A mean-field, effective medium theory of random magnetic alloys I: The random Ising model

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Abstract. Starting from a physical RKKY interaction model for magnetic alloys and a valid probabilistic description of a random substitutional alloy, the possible phases have been studied in a mean field, effective medium theory. Most of the detailed objections raised against former treatments of these systems have been removed.

Keywords. Random magnetic alloys; spin glasses.

1. Introduction

Ever since Edwards and Anderson (1975) introduced the replica formalism to study the models for the so called 'spin glass' random alloys, numerous papers on 'exactly' solvable models have appeared in the literature. These models may be classified in three categories: (i) The Sherrington-Kirkpatrick (1975) replica model, (ii) the mean field type of approaches (Kaneyoshi 1976; Plefka 1976; Thouless *et al* 1977) and (iii) the virial expansion (Morita and Horiguchi 1976). The final phase diagrams are identical in all the three cases; the procedures for obtaining them, however, were quite different. The Sherrington-Kirkpatrick work has had serious objections raised against it: the zero replica limit is not justifiable mathematically, the free energy is not a *minimum* in terms of the two defined order parameters and finally in the low temperature limit the entropy and internal energies behave in an unphysical way. The mean field and virial expansion approaches are formally correct, but the 'solvable' models assume that the exchange interaction $J(|\mathbf{r}_i - \mathbf{r}_j|)$ is a random variable, distributed as a Gaussian about a mean J_0 *independent* of the separation $R = |\mathbf{r}_i - \mathbf{r}_j|$. As a necessary consequence, the interaction is infinitely weak and long ranged: $J(|\mathbf{r}_i - \mathbf{r}_j|) \sim O(N^{-1/2})$ and $J_0 \sim O(N^{-1})$, where N is the total number of spins in the system. Such a model is instructive no doubt, but not very suitable if we want to examine the phases of realistic alloys (CuMn, AuFe, AuCr, AgMn, etc). For these, the predominant energy is due to the RKKY indirect exchange between the magnetic atoms. Even if R is random, this interaction has a mean that is *strongly* dependent on the separation R. The statistical description is also different: here the N magnetic atoms can sit on any of the M lattice sites with equal probability. We are neglecting any site occupation correlation, either due to statistical fluctuations or chemical segregation. In the thermodynamic limit $N, M \to \infty$ such that $N/M \to c$, the concentration of the magnetic constituent in the alloy.

Pr (a magnetic atom labelled k occupies the site $\mathbf{r}_k$) $= 1/M$.

Pr (a site r_k is occupied by any one magnetic atom) $= N/M \to c$.

There can be $\sim M^N$ possible random configurations all equally probable. Such a statistical description is far cry from the Gaussian distribution for the J_s.

The virial expansion is valid either at low concentrations or very high temperatures (Larkin and Khmelmitskii 1970) or when $J \sim 0$ ($N^{-1/2}$). We are here interested in low temperatures, all concentration ranges and a realistic RKKY interaction. The virial expansion is no longer valid in the entire region of the phase space we are interested in.

The purpose of this paper is to examine this more realistic model using physical RKKY interactions and a physical statistical description. The results may look similar to the earlier works, but the procedure has certain distinctive features of its own. The ideas behind these new features are non-trivial and essential if we are to go beyond the simple assumptions of the exactly solvable models and apply our theory to more realistic situations.

2. The effective medium and local magnetisation

Let us begin by studying the spin $\frac{1}{2}$, classical Ising model. Generalisations to general spins and the Heisenberg models are straightforward and will be reported in a subsequent communication. The Hamiltonian is given by

$$H = -\tfrac{1}{2} \sum_{r_j \neq r_i} \sum J(|r_i - r_j|) \, S_i \, S_j$$

$$J(R) = [A \cos(2k_F R)]/(2k_F R)^3 \tag{1}$$

r_k denotes the position of the kth magnetic atom, $\{S_k\}$ are the classical spin operators with eigenvalues ± 1. The partition function is

$$Z(\{r_k\}) = \mathrm{Tr}_{\{S_k\}} \exp(\tfrac{1}{2} \beta \sum \sum J(|r_i - r_j|) \, S_i \, S_j). \tag{2}$$

The trace is taken over all possible spin configurations. The partition function remains a function of the random configurations of the magnetic ions. At this stage no random averaging is taken.

As it stands, the trace cannot be exactly taken. We shall introduce here a single-site 'mean field' approximation, by replacing all operator products AB by $A \langle B \rangle + \langle A \rangle B - \langle A \rangle \langle B \rangle$, where $\langle A \rangle$ denotes Tr $A \exp \{-\beta H\}/Z$, or the thermal average of A. Within this approximation

$$Z(\{r_k\}) = \exp\left[-\frac{\beta}{2} \sum J(|r_i - r_j|) \, \sigma_i \, \sigma_j + \right.$$

$$\left. \sum_i \ln \cosh \left\{ \beta \sum_j J(|r_i - r_j|) \, \sigma_j \right\}, + N \ln 2 \right] \tag{3}$$

where

$$\sigma_k = \langle S_k \rangle.$$

Thus the free energy is

$$F(\{\mathbf{r}_k\}) = (1/2) \sum_i \sum_j J(|\mathbf{r}_i - \mathbf{r}_j|) \, \sigma_i \, \sigma_j$$

$$- (1/\beta) \sum_i \ln \cosh \left\{ \beta \sum_j J(|\mathbf{r}_i - \mathbf{r}_j|) \, \sigma_j \right\}. \tag{4}$$

We have shifted the zero of the free energy to eliminate the last constant term.

The interpretation of σ_k is important. In an ordered system we can define a homogeneous magnetisation per spin $\sigma = \sigma_k = (1/N) \Sigma \sigma_j$. In our system large scale inhomogeneities prevent us from introducing such a concept prior to averaging. We rather visualise the situation as follows: the net result of the random RKKY coupling is that each magnetic ion experiences a local random magnetic field, and then aligns itself to it. We shall call σ_k the *local* magnetisation. A study of local magnetisation necessarily involves the study of the probability densities of the local magnetic field. This interpretation links our work to that of Kaneyoshi (1976), Plefka (1976) and Thouless *et al* (1977). To find the stable phases we differentiate the free energy with respect to the local magnetisations: $\partial F/\partial \sigma_k = 0$. The consistent solution of these set of equations is:

$$\sigma_k = \tanh \left[\beta \sum_i J(|\mathbf{r}_i - \mathbf{r}_k|) \, \sigma_i \right], \tag{5}$$

for all k. If we differentiate this expression for $\partial F/\partial \sigma_k$ again, it is straightforward to show that if $k_B T > J^2(|\mathbf{r}_i - \mathbf{r}_j|)$ then only the solution $\{\sigma_k = 0\}$ of the set of equations (5) provide the local minimum of the free energy. For lower temperatures and low concentrations, any set of solutions which are not all zero provides the local minimum. Note however, that the sign of the various local magnetisations can be either $+$ or $-$. There may be as many local magnetisations which are $+$ as there are $-$. Thus, the total magnetisation may still be zero. Moreover, there may be several different configurations of non-zero solutions, which, at the same temperature may provide local minima for the free energy, and for each configuration the free energy may have the same value. This implies, that rather having a unique stable phase, there are a large number of metastable phases differing in local magnetisation configuration, each separated by an energy barrier. Edwards (1975) has already hinted at such a possibility. The spin glass phase may then consist of domains with differing local magnetisation configurations.

These local minima contrast with the saddle points of the averaged free energy in the Sherrington-Kirkpatrick approach.

Since the local magnetisations are still random, we need to find their probability densities. Let us call $\Sigma K(|\mathbf{r}_i - \mathbf{r}_j|) \, \sigma_j = x$, where $K(R) = J(R)/A$, A being the constant of (1). The probability density is given by the radon transform

$$P(x) = \sum_{r_1} \cdots \sum_{r_N} \Pr(\mathbf{r}_1 .. \mathbf{r}_N) \, \delta [x - \Sigma K(|\mathbf{r}_i - \mathbf{r}_j|) \sigma_j].$$

In the thermodynamic limit:

$$P(x) = \sum_{r_1}^{M} \ldots \sum_{r_N}^{M} (1/M)^N (2\pi)^{-1} \int dk \exp [ik\{x-\Sigma K(|r_i-r_j|)\sigma_j\}].$$

We cannot proceed without further approximations, because of the inextricable statistical interdependence of σ_j and the $J(|r_i-r_j|)$. In the earlier works, since the interactions J were infinitely weak, in the thermodynamic limit σ_j was replaced by $\bar{\sigma}_j$ which is independent of $J(|r_i-r_j|)$. Here, however, we cannot do this; rather we shall follow an argument similar to Klein (1968).

$$P(x) = (2\pi)^{-1} \int dk \exp (ikx) \sum_{r_1} \ldots \sum_{r_N} (1/M)^N$$

$$\int \ldots \int \int \Pi \, dx_i \, \delta \, [x_i - \Sigma K(|r_i-r_j|)\sigma_j] \times$$

$$\exp [-ik \, \Sigma \, K(|r_i-r_j|) \tanh (\beta'x_j)].$$

where $\beta' = \beta A$.

The approximation now entails the replacement of the δ-functions by their configuration averages.

$$\langle \, \Pi_i \, \delta \, [x_i - \Sigma \, K(|r_i-r_j|) \, \sigma_j] \, \rangle = \mathrm{Pr}(\{x_k\}).$$

Let us assume first that there are no *statistical* correlations between the fields at different sites. The effect of such correlations lead to mictomagnetic clusters, and will be studied later. $\mathrm{Pr}(\{x_k\}) = \Pi \, \mathrm{Pr}(x_k)$, and statistical homogeneity demands that $\mathrm{Pr}(x_k) = \mathrm{Pr}(x)$ for all k. This yields an integral equation for $P(x)$.

$$P(x) = (2\pi)^{-1} \int dk \exp (ikx) [1-F(k)/M]^N$$

where

$$F(k) = \sum_{r}^{M} \int dz \, P(z) [1-\exp\{-ikK(R) \tanh \beta'z\}]. \tag{7}$$

In the thermodynamic limit

$$P(x) = (2\pi)^{-1} \int dk \exp [ikx-cF(k)]. \tag{8}$$

The calculation of $F(k)$ as in (7) is tough as it stands. However, one of the properties of the RKKY interaction is that the moments $\Sigma K(R)^n$ decrease very rapidly with n. A specific estimate is discussed in the appendix. The $n=3$ and 4, for example, are five and seven orders of magnitude smaller than the $n=1$ term. Thus in (7) we shall expand the exponential and neglect all moments of $K(R)$ from order three onwards. Such an approximation is certainly physically valid, depending as it is on the *nature* of the decaying RKKY interaction. It is quite distinct from assuming the Js to be Gaussian right from the start. The statistics arises from the random occupation of the sites. Our approximation does not tamper with that picture.

Let us define

$$\langle \sigma \rangle = m = \int dz\, P(z) \tanh(\beta' z), \tag{9}$$

and

$$\langle \sigma^2 \rangle = q = \int dz\, P(z) \tanh^2(\beta' z) \qquad K_0 = \Sigma K(R) \tag{10}$$
$$K_1^2 = \Sigma K^2(R),$$

further assume that $K_0 > 0$ (ferromagnetic case), so that

$$P(x) = (2\pi cqK_1)^{-1/2} \exp[-(x - cK_0 m)^2/(2cK_1 q)]. \tag{11}$$

The Gaussian distribution contrasts with the Lorenzians predicted by Klein (1968) and Klein and Held (1975) and anticipated by Plefka (1976). A Lorenzian would have implied that the second moment of the RKKY is divergent, which is physically untrue. Substituting into (9) we get

$$m = (1/\sqrt{2\pi}) \int dz\, \exp(-Z^2/2) \tanh[\beta(cJ_0 m + \sqrt{cq}\, A_0 z)]$$

$$q = 1 - (1/\sqrt{2\pi}) \int dz\, \exp(-Z^2/2) \operatorname{sech}^2[\beta(cJ_0 m + \sqrt{cq} A_0 z)], \tag{12}$$

where $J_0 = AK_0$ and $A_0 = AK_1$.

These equations are similar to those of the earlier workers, with cJ_0 and cA_0^2 replacing J_0 and J^2. The scaling of both the mean and the variance linearly with the concentration arises naturally within the formalism, and does not require supplementary argument as in the earlier work. It must be emphasised again, that although the results look similar to earlier works, the procedure is throughout physical in its approximations, in its description of the statistics of the randomly placed magnetic ions.

3. The phases

Solutions of (12) describe the possible 'phases' of the system. We note that $m = q = 0$ is a possible solution. We shall describe such a phase as 'paramagnetic'. Near the boundary of this phase in the $c - T$ plane, assuming the phase transition to be of the second order, q and m are both small. Expanding (12) for small m, q

$$m[(1 - cJ_0/k_B T) + c^2 J_0 A_0^2 q/(k_B T)^3 + c^3 J_0^3 m^2/3(k_B T)^3] = 0$$

$$q[1 - cA_0^2/(k_B T)^2 + 2c^2 A_0^4 q/(k_B T)^4 + 4c^3 J_0^2 A_0^2 m^2/(k_B T)^2]$$

$$- c^2 J_0^2 m^2/(k_B T)^2 = 0$$

There are two possible solutions other than $m = q = 0$

$$\text{(i)} \quad m = 0; \quad q = [cA_0^2 - k_B^2 T^2]/(2c^2 A_0^2/k_B^2 T^2). \tag{13}$$

Since $q > 0$, this solution is possible only if $k_B T < \sqrt{c}\, A_0$.

We shall call this phase the 'spin glass' phase, characterised as it is by a short range magnetic ordering without any long range ordering. The paramagnetic-spin glass phase boundary is the parabola $k_B T = \sqrt{c}\, A_0$ and the critical temperature $T_c = \sqrt{c}\, A_0/k_B$. This agrees well with an experimental behaviour which went as $c^{0.6}$. (Mydosh 1974).

(ii) $q = (cJ_0 - k_B T)/[cJ_0(1 - cA_0^2/k_B^3 T^3)(1 + cA_0^2/\{K_B^2 T^2 - cA_0^2\})]$

$$m = (cJ_0 - k_B T)/[c^3 J_0^3(\tfrac{1}{3} + cA_0^2/\{k_B^2 T^2 - cA_0^2\})/k_B^2 T^2]. \tag{14}$$

Again, since both q and $m^2 > 0$, this solution is only possible provided $cJ_0 > k_B T$. The ferromagnetic paramagnetic phase boundary is the straight line $k_B T = cJ_0$. Again, this linear behaviour is in accord with experimental observations (Mydosh 1974).

The bi-critical point (where all the phases meet) is given by $c_b = (A_0/J_0)^2$ and $T_b = c_b J_0/k_B$. In comparison with experimental situation, knowledge of c_b, T_b will determine A_0 and J_0.

Let us now examine the low temperature phases

$$q = 1 - (1/\sqrt{2\pi})(k_B T/cA_0)\exp(-cJ_0^2 m^2/2A_0^2).$$

Thus near $T = 0$, as we approach the spin glass phase from the ferromagnetic side $m \to 0$, but $q \to 1 - k_B T/[\sqrt{2\pi} c A_0]$ which is not small.

Let us expand (12) around small m and $q \simeq 1$, $T \simeq 0°K$.

$$m = \sqrt{(2/\pi)}\,[\sqrt{c}J_0 m/A_0 - \tfrac{2}{3}(cJ_0 m/A_0)^3].$$

One possible solution is $m = 0$, which corresponds to the spin glass side. The other is

$$m^2 = [\sqrt{c}\, J_0/A_0 - \sqrt{(\pi/2)}]/[\tfrac{2}{3}(\sqrt{c}\, J_0/A_0)^3]$$

This solution corresponds to the ferromagnetic phase and is only possible if $\sqrt{c}\, J_0/A_0 > \sqrt{\tfrac{1}{2}\pi} = 1.25$ or $c > 1.25\, c_b = c_0$. If $c < c_0$, there are just not enough spins to induce a long-range order to set in. However, a word of caution here. Although we may assert the existence of c_0, we cannot be certain of its exact value. It interesting to note that a percolation theory prediction of the critical concentration is indeed higher.

4. The antiferromagnetic alloys

In the above analysis we have assumed that $K_0 > 0$. If $K_0 < 0$ then it is easy to see from (14) that $m^2 < 0$, so that it is not possible to have a solution $m \neq 0$ at any temperature. Such a situation might arise if the nearest neighbour distance in the alloys is
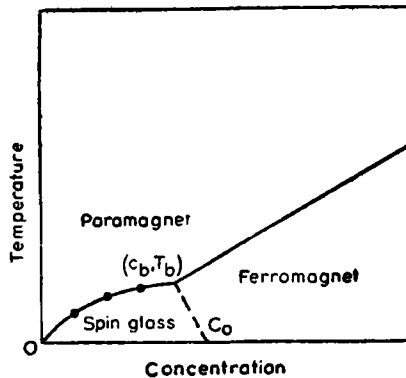
Figure 1. Phase diagram in the temperature-concentration plane.

such that the RKKY interaction tends to align the nearest neighbour spins in opposite directions. The low temperature, high concentration phase we expect is therefore antiferromagnetic rather than a ferromagnet. To examine such a phase we have to use a different order parameter. Taking our cue from the work on pure magnetic metals, let us choose this order parameter to be the staggered magnetisation.

Let us divide the underlying lattice into two sublattices A and B interpenetrating each other. They are such that if a site r_i belongs to one sublattice, its nearest neighbours belong to the other. Therefore,

$$J \left(\left| r_i - r_j \right| \right) = J_{ij}^{(1)} \text{ if } r_i, r_j \text{ lie in different sublattices,}$$

$$= J_{ij}^{(2)} \text{ if they lie in the same sublattice.}$$

In the antiferromagnetic case $J_{ij}^{(1)} < 0$, but $J_{ij}^{(2)} > 0$. It is now easy to check that J_0 is indeed negative. An illustrative example is shown in the appendix.

We now define the local staggered spin operator

$$M_i = S_i, \text{ if } r_i \text{ belongs to sublattice } A$$

$$= -S_i, \text{ if } r_i \text{ belongs to the sublattice } B$$

and a staggered interaction
$J^s \left(\left| r_i - r_j \right| \right) =$ either $-J_{ij}^{(1)}$ or $J_{ij}^{(2)}$ depending on whether the two sites lie in different or the same sublattices. We can now satisfy ourselves that $\Sigma J^s (R) > 0$. The Hamiltonian may be written as

$$H = - (1/2) \sum_{r_i} \sum_{r_j} J_S \left(\left| r_i - r_j \right| \right) M_i M_j. \tag{15}$$

If we compare this with the Hamiltonian of the ferromagnetic system (2), we note that they are identical with M_i, JS_{ij} taking the plane of $S_i J_{ij}$. The entire analysis will then go over unchanged with the two new order parameters: staggered magnetisation $m^s = [\langle M_i \rangle]$ and short-ranged magnetisation $q^s = [\langle M_i \rangle^2]$ and $J_0^s > 0$ replacing J_0

(however $A_0{}^2$ is the same for both the cases). The phase diagram is again identical.   The three phases are: (i) the paramagnetic phase $m^s = q^s = 0$ the (ii) antiferromagnetic phase $m^s \neq 0$, $q^s \neq 0$ and the (iii) antiferromagnetic spin glass phase $m^s = 0$, $q^s \neq 0$.   Since in our present analysis we have taken the short range ordering to be really single site, we cannot distinguish between a ferromagnetic spin glass and an antiferromagnetic spin glass in this phase.   In reality, we expect randomly oriented mictomagnetic clusters and these clusters will have an anti-ferromagnetic ordering.   Note also, that it is not really necessary to have an underlying crystalline lattice.   A randomly distorted network with its sites labelled by r_i can also be treated exactly similarly.   The specific values of J_0, A_0, etc will of course be different, depending on the amount of distortion.   The ferromagnetic case presents no difficulty. In the antiferromagnetic case, there might be an ambiguity in assigning the network sites to the two subnetworks A and B.   For example, if an odd member ring occurs (figure 2), then the site 5 for example can be assigned either to A or to B, if the site 1 is assigned to A.   However, if the distortion is not much, such mismatches are rare and we can still visualise an antiferromagnetic phase, albeit with a few mismatches. Recently Agarwal and Mookerjee (1977) have used this idea of an antiferromagnetic spin glass phase to explain the experimentally observed (Hudgens 1975) magnetic behaviour of amorphous Ge films.   The structure of amorphous Ge films is a four-fold coordinated random network and the spins arising out of the dangling bonds at the microvoids interact with one another via a short ranged antiferromagnetic super-exchange interaction mediated by the localised electronic overlaps.

5. Conclusion

Starting from the physical RKKY interaction and a physical description of the disorder in the alloy, we have shown that under appropriate approximations which are valid in the temperature range of interest, it is possible to set up a mean-field, effective medium theory exhibiting the three possible phases.   These phases are stable, since the local magnetisations which characterise them properly *minimise* the free energy for all random configurations.

It is not certain whether a fluctuating exchange is necessary for the existence of a spin glass phase.   In our theory only parameters which enter the equations are the
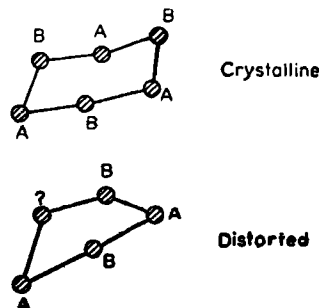

Figure 2.   Odd membered rings in a distorted network, showing ambiguity in subnetwork assignation.

mean, variation and in better approximations only the higher moments of the inter-action. Certain amorphous alloys tend to show behaviour like spin glasses (Korn 1965). The interaction in these are heavily damped. In the amorphous Ge too, the superexchange is not fluctuating. Although the existing unexplained magnetic behaviour of these films could be explained based on a spin glass model, enough experimental data are not available to confirm the model.

Our analysis is that for a random Ising model. A similar analysis will be reported in a subsequent communication, for Heisenberg models. Although the ideas and approximation schemes will be identical, the problem has specific features of its own.

Acknowledgements

Appendix

We want to estimate the spatial moments of the RKKY interaction for a particular model. If the underlying lattice is a simple cubic one, the nth neighbour shells are octahedra with six vertices, twelve edges with $(n-1)$ sites and eight sides with $\frac{1}{2} n^2 - (3/2) n+1$ sites. Thus if R is the nearest neighbour distance and

$$\gamma = 2k_F R$$

$$I_p = \Sigma J^p(R) = \Sigma (\cos^p \gamma)/\gamma^{3p}$$

$$= \sum_{n=1}^{\infty} \left[6 (\cos \gamma n/\gamma n) \, p + 12 \sum_{r=1}^{n-1} \frac{\cos^p \{\gamma (r^2 + (n-r)^2)^{1/2}\}}{\{\gamma (r^2 + (n-r)^2)^{1/2}\}^{3p}} \right.$$

$$\left. + 8 \sum_{r=1}^{n-1} \sum_{s=1}^{n-2} \frac{\cos^p \{\gamma (r^2+s^2 + (n-r-s)^2)^{1/2}\}}{\{\gamma (r^2+s^2 + (n-r-s)^2)^{1/2}\}^{3p}} \right].$$

Table 1. The ratios I_p/I_1 for the two cases ferromagnetic and antiferromagnetic alloys with simple cubic lattice.

	Case I		Case II	
P	Numerical	Approximation	Numerical	Approximation
1	1·0	1·0	−1·0	−1·0
2	0·5 × 10⁻²	0·62 × 10⁻²	0·18 × 10⁻²	0·21 × 10⁻²
3	2·2 × 10⁻⁵	2·34 × 10⁻⁵	−1·95 × 10⁻⁵	−2·0 × 10⁻⁵
4	0·9 × 10⁻⁷	0·9 × 10⁻⁷	3 × 10⁻⁹	3 × 10⁻⁹

This sum can be numerically estimated and the results are shown in table 1. We may also estimate such sums by replacing the complicated octahedral shells by spherical ones. Each cell has for large $n \sim 4\pi n^2$ sites.

$$I_p = 4\pi \sum_{n=1}^{\infty} \cos^p (\gamma n) \gamma^{-3p} n^{2-3p}.$$

Let us choose two cases $\gamma = 2\pi$ and $\gamma = 3\pi$. Two real alloys $\underline{Au}$ Fe and $\underline{Cu}$ Mn have almost near these values (respectively 6·3 and 9·2). The former exhibits a ferromagnetic phase, while the latter an antiferromagnetic phase at low temperature, high concentration regions.

In case 1, $I_p = 2(2\pi)^{3p-1} \zeta (3p-2)$

$\zeta(r)$ being the Riemann zeta function.

Similarly, for case II, $I_p = (-1)^p 2(2\pi)^{3p-1} Z(3p-2)$

$Z(r)$ being either the zeta or the eta functions according as p is even or odd. The table shows that for a simple cubic such an approximation overestimates the number and distance of the sites in the nearer shells. But for the higher moments the approximations are good.

Further the ratios I_p/I_1 very rapidly decrease, so that our approximation of ignoring I_p for $p > 3$ is not bad.

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