}'}{2\Delta'}(M_2+M_3) \ , \\ M_3 &= \frac{\mathcal{J}'}{2\Delta'}M_1 \ , \\ M_2 &= \frac{\mathcal{J}'}{2\Delta'}M_1 \ , \\ M_0 &= \frac{2\mathcal{J}}{\Delta}M_1 \ , \end{aligned} \tag{16}$$

but

$$M_1 = \frac{\mathcal{J}M_0}{2\Delta} + \frac{\mathcal{J}'(2M_2+M_3)}{2\Delta'} \ .$$

Therefore, if we substitute the expressions (16) into the equation for M_1, we get the condition

$$\frac{\mathcal{J}^2}{\Delta\Delta'} + \frac{5}{4}\frac{\mathcal{J}'^2}{\Delta'^2} = 1 \ . \tag{17}$$

For a given $\mathcal{J}/\Delta$ and a given Δ/Δ' we can calculate for what value of $\mathcal{J}'/\Delta'$ all moments go the zero. There will be a certain value of $\mathcal{J}'/\Delta'$ for which $\mathcal{J}$ becomes greater that $\mathcal{J}'$. At this point, the remaining magnetization on the impurity atom will decrease rapidly as the host becomes strongly nonmagnetic, until it vanishes according to Eq. (17). We can give an upper limit for this particular situation to occur, i.e., there will be a value of $\mathcal{J}/\Delta$ above which the moments will not fall to zero. Indeed, this further condition is

$$1 - \frac{\mathcal{J}^2}{\Delta\Delta'} > 0 \ ,$$

otherwise there cannot be a solution for Eq. (17). In the case of Fig. 2(c), this becomes $\mathcal{J}^2/\Delta^2 < 2$, or $\mathcal{J}/\Delta < \sqrt{2}$. This means that for $\mathcal{J}/\Delta > \sqrt{2}$ this behavior of the moments will cease.

This is clearly illustrated by Fig. 2(c), for $\mathcal{J}/\Delta = 3$, with $\mathcal{J} = 1.5$ and $\Delta = \frac{1}{2}\Delta$. The impurity is now strongly magnetic and M_0 and M_1 still have finite nonzero values for small values of $\mathcal{J}'\Delta'$. If we compare this curve to that which had the same $\mathcal{J}/\Delta$ but instead a value of $\mathcal{J}$ double than the one it has now, i.e., with Fig. 2(b), the first neighbor feels a weaker effect from the magnetic impurity than before, because $\mathcal{J}$ is now lower.

Let us now turn to the case of a nonmagnetic impurity in a magnetic host, always assuming a short-range exchange imteraction between any pair of atoms in a square lattice. The equations giving the effective fields at each site i are similar to Eq. (15); here, we have assumed that the atoms outside the fourth-neighbor shell have a moment equal to $\frac{1}{2}$, i.e., the bulk value. We thus expect the moments to increase from the value at the impurity site towards the maximum value $\frac{1}{2}$ as one moves towards the outer shells of atoms. The system of equations (15) now becomes

$$\begin{aligned} H_0 &= 4\mathcal{J}M_1 \ , \\ H_1 &= \mathcal{J}M_0 + \mathcal{J}'(2M_2+M_3) \ , \\ H_2 &= 2\mathcal{J}'(M_1+M_4) \ , \\ H_3 &= \mathcal{J}'(M_1+2M_4+\tfrac{1}{2}) \ , \\ H_4 &= \mathcal{J}'(M_2+M_3+1) \ , \end{aligned} \tag{18}$$

with the moments given by Eq. (13). The following curves, i.e., Fig. 2(d), shows the results obtained from the computer calculation.

We can see that $M_0 < M_1 < \cdots < M_4$ for all values of $\mathcal{J}/\Delta < 1$ and $\mathcal{J}'/\Delta' > 1$. The effect of the vacancy is to depolarize the four atoms of the first-neighbor shell, thus lowering the value of M_1. This effect is also felt, to a lesser extent, by the atoms of outer shells which consequently have larger moments than M_1 but lower than the bulk value. The moment M_0 at the vacancy site will be maximum for $\mathcal{J}/\Delta$ near the magnetic condition. As this parameter decreases, the effect of the magnetic host atoms polarizing the impurity becomes weaker, therefore M_0 decreases. We see that as $\mathcal{J}'/\Delta'$ increases, M_0 also increases. The effect on the moments $M_1, \ldots, M_4$ will be of an increase in their values, which will gradually tend to $\frac{1}{2}$. The depolarization effect of the vacancy is felt by the other atoms to a lesser extent as the main effect is the polarization of the impurity atom by the strongly magnetic host. A similar effect is achieved for the same $\mathcal{J}'/\Delta'$ but now with $\mathcal{J}'$, Δ' being twice the values they had previously. The following curve [Fig. 2(e)], shows the moment distribution for various ratios of the parameters $\mathcal{J}$, $\mathcal{J}'$, Δ, and Δ'. Figure 2(e) is an interesting physical situation: $\mathcal{J}'/\Delta'$ is still greater than 1 but $\mathcal{J}$, Δ are twice their values such that the ratio $\mathcal{J}/\Delta$ is unchanged, i.e., < 1. Now, $\mathcal{J} > \mathcal{J}'$ and $\Delta/\Delta' = 2$. Here again, the main effect is

seen on M_1. As $\mathcal{J} > \mathcal{J}'$, the effective field H_1 can be very large, the more so the nearer $\mathcal{J}/\Delta$ is from the magnetic condition, as M_0 will be larger in that region. Therefore, we can have a range of values of $\mathcal{J}'$, Δ' for which $M_1 > M_4$ but still be smaller than the bulk value $\frac{1}{2}$. The moment M_1 is thus greater than M_2, M_3, M_4, always being that $M_2 > M_3 > M_4$ because these atoms will not feel the effect of the impurity. Only when $\mathcal{J} = \mathcal{J}'$ does M_1 become smaller than M_2, M_3, and M_4. If we decrease $\mathcal{J}/\Delta$ further than that value, we regain the picture of Fig. 2(d), i.e., the moments increase monotonically from M_0 to M_4, maximum value of all the moments and smaller than $\frac{1}{2}$. As $\mathcal{J}'/\Delta'$ increases, the host becomes more strongly magnetic and it reaches a value $\mathcal{J}' > \mathcal{J}$; at that point, for all values of $\mathcal{J}/\Delta$ the condition $M_0 < M_1 < \cdots < M_4$ holds again.

If we reduce $\mathcal{J}$ by one-half and Δ by the same amount so that the ratio $\mathcal{J}/\Delta$ is left unchanged, we see very little change in M_0. But because $\mathcal{J}$ is now weaker, M_1 will take slightly lower values because it will feel the depolarization effect of the vacancy. Again, the main effect is observed on M_1, the other moments showing very little change.

1. *Simulation of a metal (long-range interaction)*

We simulate now the case of a metal, always for a square lattice at $T = 0$. The exchange interaction is now long range; i.e., it allows for interactions of an atom with neighbors of up to the fourth-neighbor shell. The exchange interaction is taken proportional to $1/r^3$ in order to simulate the situation in a real metal. The system of equations which gives the effective fields is now the following:

$$
\begin{aligned}
H_0 &= \mathcal{J}\left(4M_1 + \sqrt{2}M_2 + \tfrac{1}{2}M_3 + \frac{8M_4}{5\sqrt{5}}\right) , \\
H_1 &= \mathcal{J}M_0 + \mathcal{J}'\left(2M_2 + M_3 + \frac{1}{\sqrt{2}}(M_1 + M_4)\right. \\
&\qquad \left. + \tfrac{1}{8}(M_1 + 2M_4) + \frac{M_2 + M_3}{8\sqrt{2}}\right) , \\
H_2 &= \frac{\mathcal{J}M_0}{2\sqrt{2}} + \mathcal{J}'\left(2(M_2 + M_3) + \frac{M_3}{\sqrt{2}} + \frac{M_3}{4}\right. \\
&\qquad \left. + \frac{1}{8\sqrt{2}}(M_1 + M_4)\right) , \qquad (19) \\
H_3 &= \mathcal{J}\frac{M_0}{8} + \mathcal{J}'\left(2M_4 + M_1 + \frac{M_2}{\sqrt{2}} + \frac{1}{8\sqrt{2}}(M_1 + M_4)\right) , \\
H_4 &= \mathcal{J}\frac{M_0}{16\sqrt{2}} + \mathcal{J}'\left(M_2 + M_3 + \frac{1}{2\sqrt{2}}(M_1 + M_4)\right. \\
&\qquad \left. + \tfrac{1}{8}(M_1 + M_4) + \frac{1}{16\sqrt{2}}(M_2 + M_3)\right) .
\end{aligned}
$$

The following curves [Figs. 3(a) and 3(b)], show the case of a magnetic impurity in a nonmagnetic host, for the physical situation in which the exchange interaction is long range. The curves show that the effect of the long-range interaction is that of increasing all moments, as compared with their values in the case of a short-range interaction. The polarization effect of the impurity is felt by all atoms within the first four-neighbor shells that were considered. As $\mathcal{J}'/\Delta'$ decreases, all moments decrease; but we can see that M_2, M_3, and M_4 take values which are at least two orders of magnitude higher than in the case of the short-range interaction. As $\mathcal{J}/\Delta$ increases, the magnetic impurity has a stronger moment, and it feels less the effect of the vacancies surrounding it; indeed, the effect is that of a considerable enhancement of all moments.

Consider now the case for which $\mathcal{J}/\Delta$ is such that $\mathcal{J}$, Δ have twice their values in the previous case [see Fig. 3(b)], so that the ratio $\mathcal{J}/\Delta$ is unchanged. This results in an even stronger enhancement of all moments. If we consider the case of $\mathcal{J}'$, Δ' having twice their values of the previous case but still $\mathcal{J}'/\Delta'$ remains unchanged [Fig. 3(b)], we arrive at another interesting physical situation for $\mathcal{J}/\Delta > 1$. The strong $\mathcal{J}'$ will compete with the corresponding $\mathcal{J}$, producing a depolarization effect which will cause a gradual falloff of all moments, though in a less abrupt fashion than in the situation with short-range interaction. The effect is due to the two competing interactions: $\mathcal{J}$, now long range, slightly larger than the critical ratio 1, and $\mathcal{J}'$ greater than $\mathcal{J}$ until it reaches a value $\mathcal{J}'/\Delta'$ which is equal to $\mathcal{J}/\Delta$. At this point, all moments fall rapidly to zero. A condition for this to occur can be got along similar lines to Eq. (17): For the case of the long-range exchange, the expression becomes more cumbersome than in the case of the short-range exchange because the effective fields have more complicated expressions, as can be seen from Eq. (19). We therefore present a curve [Fig. 3(b)], which clearly shows the physical situations just described. We now consider a nonmagnetic impurity in a magnetic host, for a long-range exchange interaction, always in a two-dimensional square lattice. Here again, we have assumed that all the moments in the outer shells have a value equal to the bulk value; i.e., $M_i = \frac{1}{2}$ for all atoms outside the first four shells of neighbors to the impurity. The most important effect to be observed is that of the strong polarization of the vacancy impurity by the host atoms. For all values of $\mathcal{J}/\Delta$, given a certain $\mathcal{J}'/\Delta'$, we see that $M_0 < M_1 < \cdots < M_4$. The moment M_1 will be accordingly larger than corresponding to the short-range interaction, and so all moments will be enhanced. The moments $M_1, \ldots, M_4$ are now almost unaffected by the vacancy for all values of $\mathcal{J}/\Delta$.

The case for which $\mathcal{J}$, Δ have twice their previous values but still $\mathcal{J}/\Delta$ is unchanged is again an interest-

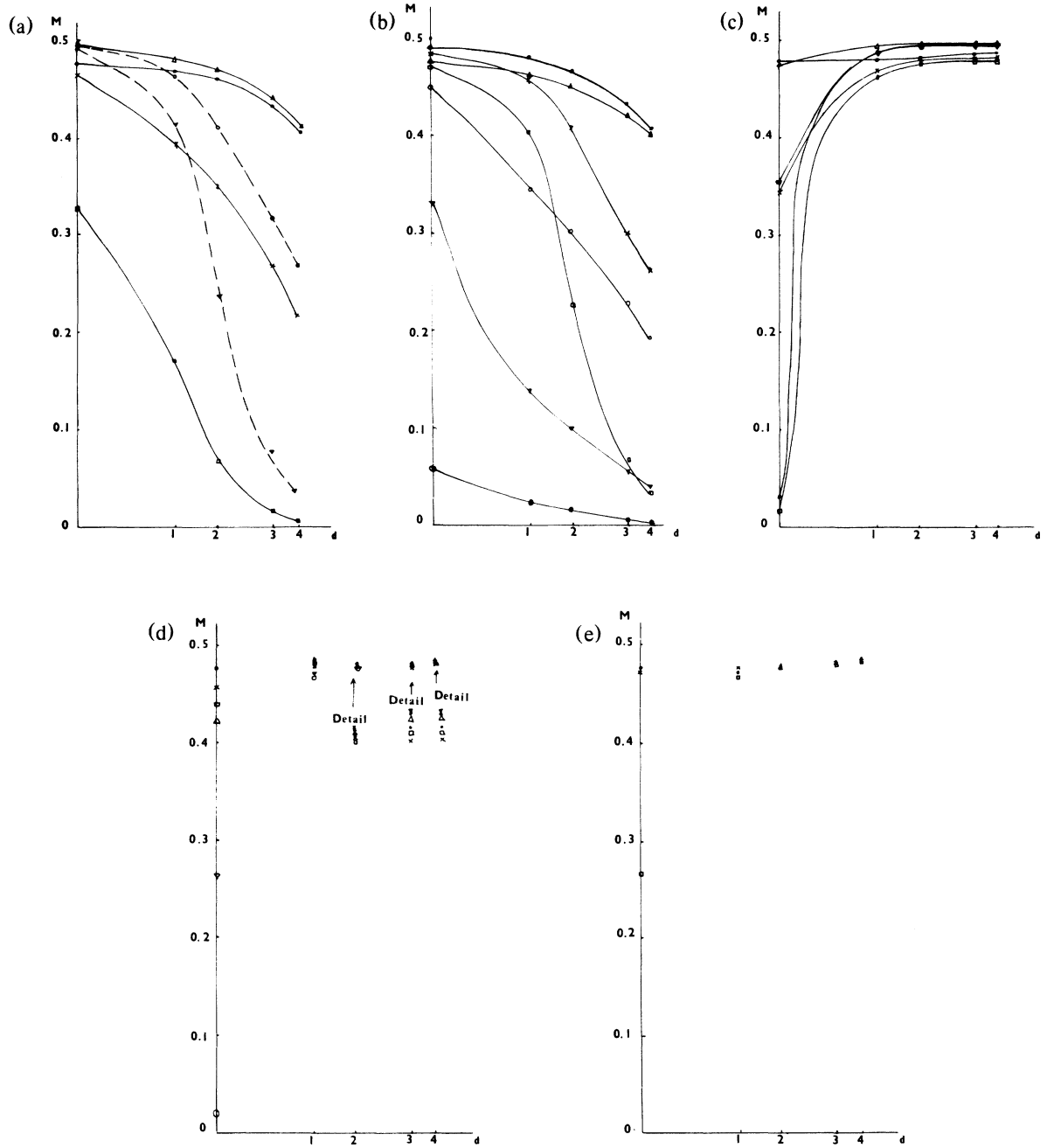

FIG. 3. Singlet-singlet systems, 1 impurity ($T=0$). Two-dimensional case. $1/r^3$ interaction. (a) Magnetic impurity in nonmagnetic host: ●$-\mathcal{J}/\Delta=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=1.1/0.99$; ×$-\mathcal{J}/\Delta=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=1.1/0.5$; □$-\mathcal{J}/\Delta=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=1.1/0.01$; Δ$-\mathcal{J}/\Delta=3$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=3/0.99$; ○$-\mathcal{J}/\Delta=3$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=3/0.5$; and ▽$-\mathcal{J}/\Delta=3$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=3/0.01$. (b) Magnetic impurity in nonmagnetic host: ●$-\mathcal{J}/\Delta=1.5$, $\Delta/\Delta'=2$, $\mathcal{J}/\mathcal{J}'=3/0.99$; ×$-\mathcal{J}/\Delta=1.5$, $\Delta/\Delta'=2$, $\mathcal{J}/\mathcal{J}'=3/0.5$; □$-\mathcal{J}/\Delta=1.5$, $\Delta/\Delta'=2$, $\mathcal{J}/\mathcal{J}'=3/0.01$; Δ$-\mathcal{J}/\Delta=1.1$, $\Delta/\Delta'=1/2$, $\mathcal{J}/\mathcal{J}'=1.1/1.98$; ○$-\mathcal{J}/\Delta=1.1$, $\Delta/\Delta'=1/2$, $\mathcal{J}/\mathcal{J}'=1.1/1$; ▽$-\mathcal{J}/\Delta=1.1$, $\Delta/\Delta'=1/2$, $\mathcal{J}/\mathcal{J}'=1.1/0.6$; and ○$-\mathcal{J}/\Delta=1.1$, $\Delta/\Delta'=1/2$, $\mathcal{J}/\mathcal{J}'=1.1/0.4$. (c) Nonmagnetic impurity in magnetic host: ●$-\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=0.99/1.1$; ×$-\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=0.3/1.1$; □$-\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=0.01/1.1$; Δ$-\mathcal{J}'/\Delta'=3$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=0.99/3$; ▽$-\mathcal{J}'/\Delta'=3$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=0.3/3$; and ○$-\mathcal{J}'/\Delta'=3$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=0.01/3$. (d) Nonmagnetic impurity in magnetic host: ●$-\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=2$, $\mathcal{J}/\mathcal{J}'=1.98/1.1$; ×$-\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=2$, $\mathcal{J}/\mathcal{J}'=1.4/1.1$; □$-\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=2$, $\mathcal{J}/\mathcal{J}'=1.2/1.1$; Δ$-\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=2$, $\mathcal{J}/\mathcal{J}'=1/1.1$; ▽$-\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=2$, $\mathcal{J}/\mathcal{J}'=0.4/1.1$; and ○$-\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=2$, $\mathcal{J}/\mathcal{J}'=0.02/1.1$. (e) Nonmagnetic impurity in magnetic host: ●$-\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=0.5/1$, $\mathcal{J}/\mathcal{J}'=0.495/1.1$; ×$-\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=0.5/1$, $\mathcal{J}/\mathcal{J}'=0.475/1.1$; and □$-\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=0.5/1$, $\mathcal{J}/\mathcal{J}'=0.1/1.1$.

ing one. Just as in the short-range situation, for values of $\mathcal{J} > \mathcal{J}'$ and until $\mathcal{J}/\Delta$ becomes equal to $\mathcal{J}'/\Delta'$, the moment M_1 can be greater than M_4. As we let $\mathcal{J}$ decrease, M_1 will gradually decrease and become only greater than M_2, and finally when $\mathcal{J}=\mathcal{J}'$ we regain the situation $M_0 < M_1 < \cdots < M_4$ [Figs. 3(c) and 3(d)]. The following curves, Figs. 3(c), 3(d), and 3(e), show graphically the cases just discussed.

C. Three-dimensional crystal

Let us now consider the case of a three-dimensional crystal. For simplicity, we have only considered nearest-neighbor interaction between any pair of atoms in the lattice. The equations for the effective fields at each site will depend on the geometrical configuration of the lattice. If we take first a simple cubic lattice, we get

$$\begin{aligned} H_0 &= 6\mathcal{J}M_1 \ , \\ H_1 &= \mathcal{J}M_0 + \mathcal{J}'(4M_2 + M_4) \ , \\ H_2 &= 2\mathcal{J}'(M_1 + M_3 + M_4) \ , \\ H_3 &= 3\mathcal{J}'M_2 \ , \\ H_4 &= \mathcal{J}'M_1 \ . \end{aligned} \tag{20}$$

We have again assumed that all moments outside the fourth-neighbor shell vanish. This is a good approximation for small values of $\mathcal{J}'/\Delta'$; i.e., $\mathcal{J}'/\Delta' \lesssim 0.5$. For larger values of this parameter, M_4 is no longer small and we have to turn to the continuum expression and calculate down to which site the effect of the magnetic impurity is still felt by the host. Alternatively, one can look for a value of $\mathcal{J}/\Delta$ for which the moments just form; in this case, a linear Ginzburg-Landau equation can be used. The computer calculation was done using the same iteration procedure as for the linear-chain and the square-lattice cases. For $\mathcal{J}/\Delta$ greater than 1, and due to the fact that the magnetic impurity has now six nearest neighbors which will feel its effect, we can see that the moments are considerably enhanced if we compare them to their values in the two-dimensional case. For $\mathcal{J}/\Delta$ large, the impurity is strongly magnetic and feels very little the effect of its nonmagnetic surroundings; the impurity moment is thus nearly unchanged for all values of $\mathcal{J}'/\Delta'$. We can see that the general behavior of the moments is similar to the behavior observed in the one- and two-dimensional cases. This is clearly shown in the following set of curves, Figs. 4(a) and 4(b).

If we now look at the case for which $\mathcal{J}/\Delta$ is slightly above the magnetic condition but $\mathcal{J}, \Delta$ have values equal to one-half of their previous values, we see that for $\mathcal{J} < \mathcal{J}'$ all moments decrease and after the condition $\mathcal{J}=\mathcal{J}'$ is reached, all moments fall to zero very rapidly. We can again get a relationship, by means of a small moment expansion, that will tell us, for a given $\mathcal{J}/\Delta$, the value of $\mathcal{J}'/\Delta'$ for which all moments just vanish. We thus have, to first order in $\mathcal{J}/\Delta$ and $\mathcal{J}'/\Delta'$,

$$\begin{aligned} M_4 &\simeq \frac{\mathcal{J}'M_1}{2\Delta'} \ , \\ M_3 &\simeq \frac{3}{2}\frac{\mathcal{J}'M_2}{\Delta'} \ , \\ M_2 &\simeq \frac{\mathcal{J}'M_1}{\Delta} \ , \\ M_0 &\simeq \frac{3\mathcal{J}M_1}{\Delta} \ . \end{aligned}$$

If we now substitute these into the expression for M_1, we get the condition for all moments to vanish, viz.,

$$\frac{3}{2}\frac{\mathcal{J}^2}{\Delta\Delta'} + \frac{15}{4}\frac{\mathcal{J}'^2}{\Delta'^2} = 1 \ . \tag{21}$$

If we take $\mathcal{J}/\Delta > 1$ but let $\mathcal{J}, \Delta$ have values equal to twice their original values, we go back to the original behavior of the system, i.e., a strong enhancement of all moments due to the presence of the magnetic impurity. The main effect is clearly felt by the first-neighbor shell, as we have assumed a short-range interaction. We can see again the same outstanding feature as before: In such a system, a single magnetic impurity is enough to create strong moments, for $\mathcal{J}'/\Delta'$ near the magnetic condition and for values of $\mathcal{J}/\Delta$ just above the magnetic condition. The calculation shows that a value of $\mathcal{J}/\Delta = 1.01$ is enough to create nonzero moments in the system, for values of $\mathcal{J}'/\Delta'$ near the critical ratio 1. We will make use of this result when we treat the continuum solution. The following curve, Fig. 4(b), shows the two situations discussed above.

Let us now turn to the case of a nonmagnetic impurity in a magnetic host, for the case of a simple cubic lattice with nearest-neighbor interaction. We have again assumed that all the moments outside the fourth-neighbor shell are equal to the bulk value $\frac{1}{2}$. Accordingly, the equations for the effective fields become

$$\begin{aligned} H_0 &= 6\mathcal{J}M_1 \ , \\ H_1 &= \mathcal{J}M_0 + \mathcal{J}'(4M_2 + M_4) \ , \\ H_2 &= 2\mathcal{J}'(M_1 + M_3 + M_4) \ , \\ H_3 &= 3\mathcal{J}'M_2 + 1.5 \ , \\ H_4 &= \mathcal{J}'M_1 + 1.5 \ . \end{aligned} \tag{22}$$

The magnetic host polarizes the vacancy impurity, and all moments increase monotonically from the impurity value to the value at the fourth-neighbor shell. As $\mathcal{J}'/\Delta'$ increases, and although the condition $M_0 < M_1 < \cdots < M_4$ still holds, the values of M_1,

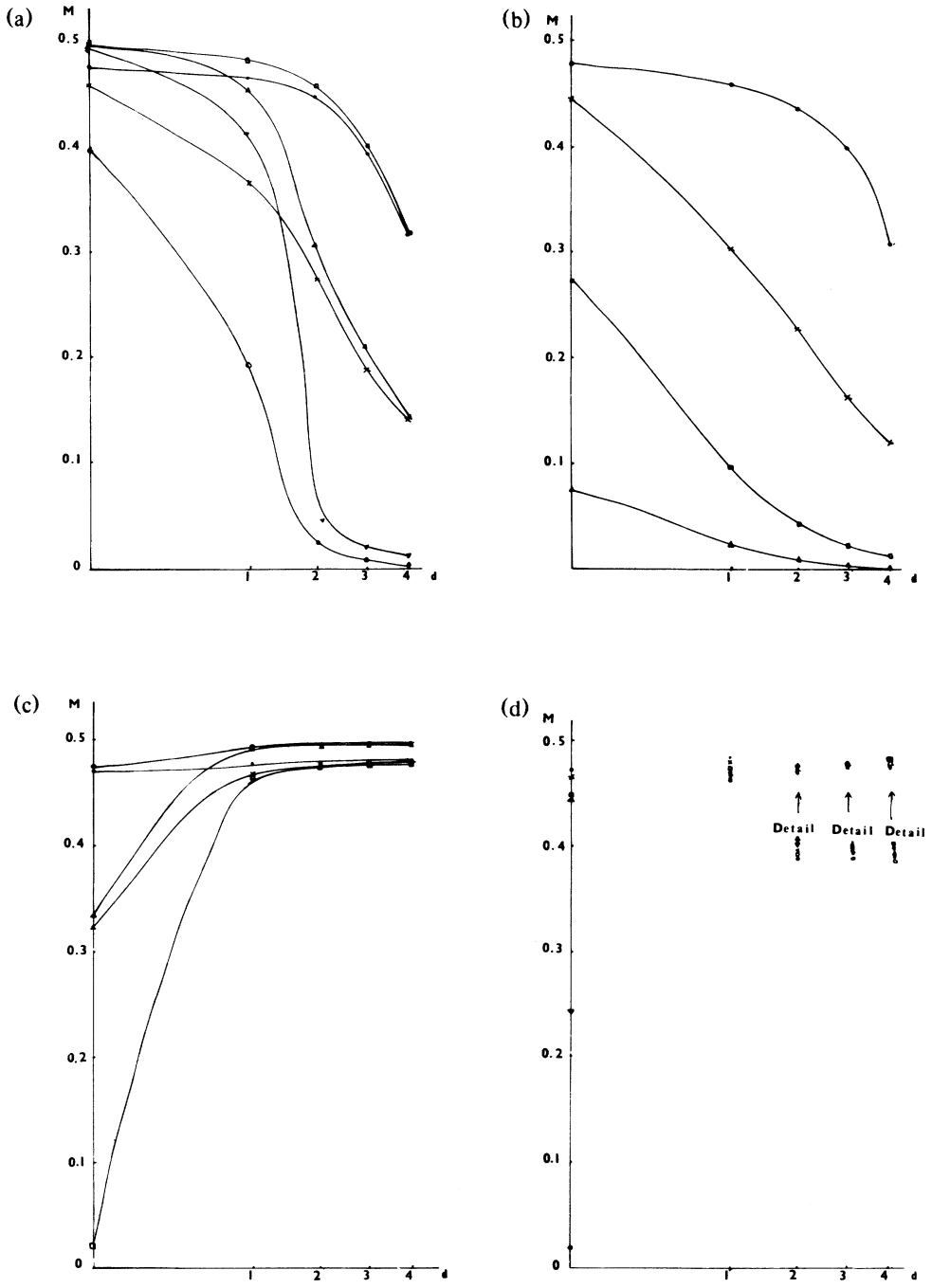

FIG. 4. Singlet-singlet systems, 1 impurity ($T=0$). Three-dimensional case. Nearest-neighbor interaction. (a) Simple cubic lattice. Magnetic impurity in nonmagnetic host: ●—$\mathcal{J}/\Delta=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=1.1/0.99$; ×—$\mathcal{J}/\Delta=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=1.1/0.5$; ○—$\mathcal{J}/\Delta=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=1.1/0.01$; □—$\mathcal{J}/\Delta=3$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=3/0.99$; Δ—$\mathcal{J}/\Delta=3$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=3/0.5$; and ▽—$\mathcal{J}/\Delta=3$, $\Delta/\Delta'=1$, $\mathcal{J}/\mathcal{J}'=3/0.01$. (b) Magnetic impurity in nonmagnetic host: ●—$\mathcal{J}/\Delta=0.55/0.5$, $\Delta/\Delta'=0.5/1$, $\mathcal{J}/\mathcal{J}'=0.55/0.99$; ×—$\mathcal{J}/\Delta=0.55/0.5$, $\Delta/\Delta'=0.5/1$, $\mathcal{J}/\mathcal{J}'=0.55/0.2$; □—$\mathcal{J}/\Delta=0.55/0.5$, $\Delta/\Delta'=0.5/1$, $\mathcal{J}/\mathcal{J}'=0.55/0.3$; and Δ—$\mathcal{J}/\Delta=0.55/0.5$, $\Delta/\Delta'=0.5/1$, $\mathcal{J}/\mathcal{J}'=0.55/0.2$. (c) Nonmagnetic impurity in magnetic host: ●—$\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}'/\mathcal{J}=1.1/0.99$; ×$\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}'/\mathcal{J}=1.1/0.3$; □—$\mathcal{J}'/\Delta'=1.1$, $\Delta/\Delta'=1$, $\mathcal{J}'/\mathcal{J}=1.1/0.01$; ○—$\mathcal{J}'/\Delta'=3$, $\Delta/\Delta'=1$, $\mathcal{J}'/\mathcal{J}=3/0.99$; and Δ—$\mathcal{J}'/\Delta'=3$, $\Delta/\Delta'=1$, $\mathcal{J}'/\mathcal{J}=3/0.3$. (d) Nonmagnetic impurity in magnetic host: ●—$\mathcal{J}'/\Delta'=1.1$, $\Delta'/\Delta=1/2$, $\mathcal{J}'/\mathcal{J}=1.1/1.98$; ×—$\mathcal{J}'/\Delta'=1.1$, $\Delta'/\Delta=1/2$, $\mathcal{J}'/\mathcal{J}=1.1/1.8$; □—$\mathcal{J}'/\Delta'=1.1$, $\Delta'/\Delta=1/2$, $\mathcal{J}'/\mathcal{J}=1.1/1.4$; Δ—$\mathcal{J}'/\Delta'=1.1$, $\Delta'/\Delta=1/2$, $\mathcal{J}'/J=1.1/1.2$; ▽—$\mathcal{J}'/\Delta'=1.1$, $\Delta'/\Delta=1/2$, $\mathcal{J}'/\mathcal{J}=1.1/0.4$; and ○—$\mathcal{J}'/\Delta'=1.1$, $\Delta'/\Delta=1/2$, $\mathcal{J}'/\mathcal{J}=1.1/0.02$.

M_2, M_3, M_4 get very close to each other and tend to the bulk value $\frac{1}{2}$. We thus see that the effect of the vacancy is too weak to affect the magnetic host. This situation is clearly seen in Figs. 4(c) and 4(d).

If we let $\mathcal{J}'/\Delta'$ be just above the magnetic condition, and we take $\mathcal{J}, \Delta$ equal to twice their values in the previous cases still keeping the ratio $\mathcal{J}/\Delta$ unchanged, i.e., $\mathcal{J}/\Delta < 1$, We have again the situation encountered in the case of the two-dimensional lattice. Namely, M_1 can be larger than M_4 for all values of $\mathcal{J}/\Delta$ for which $\mathcal{J} > \mathcal{J}'$. When one gets to the point where $\mathcal{J} = \mathcal{J}'$, one comes back to the condition $M_0 < M_1 < \cdots < M_4$. Figure 4(d) shows this situation. Another interesting configuration is that of a bcc crystal. We discuss this case again assuming nearest-neighbor interaction for simplicity.

In the case of a magnetic impurity in a nonmagnetic host, we write the effective fields assuming that the moments outside the fourth-neighbor shell are zero. This will be seen to be valid again for $\mathcal{J}'/\Delta' \lesssim 0.5$; for $\mathcal{J}'/\Delta'$ near the magnetic condition, where all moments will be seen to be large even for $\mathcal{J}/\Delta$ just above the magnetic condition, we will have to use the continuum expression. We thus have

$$\begin{aligned} H_0 &= B8M_1 \ , \\ H_1 &= \mathcal{J}M_0 + \mathcal{J}'(3M_2 + 4M_3) \ , \\ H_2 &= 4\mathcal{J}'(M_1 + M_4) \ , \\ H_3 &= 2\mathcal{J}'(M_1 + 2M_4) \ , \\ H_4 &= \mathcal{J}'(2M_3 + M_2) \ . \end{aligned} \tag{23}$$

The following curve, i.e., Fig. 5(a), shows the moment distribution in space, from the impurity site down to the fourth-neighbor shell of atoms. The behavior is very similar to the simple cubic lattice case; in the bcc, the moments M_0 and M_1 are larger as there are now more neighbors that are going to contribute to achieve the enhancement.

In the case of a nonmagnetic impurity in a magnetic crystal, we have again assumed that all moments outside the fourth-neighbor shell are equal to the bulk value $\frac{1}{2}$. The corresponding expression for the

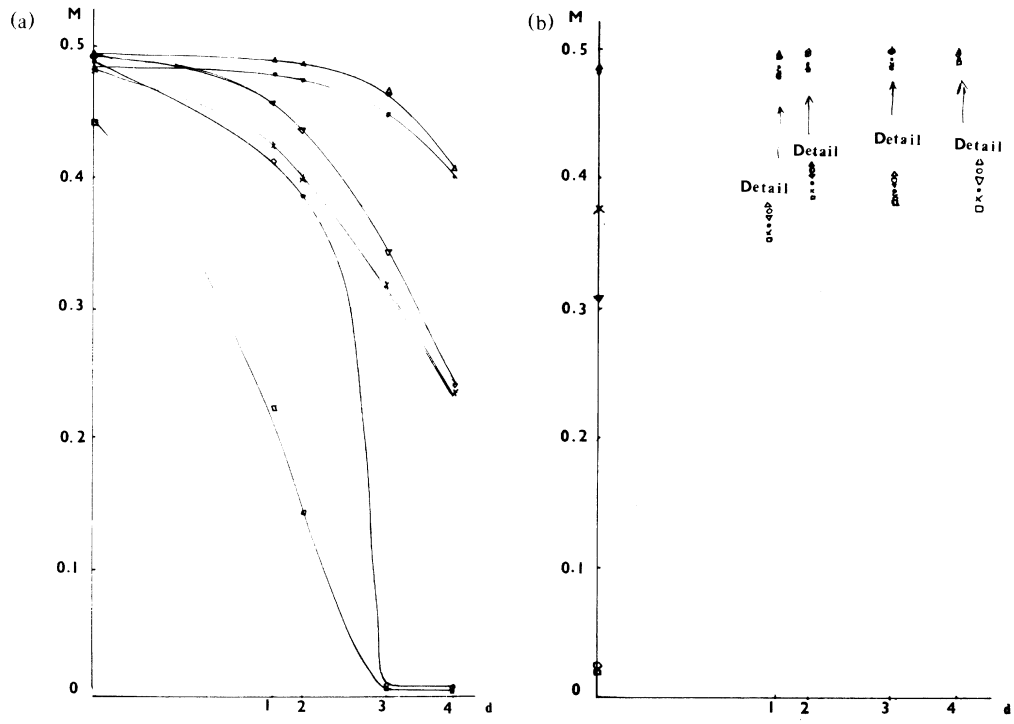

FIG. 5. Singlet-singlet systems, 1 impurity ($T = 0$). Three-dimensional case. Nearest-neighbor interaction. (a) bcc lattice. Magnetic impurity in nonmagnetic host: ● $-\mathcal{J}/\Delta = 1.1$, $\Delta'/\Delta = 1$, $\mathcal{J}/\mathcal{J}' = 1.1/0.99$; × $-\mathcal{J}/\Delta = 1.1$, $\Delta'/\Delta = 1$, $\mathcal{J}/\mathcal{J}' = 1.1/0.5$; □ $-\mathcal{J}/\Delta = 1.1$, $\Delta'/\Delta = 1$, $\mathcal{J}/\mathcal{J}' = 1.1/0.01$; Δ $-\mathcal{J}/\Delta = 3$, $\Delta'/\Delta = 1$, $\mathcal{J}/\mathcal{J}' = 3/0.99$; ▽ $-\mathcal{J}/\Delta = 3$, $\Delta'/\Delta = 1$, $\mathcal{J}/\mathcal{J}' = 3/0.5$; and ○ $-\mathcal{J}/\Delta = 3$, $\Delta'/\Delta = 1$, $\mathcal{J}/\mathcal{J}' = 3/0.01$. (b) Nonmagnetic impurity in magnetic host: Δ $-\mathcal{J}'/\Delta' = 1.1$, $\Delta/\Delta' = 1$, $\mathcal{J}/\mathcal{J}' = 0.99/1.1$; ○ $-\mathcal{J}'/\Delta' = 1.1$, $\Delta/\Delta' = 1$, $\mathcal{J}/\mathcal{J}' = 0.5/1.1$; ▽ $-\mathcal{J}'/\Delta' = 1.1$, $\Delta/\Delta' = 1$, $\mathcal{J}/\mathcal{J}' = 0.01/1.1$; ● $-\mathcal{J}'/\Delta' = 3$, $\Delta/\Delta' = 1$, $\mathcal{J}/\mathcal{J}' = 0.99/3$; × $-\mathcal{J}'/\Delta' = 3$, $\Delta/\Delta' = 1$, $\mathcal{J}/\mathcal{J}' = 0.5/3$; and □ $-\mathcal{J}'/\Delta' = 3$, $\Delta/\Delta' = 1$, $\mathcal{J}/\mathcal{J}' = 0.01/3$.

effective fields becomes

$$
\begin{aligned}
H_0 &= 8\mathcal{J}M_1 \ , \\
H_1 &= \mathcal{J}M_0 + \mathcal{J}'(3M_2 + 4M_3) \ , \\
H_2 &= 4\mathcal{J}'(M_1 + M_4) \ , \\
H_3 &= \mathcal{J}'(2M_1 + 4M_4 + 1) \ , \\
H_4 &= \mathcal{J}'(M_2 + 2M_3 + 2.5) \ .
\end{aligned} \tag{24}
$$

The following curve, Fig. 5(b), shows that the behavior $M_0 < M_1 < \cdots < M_4$ still holds, although the values of $M_1, \ldots, M_4$ are somewhat higher than in the case of the simple cubic crystal. As the host becomes more strongly magnetic, we can see that the effect of the vacancy is very weak; i.e., it is hardly felt by the neighbors, and the moments $M_1, \ldots, M_4$ have values very close to the bulk value $\frac{1}{2}$.

D. Conclusions from the computer simulation

We have studied the moment distribution for the one-, two-, and three-dimensional lattices, both for the case of a magnetic impurity placed in a nonmagnetic host, and for a nonmagnetic impurity in a magnetic host. We have treated the case of a short-range interaction (nearest neighbor) and that of a long-range exchange ($\propto 1/r^3$), in order to simulate, respectively, the case of an insulator and that of a real metal.

The general behavior of the curves corresponding to the case of a magnetic impurity in a nonmagnetic host lattice is that of a decrease of the moment, from the maximum value at the impurity site to the lowest at the fourth-neighbor site. If the $\mathcal{J}/\Delta$ parameter is just above the magnetic condition, we have seen that the presence of a single impurity is enough to create moments at all sites, for values of $\mathcal{J}'/\Delta'$ near the critical value for magnetism. It is in this region that a continuum expression should be used. A Ginzburg-Landau equation will be used, in its linear form (i.e., for small moments, as will be explained below) for values of $\mathcal{J}/\Delta$ which just set moments on all sites. The calculation has showed that a value of $\mathcal{J}/\Delta$ equal to 1.01 is sufficient to produce the formation of nonzero moments. For values of $\mathcal{J}'/\Delta'$ smaller than 0.5, we will use discrete solutions, as given by the equations corresponding to each case dealt with in the previous sections.

The behavior of the curves corresponding to the case of a nonmagnetic impurity in a magnetic-host lattice is that of an increase of the moments from the lowest value at the impurity site to the maximum value at the fourth-neighbor site. Here again, for values of $\mathcal{J}/\Delta$ near the critical ratio 1 we will use the continuum expression. But for lower values of that parameter, we will use the discrete solutions by means of the equations corresponding to each case.

III. CONTINUUM REPRESENTATION

Bearing in mind the results of the computer simulation, we can now go back to the general equation for the singlet-singlet problem. We will show that one can transform the discrete Landau expansion into an integro-differential Ginzburg-Landau equation. We will get a self-consistent equation for $M(\vec{r})$ which involves such parameters as the exchange interaction $\mathcal{J}(\vec{r}, \vec{r}')$ and the crystal-field $\Delta(\vec{r})$. The temperature dependence will also be included in this expression.

We will then show the similarity between our expression for the Ginzburg-Landau equation deduced from first principles, and that used by Edwards, Mathon, and Wohlfarth[28] in treating the Pd-Ni alloys. We shall first treat the case of a single impurity of the magnetic type in a nonmagnetic host. Using the experimental results of Sarkissian[29] for the Pr-Tb susceptibility, we will derive the value of the constant that we need in order to proceed to the finite concentration case. We then calculate the critical concentration for magnetism to occur. Although caution must be applied to the results, we will finally give a comparison between this critical concentration and the result found in the computer simulation.

We therefore start by transforming the discrete Landau expansion into the continuum representation. This expansion is then the following one:

$$
\begin{aligned}
M(\vec{r}) = {} & \frac{1}{2\Delta(\vec{r})} \tanh\left[\frac{\Delta(\vec{r})}{2kT}\right] \int \mathcal{J}(\vec{r}, \vec{r}') M(\vec{r}')\, d\vec{r}' \\
& + \frac{1 - \tanh^2(\Delta(\vec{r})/2kT)}{4\Delta(\vec{r})(2kT)} \\
& \times \int\int\int \mathcal{J}^3(\vec{r}, \vec{r}') M^3(\vec{r}')\, d\vec{r}' \ .
\end{aligned} \tag{25}
$$

The exchange interaction has been written as $\mathcal{J}(\vec{r}, \vec{r}')$ and not as $\mathcal{J}(\vec{r} - \vec{r}')$ because, due to the presence of the impurity, the translational invariance does not hold any more. We now have two different types of exchange, that between host-host atoms and that between host-impurity atoms. We also have two different crystal fields, a $\Delta(\vec{r})$ for all host atoms where $r \neq 0$ if we take the impurity to be at the origin $r = 0$, and $\Delta(r = 0)$ for the impurity atom. We write the equation for $M(\vec{r})$, which is a Schrödinger-like differential equation, and then use an effective potential which allows us to deal with our physical situation.

There are two ways of transforming an integro-differential equation such as Eq. (25) into a differential equation. The first method uses a convolution integral and the expansion of $\mathcal{J}(\vec{k})$, the Fourier transform of the exchange interaction, for small $\vec{k}$.

The other method, a rather more sophisticated one, was used by Werthamer.[30] We shall here use the first method.

We start then with Eq. (25) but we write the exchange as $\mathcal{J}(\vec{r}-\vec{r}')$; the fact that there is an impurity will be introduced later in the formalism when solving the differential equation, and for the time being we can keep the translational invariance as if we were dealing with a pure system.

The linear term in Eq. (25) can be written as

$$\int \mathcal{J}(\vec{r}-\vec{r}')M(\vec{r}')\,d\vec{r}' = \int d\vec{r}' \frac{1}{(2\pi)^3}\int \exp[i\vec{k}\cdot(\vec{r}-\vec{r}')]\mathcal{J}(\vec{k})\,d\vec{k}\frac{1}{(2\pi)^3}\int \exp[i\vec{k}'\cdot\vec{r}']M(\vec{k}')\,d\vec{k}' .$$

We can commute the order of integration as

$$\frac{1}{(2\pi)^6}\int d\vec{k}\int d\vec{k}'\int d\vec{r}'\exp[-i(\vec{k}-\vec{k}')\cdot\vec{r}']\exp[i\vec{k}\cdot\vec{r}]\mathcal{J}(\vec{k})M(\vec{k}') .$$

Using the definition of the δ function, in this case $\delta(\vec{k}-\vec{k}')$, we get

$$\int \mathcal{J}(\vec{r}-\vec{r}')M(\vec{r}')\,d\vec{r}' = \frac{1}{(2\pi)^3}\int \mathcal{J}(\vec{k})M(\vec{k})\exp(i\vec{k}\cdot\vec{r})\,d\vec{k} .$$

Assuming that we can expand $\mathcal{J}(\vec{k})$ around $k=0$, we get

$$\mathcal{J}(\vec{k}) = \mathcal{J}(0) + k^2\frac{\partial^2 J}{\partial k^2}\Big|_{k=0} + \cdots .$$

Calling $\mathcal{J}''(0) = \partial^2 J/\partial k^2|_{k=0}$, we get

$$\frac{1}{(2\pi)^3}\int \mathcal{J}(0)M(\vec{k})\exp(i\vec{k}\cdot\vec{r})\,d\vec{k} + \frac{1}{(2\pi)^3}\int k^2 M(\vec{k})\exp(i\vec{k}\cdot\vec{r})\mathcal{J}''(0)\,d\vec{k} + \cdots$$

Going back to real space, this expression is

$$\mathcal{J}(0)M(\vec{r}) - \mathcal{J}''(0)\nabla^2 M(\vec{r}) + \cdots . \tag{26}$$

We can write explicitly $\mathcal{J}(0)$ as

$$\mathcal{J}(0) = \frac{1}{(2\pi)^3}\int \mathcal{J}(\vec{r}-\vec{r}')\,d\vec{r}' .$$

As we are going to discuss only the paramagnetic regime in these alloys, we can omit the term in M^3 for as being much smaller than the linear term. Using Eqs. (25) and (26), we get

$$M(\vec{r}) = \frac{1}{2\Delta(\vec{r})}\tanh\left(\frac{\Delta(\vec{r})}{2kT}\right) \times [M(\vec{r})\mathcal{J}(0) - \mathcal{J}''(0)\nabla^2 M(\vec{r})] . \tag{27}$$

The temperature-dependent coefficient in Eq. (27) depends upon r because the crystal field is r dependent. Thus we get, calling

$$A(\vec{r}) = \frac{1}{2\Delta(\vec{r})}\tanh\left(\frac{\Delta(\vec{r})}{2kT}\right) ,$$

the expression

$$-\mathcal{J}''(0)A(\vec{r}) + \nabla^2 m(\vec{r}) + [A(\vec{r})\mathcal{J}(0) - 1]M(\vec{r}) = 0 ,$$

which can be written as

$$-\nabla^2 M(\vec{r}) + \left(\frac{\mathcal{J}(0)}{\mathcal{J}''(0)} - \frac{1}{\mathcal{J}''(0)A(\vec{r})}\right)M(\vec{r}) = 0 . \tag{28}$$

Let us define a correlation length κ^{-1} which will give an idea of how the magnetization varies in space. Following Edwards, Mathon, and Wohlfarth,[28] we define

$$\kappa^{-1} = \left(\frac{\mathcal{J}''(0)A(\vec{r})}{\mathcal{J}(0)A(\vec{r})-1}\right)^{1/2} = \left(\frac{J''(0)}{J(0)-1/A(\vec{r})}\right)^{1/2} . \tag{29}$$

We are now going to introduce into the equation the information about there being an impurity. We can think of the coefficient of the linear term in Eq. (28) as being composed of a constant value V_0 corresponding to the host matrix, and a term $V(\vec{r})$ which is the effective potential around the impurity atom, i.e., in a sphere of radius calculated from the atomic volume of the host atom. If we call $B = -1 + A(\vec{r})J(0)$, then $B = V_0 + V(\vec{r})$. Thus, for $r > a$, $V(\vec{r}) = 0$. As our impurity will be taken to be magnetic, this will impose a condition on the coefficient of the linear term. This coefficient is in fact κ^2, the square of the correlation length previously defined. The condition of the impurity being magnetic shows that the fact of $\mathcal{J}/\Delta$ being > 1 is consistent with a positive value of κ^2, as it should be. We can pursue the analogy with the Schrödinger equation in order to solve Eq. (28), which has now the form

$$-\nabla^2 M(\vec{r}) + \kappa^2 M(\vec{r}) = 0 . \tag{30}$$

As Edwards, Mathon, and Wohlfarth,[28] point out, it

is unreasonable to apply the Ginzburg-Landau equation within the atomic sphere that surrounds the impurity. Therefore, we are only interested in the exterior solution of Eq. (30); i.e., we will calculate $M(\vec{r})$ for $r > a$. In that case,

$$\kappa^{-1} = \left[\frac{\mathcal{J}''(0) A(\vec{r})}{V_0} \right]^{1/2} .$$

Using the notation of Edwards, Mathon, and Wohlfarth,[28] namely,

$$\rho = \kappa r ; \psi = \rho M ,$$

Eq. (30) becomes

$$\frac{-d^2\psi}{d\rho^2} + \psi = 0 . \tag{31}$$

This equation must satisfy the boundary condition for $\rho = \kappa a$; i.e., it has to match the logarithmic derivative $(1/\psi)(d\psi/d\rho)$ at $\rho = \kappa a$ must be finite for $\rho \to \infty$ and equal to a constant, which we call γ. This is

$$(1/\psi)(d\psi/d\rho)_{\rho=\kappa a} = \gamma .$$

The solution is thus written as

$$M(r) = \frac{H}{A_0} \left[1 + \frac{1 - \gamma\kappa a}{1 + \gamma} \left[\frac{\exp[-\kappa(r-a)]}{\kappa r} \right] \right] . \tag{32}$$

If we integrate $M(r)$ over all space outside the sphere of radius a, we get χ_{si}, the susceptibility for the single impurity:

$$\chi_{si} = \int_a^\infty \frac{M(\vec{r})}{H} d\vec{r} .$$

We can write a relationship between χ_{host} and χ_{si} as

$$\frac{\chi_{si}}{\chi_{host}} = 1 + \frac{3(1 - \gamma\kappa a)(1 + \kappa a)}{\kappa^3 a^3 (1 + \gamma)} . \tag{33}$$

We now use the experimental results of Sarkissian;[29] the low-field susceptibility of Pr-Tb alloys for very small concentrations of terbium is $\chi_{Pr-Tb} = 15 \times 10^{-4}$ emu/g, which we will use as the value of our χ_{si}. Koehler[31] gives a value of

$$\chi_{Pr} \simeq 15.5 \times 10^{-4} \text{ emu/g} .$$

We can now use the simplified expression for κa given by Shender in terms of the parameter $\mathcal{J}/\Delta$ of the magnetic impurity, viz., $\kappa a = (\mathcal{J}/\Delta - 1)^{1/2}$, and using the result of our computer simulation that tells us that the threshold value of $\mathcal{J}/\Delta$ for which there is just formation of moments is $\mathcal{J}/\Delta = 1.01$, i.e., when the moments are very small as they are just forming, we find that $\kappa a = 0.1$. With these values, we find that $\gamma = 10$, using Eq. (33). With this result, we can now go to the finite-concentration case in order to get the critical concentration for the onset of magnetism. We use the same model as that of Edwards, Mathon, and Wohlfarth[28] which is to put the magnetic impurities in a lattice whose lattice constant is determined by the concentration c. The boundary condition for the Schrödinger-like equation, using a Wigner-Seitz approximation, is then

$$\frac{1}{\psi}\frac{d\psi}{d\rho} = \gamma \quad \text{for } \rho = \kappa a$$
$$= 1/\rho \quad \text{for } \rho = \kappa b ,$$

where b is the radius of the Wigner-Seitz sphere. The relationship between this radius and the concentration is given by $c = 100(a/b)^3$, where c is given in percent. The solution of the Schrödinger-like equation for the magnetization is, for $a < r < b$

$$M(r) = \frac{H}{A_0} \left[1 + \frac{Ae^{\kappa r}}{\kappa r} + \frac{Be^{-\kappa r}}{\kappa r} \right] , \tag{34}$$

where

$$\frac{A}{B} = \delta \exp(-2\kappa b) ,$$

$$B = \frac{(\gamma\kappa a - 1)\exp(\kappa a)}{\delta(1-\gamma)\exp[-2\kappa(b-a)] - (1+\gamma)} ,$$

and

$$\delta = (\kappa b + 1)/(\kappa b - 1) .$$

The susceptibility of the alloy can be calculated by integrating $M(r)$ between $r = a$ and $r = b$. This gives

$$\frac{\chi_{alloy}}{\chi_{host}} = 1 + \frac{3(1 - \gamma\kappa a)}{\kappa^3 (b^3 - a^3)} \left[\kappa a + (\gamma\kappa a - 1) \frac{\delta \exp[-2\kappa(b-a)] + 1}{\delta \exp[-2\kappa(b-a)](1-\gamma) - (1+\gamma)} \right] . \tag{35}$$

The pole of χ gives the condition for the phase transition and thus the value of the critical concentration for the onset of magnetism. The condition is

$$[(\kappa b + 1)/(\kappa b - 1)] \exp[-2\kappa(b-a)] = 1 + \gamma/1 - \gamma .$$

We now use the values of κ, γ that we already obtained from the single impurity problem and we finally get $c_{crit} = 3.2\%$. With this result, we can now go back to the computer simulation and see whether this critical concentration is comparable to that found numerically. The critical ratio $\mathcal{J}/\Delta$ for the formation of moments was $\mathcal{J}/\Delta = 1.01$; for the case of a simple cubic lattice and taking account of neighbors down to

the fourth-neighbor shell from the impurity, having a single impurity in such a lattice meant a concentration 1/30, i.e., $c \simeq 3.3\%$. This result has to be treated with great caution as we have only accounted for nearest-neighbor interaction in the three-dimensional lattice in our computer simulation, which is not the most realistic model for a metal. Also, our calculation was done using mean-field theory (see Sec. II). Nevertheless, the result of the critical concentration found by means of the Ginzburg-Landau equation gives a value which is comparable to that found numerically. A more exact model should account for interactions between the impurities and deal with the disorder associated with these. Finally, the more general Ginzburg-Landau equation which includes the cubic term should be used to account for situations in which the moments are not necessarily small and where that nonlinear term may become important.

IV. GENERAL CONCLUSIONS

A computer simulation of the distribution of the magnetic moments in one-, two-, three-dimensional lattices was carried out for singlet-singlet systems with a single impurity, using a molecular-field-theory approach. A discrete solution for the value of the moment was obtained, as a function of the parameters $\mathcal{J}/\Delta$ and $\mathcal{J}'/\Delta'$, ratio of the exchange to the crystal field in the impurity and the host sites, respectively. For the continuum solution, a linear Ginzburg-Landau equation for the magnetic moment as a function of r was derived from first principles. Within the limits of the theory, a solution was obtained for the spatial distribution of the moment, and a value for the critical concentration of impurities for the onset of magnetism was calculated, the latter to be found in good agreement with the computer simulation. Although future studies should account for the various approximations introduced in the model, we believe that the behavior of singlet-singlet systems containing impurities has been found to be the correct one.

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