

ments of the sparking potential in oxygen were made in chamber B at a base pressure of 10^{-2} Torr and the values are in excellent agreement with those found in chamber A at a base pressure of 10^{-2} Torr; the values are in excellent agreement with those found in chamber A at a base pressure of 10^{-9} Torr. This indicates that the sparking potential of oxygen is unaffected by the presence of the ordinary background impurities up to partial pressures of about 10^{-2} Torr. Also, in all the measurements it was found that the sparking potential is independent of whether or not the cathode was irradiated with uv light, indicating that the sparking

potential is independent of the magnitude of I_0 , at least up to values of $I_0=10^{-11}$ A.

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Critical Percolation Probabilities by Series Methods

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Series estimates of the critical percolation probabilities for the "bond problem" and the "site problem" are presented for two- and three-dimensional lattices. Good agreement with the Monte Carlo estimates of Frisch *et al.* and of Dean is obtained. The series method gives information on the critical behavior of the mean cluster size and it is found that there is a much sharper growth of large clusters in two dimensions than in three dimensions as the critical concentration is approached from below.

1. INTRODUCTION

RECENTLY, numerical estimates for the critical probabilities that arise in two percolation problems of physical interest have been given by a number of authors.¹⁻⁵ In the *bond problem*, described by Broadbent and Hammersley,⁶ one studies percolation through a "random maze" of paths (bonds) which are "open" with probability p and "blocked" with probability $q=1-p$. We shall treat the case when the "maze" is an infinite crystal lattice. In the *site problem* introduced by Domb,⁷ one supposes the sites of the lattice to be occupied with probability p and vacant with probability q . Site problems are more general since every bond problem can be made isomorphic with a site problem on a suitably chosen *covering lattice* by the bond-to-site transformation.⁸ A very complete discussion of the underlying

physical problems is given in the references quoted above.

Percolation problems on infinite lattices are characterized by the occurrence of a critical probability p_c , above which there is a nonzero probability of a site being a member of an infinite "cluster" of connected sites. Estimates for p_c for the more important crystal lattices have been obtained by Monte Carlo methods¹⁻⁴ and from exact series expansions.⁵ It is the object of the present paper to develop the series method.

It was suggested by Domb⁹ that the method of exact series expansions could be applied to a study of percolation problems. In particular the critical probability could be investigated by expanding the mean size of finite clusters $S(p)$ as a power series in p . A series of positive terms results and hence the radius of convergence of the expansion can be identified with the critical percolation probability p_c . Subsequently, series developments for $S(p)$ were published and examined briefly by Domb and Sykes⁵ who also suggested that the mean cluster size in the critical region could be investigated by series methods. Domb and Sykes found the behavior of the coefficients not altogether smooth, particularly in

¹ H. L. Frisch, J. M. Hammersley, and D. J. A. Welsh, *Phys. Rev.* **126**, 949 (1962).

² V. A. Vyssotsky, S. B. Gordon, H. L. Frisch, and J. M. Hammersley, *Phys. Rev.* **123**, 1566 (1961).

³ H. L. Frisch, E. Sonnenblick, V. A. Vyssotsky, and J. M. Hammersley, *Phys. Rev.* **124**, 1021 (1961).

⁴ P. Dean, *Proc. Cambridge Phil. Soc.* **59**, 397 (1963).

⁵ C. Domb and M. F. Sykes, *Phys. Rev.* **122**, 77 (1961).

⁶ S. R. Broadbent and J. M. Hammersley, *Proc. Cambridge Phil. Soc.* **53**, 629 (1957).

⁷ C. Domb, *Nature* **184**, 509 (1959).

⁸ J. W. Essam and M. E. Fisher, *J. Math. Phys.* **2**, 609 (1961).

⁹ Conference of the Physical Society on "Fluctuation Phenomena and Stochastic Processes" held at Birkbeck College, London, England, 19-20 March, 1959, and briefly reported in Ref. 7.

the initial stages, and this makes extrapolation difficult. The successful exploitation of information in series expansion form demands an accurate knowledge of their radius of convergence and for this reason the location of the critical concentration p_c is of prime importance.

We have derived extra coefficients for the expansions of $S(p)$ by applying the theory of cooperative phenomena in crystals¹⁰ to these problems. Details of the various devices that have been employed will be published separately and we shall confine our present treatment to stating only those aspects of the technique that are germane to our present purpose of extrapolation. If the mean size of finite clusters be expanded in powers of p

$$S(p) = \sum_n a_n p^n, \quad (1.1)$$

it can be shown that the successive coefficients a_n are equivalent to an enumeration problem on the lattice considered, and in particular the n th coefficient for the bond problem can be expressed as a linear sum of high-temperature lattice constants of n lines. For a precise treatment of lattice constants reference should be made to the review by Domb.¹⁰ Expansions whose coefficients depend on lattice constant data also occur in two other physical problems; the high-temperature expansion for the susceptibility of the Ising model^{11,12} in powers of the usual counting variable $v = \tanh K$

$$\chi(v) = \sum_n b_n v^n, \quad (1.2)$$

and the non-self-intersecting chain generating function

$$C(x) = \sum_n c_n x^n \quad (1.3)$$

which arises in the simple lattice model of a polymer.¹³⁻¹⁵ Expansions of this type have been much studied and an account of their salient features has been given by Domb and Sykes.¹⁶ It seems well established that we may write

$$b_n \sim n^g v_c^{-n}, \quad (1.4)$$

$$c_n \sim n^h x_c^{-n}, \quad (1.5)$$

where in (1.4) v_c is related to the Curie temperature through the relation $v_c = \tanh J/kT_c$ and in (1.5) x_c is the inverse of the limiting or critical attrition parameter, μ .

The indices g, h are found to be only dependent on the dimensionality (D) of the lattice considered and

$$\begin{aligned} g &= \frac{3}{4} & \text{for } D=2, \\ g &= \frac{1}{4} & \text{for } D=3, \\ h &= \frac{1}{3} & \text{for } D=2, \\ h &= \frac{1}{6} & \text{for } D=3. \end{aligned} \quad (1.6)$$

Confidence in the results (1.6) is based partly on the existence of exact results for some two-dimensional lattices in the Ising case¹⁷ and partly on the fact that the same parameters g, h give satisfactory results for lattices such as the face-centered cubic and diamond lattice which differ greatly in structure.¹⁸ For loose-packed lattices it proves necessary to average odd and even terms to smooth out an oscillation which, however, decays with increasing n . In view of the close similarity in the underlying enumeration problems it seems a reasonable hypothesis that we may write in (1.1)

$$a_n \sim n^j p_c^{-n}, \quad (1.7)$$

and this would imply a singularity in the mean cluster size $S(p)$ of the form $1/(p_c - p)^{j+1}$. The relatively poor behavior of the early terms already noticed is aggravated by the fact that loose-packed lattices exhibit a persistent even-odd oscillation which does not fall off as n increases. Our object has been to seek a suitable smoothing procedure and an over-all asymptotic behavior which is only dimensionality dependent. As a result of extensive numerical experiments, including the Pade-approximant^{19,20} method, we have been led to quite a simple result of the form (1.7) and we shall give the evidence in this paper and use it to estimate p_c for the more usual crystal lattices. We shall conclude that we may write in (1.7)

$$\begin{aligned} j &= 11/8 & \text{for } D=2, \\ j &= 11/16 & \text{for } D=3. \end{aligned} \quad (1.8)$$

Recently, exact values for the critical probabilities of a number of two dimensional problems have been given²¹ and this enables the validity of series expansion extrapolations to be gauged. Once the index j in (1.7) is fixed, more accurate estimates of p_c can be made.

2. TRIANGULAR LATTICE

As a detailed example we study the triangular lattice since the critical probability for the site ($p_c^S = \frac{1}{2}$) and the bond ($p_c^B = 0.347296$) problem are known exactly.²¹ Denoting the site and the bond problem by superscripts S and B we find

$$\begin{aligned} S(p)^S &= 1 + 6p + 18p^2 + 48p^3 + 126p^4 + 300p^5 + 750p^6 \\ &\quad + 1686p^7 + 4074p^8 + 8868p^9 + \dots, \end{aligned} \quad (2.1)$$

$$\begin{aligned} S(p)^B &= 1 + 10p + 46p^2 + 186p^3 + 706p^4 + 2568p^5 + 9004p^6 \\ &\quad + 30894p^7 + 103832p^8 + 343006p^9 + \dots \end{aligned} \quad (2.2)$$

The successive ratios $\rho_n^* = a_n/a_{n-1}$ in the bond series can be studied as they stand while the corresponding site ratios oscillate somewhat. To maintain a consistent

¹⁰ C. Domb, *Advan. Phys.* **9**, 149 (1960).

¹¹ T. Oguchi, *J. Phys. Soc. Japan* **6**, 31 (1951).

¹² M. F. Sykes, *J. Math. Phys.* **2**, 52 (1961).

¹³ M. E. Fisher and M. F. Sykes, *Phys. Rev.* **114**, 45 (1959).

¹⁴ M. F. Sykes, *J. Chem. Phys.* **39**, 410 (1963).

¹⁵ B. J. Hiley and M. F. Sykes, *J. Chem. Phys.* **34**, 1531 (1961).

¹⁶ C. Domb and M. F. Sykes, *J. Math. Phys.* **2**, 63 (1961).

¹⁷ M. E. Fisher, *Physica* **25**, 521 (1959).

¹⁸ J. W. Essam and M. F. Sykes, *Physica* **29**, 378 (1963).

¹⁹ G. A. Baker, Jr., *Phys. Rev.* **124**, 768 (1961).

²⁰ J. W. Essam and M. E. Fisher, *J. Chem. Phys.* **38**, 802 (1963).

²¹ M. F. Sykes and J. W. Essam, *Phys. Rev. Letters* **10**, 1 (1963).

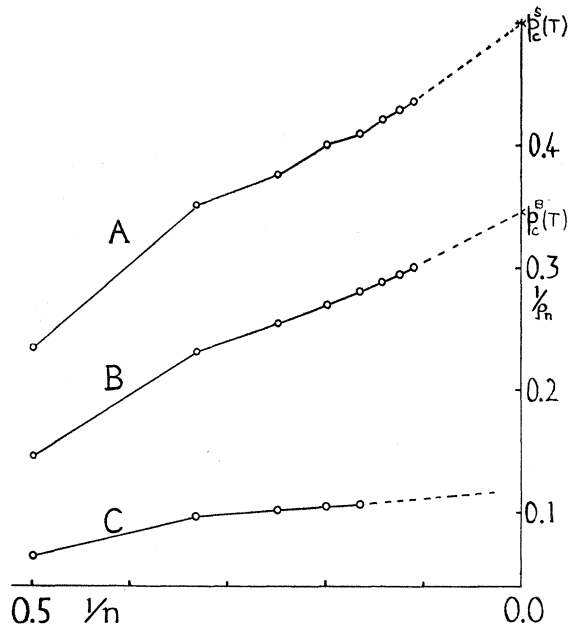


FIG. 1. Successive estimates for the critical percolation probability ($1/\rho_n \rightarrow p_c$) plotted against $1/n$. (A) Triangular lattice site problem. Exact limit $p_c^S(T)=0.5$. (B) Triangular lattice bond problem. Exact limit $p_c^B(T)=0.3473$. (C) Face-centered cubic bond problem.

treatment throughout, we shall, therefore, define the ratios of alternate terms by

$$\rho_n = (a_n/a_{n-2})^{1/2} \tag{2.3}$$

and always work with this quantity. In Fig. 1 we plot the successive $1/\rho_n$ for the bond and site problem against $1/n$. It will be seen that the over-all behavior is tending to linearity and that the true critical values are well indicated. We shall assume that

$$\lim_{n \rightarrow \infty} \rho_n = \rho = 1/p_c. \tag{2.4}$$

We next evaluate the successive approximations to the index j defined by

$$j_n = n(\rho_n - \rho)/\rho \tag{2.5}$$

and present these in Table I. The entries are plotted in Fig. 2 against $1/n$. Convergence is seen to be relatively slow, but the data are consistent with the assumption that the two sequences have a common

TABLE I. Estimates for j for the site and bond problems on the triangular lattice.

n	j_n sites	j_n bonds
4	1.2915	1.4423
5	1.2500	1.4523
6	1.3193	1.4416
7	1.2973	1.4321
8	1.3227	1.4349
9	1.3204	1.4149

TABLE II. Bond problem. Two-dimensional lattices: successive estimates for the critical probability $\beta_n = (n+j)/n\rho_n$, $j=11/8$.

n	Triangular	Simple quadratic	Honeycomb
4	0.3430	0.5079	0.6719
5	0.3431	0.5100	0.6940
6	0.3442	0.4998	0.6953
7	0.3449	0.5074	0.6517
8	0.3451	0.4982	0.6471
9	0.3460	0.5057	0.6997
10		0.4981	0.6567
11		0.5048	0.6307
12			0.6784
13			0.6755
14			0.6265
p_c exact	0.3473	0.5000	0.6527
Monte Carlo estimates ^a	0.341 ± 0.011	0.493 ± 0.013	0.640 ± 0.018
Monte Carlo estimates ^b	0.329 ± 0.021	0.492 ± 0.011	0.635 ± 0.020

^a See Ref. 2.
^b See Ref. 4.

limit, close to $1.375 = 11/8$. Because of the simple nature of this result we shall make the hypothesis that j is exactly $11/8$, although the numerical evidence is not conclusive on this point.

As remarked in the Introduction, any bond problem can be made isomorphic with a site problem on a suitably chosen covering lattice. Because of this we have sought a common limit for the indices for the site and bond problem on the triangular lattice. It may be objected that, in general, the covering lattice of a two-dimensional bond problem is a two-dimensional lattice with crossing bonds and the evidence for the existence of an index which is only dimensionality dependent comes from lattices without crossing bonds. However, the covering lattice for the honeycomb lattice (bond problem) is the Kagomé lattice (site problem), and the indices for these two problems must be identical. It follows that if our hypothesis of indices for the site and bond problem is viable then these indices must be equal. We would further suggest that the evidence of this section supports the view that the indices found for the Ising problem and quoted in (1.6) will be valid for

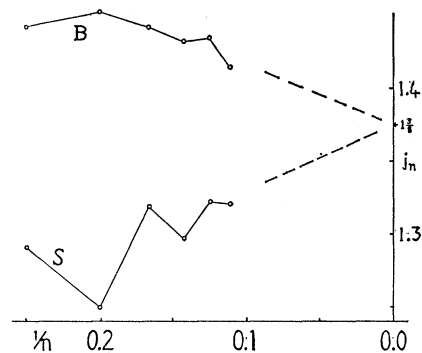


FIG. 2. Successive estimates j_n for the index for the site (S) and bond (B) problems on the triangular lattice plotted against $1/n$.

TABLE III. Site problem. Two-dimensional lattices: successive estimates for the critical probability $\beta_n = (n+j)/n\rho_n$, $j=11/8$.

n	Honeycomb matching lattice	Simple quadratic matching lattice	Triangular	Simple quadratic	Honeycomb
3	0.2860	0.3969	0.5156	0.5954	0.6889
4	0.2952	0.4075	0.5079	0.6455	0.7292
5	0.2987	0.4054	0.5100	0.6010	0.6719
6		0.4066	0.5038	0.5922	0.7689
7		0.4073	0.5047	0.6126	0.7774
8			0.5028	0.6037	0.6908
9			0.5026	0.5986	0.7268
10				0.5886	0.6904
11					0.6789
12					0.7649
Limits	0.30 ± 0.01	0.410 ± 0.010	0.5000 (exact)	0.590 ± 0.010	0.70 ± 0.01
Monte Carlo ^a			0.493 ± 0.018	0.581 ± 0.015	0.688 ± 0.017
Monte Carlo ^b		0.387 ± 0.014	0.486 ± 0.017	0.580 ± 0.018	0.688 ± 0.015

^a See Ref. 3.
^b See Ref. 4.

lattices with crossing bonds. This is equivalent to the statement that "second neighbor" interactions or longer range forces do not affect qualitatively the critical behavior of the Ising model.

3. CRITICAL PROBABILITIES FOR TWO-DIMENSIONAL LATTICES

Following closely the procedure given by Domb and Sykes¹⁶ we now estimate the radius of convergence of the expansions (2.1) and (2.2) by calculating

$$\beta_n = (n+j)/n\rho_n \tag{3.1}$$

for $j=1.375$. The quantity β_n should converge to p_c with almost negligible slope. Even if the estimate for j is incorrect β_n must still converge to p_c . In Table II we give the β_n for the bond problem on the triangular, simple quadratic, and honeycomb lattices. The expansions on which this and subsequent tables are based are given in the Appendix. The values for the triangular lattice are converging smoothly and the last entry is within $\frac{1}{2}\%$ of the limit. Graphical extrapolation of the entries against $1/n$ could be used to improve this if p_c were not known. The simple quadratic estimates oscillate about the exact limit of $\frac{1}{2}$ with a final amplitude of 1% , the average of the last pair being 0.5014. The honeycomb lattice yields a comparatively poor sequence. The slow convergence of expansions for this very loosely packed lattice is well known.²² The average of the last four entries (0.6528) is close to the true limit.

In Table III we give corresponding estimates for the site problem on a number of two-dimensional lattices. For the triangular, the exact value of $p_c = \frac{1}{2}$ is closely indicated, the last entry being within $\frac{1}{2}\%$. For the simple quadratic lattice the exact value has not been given but

it is possible to show²³ that if $p_c(\text{S.Q.})$ is the limit for this lattice and $p_c(\text{S.Q.M.})$ is the limit for the simple quadratic lattice with second neighbors, which we shall call the simple quadratic matching lattice, then

$$p_c(\text{S.Q.}) + p_c(\text{S.Q.M.}) = 1. \tag{3.2}$$

As might be expected from the behavior of the bond series for the triangular and honeycomb lattices which also form a matching pair, the lattice with the lower critical value is the more rapidly convergent. By graphical extrapolation for the simple quadratic we estimate

$$p_c(\text{S.Q.}) = 0.580 \pm 0.020, \tag{3.3}$$

but this can be improved by extrapolating instead the matching lattice as

$$p_c(\text{S.Q.M.}) = 0.410 \pm 0.010 \tag{3.4}$$

and using the relation (3.2) to give

$$p_c(\text{S.Q.}) = 0.590 \pm 0.010. \tag{3.5}$$

In a similar way we have estimated the limit for the honeycomb lattice from its matching lattice which is the honeycomb lattice with second and third neighbors.

The final estimates in Tables II and III would seem consistently better than the Monte Carlo estimates of Refs. 2, 3, and 4 for all but the most loosely packed honeycomb lattice although the differences are very small in all cases. As noticed by these authors the Monte Carlo estimates are slightly biased towards too small a value. For example, for the matching pair of the site problem on the simple quadratic lattice and the corresponding second neighbor lattice Dean quotes 0.580 ± 0.018 and 0.387 ± 0.014 , respectively. If we take the

²² M. F. Sykes and M. E. Fisher, *Physica* **28**, 919 (1962).

²³ Unpublished. The result can be established by the methods outlined in Ref. 21.

TABLE IV. Bond problem. Three-dimensional lattices: successive estimates for the critical percolation probability $\beta_n = (n+j)/n\rho_n$, $j=11/16$.

n	fcc	bcc	sc	Diamond
3	0.11 898	0.1804	0.2520	0.4097
4	0.11 891	0.1790	0.2483	0.3906
5	0.11 897	0.1793	0.2482	0.3914
6	0.11 902	0.1789	0.2477	0.3932
7		0.1791	0.2478	0.3892
8		0.1788	0.2473	0.3888
9			0.2476	0.3900
10				0.3881
11				0.3888
Limit	0.119±0.002	0.178±0.005	0.247±0.005	0.388±0.005
Monte Carlo*	0.125±0.005		0.254±0.013	0.390±0.011

* See Ref. 2.

sum as 0.967 ± 0.032 the true value (1.00) falls just outside those limits.

4. CRITICAL PROBABILITIES FOR THREE-DIMENSIONAL LATTICES

No exact result has been given for the critical probability of any three-dimensional lattice. Proceeding in a manner analogous to Sec. 2 we examine the expansion for the bond problem on the face-centered cubic lattice since this is the most rapidly convergent of the usual three-dimensional lattices. We find

$$S(p)^B = 1 + 22p + 234p^2 + 2348p^3 + 22726p^4 + 214642p^5 + 1993002p^6 + \dots \quad (4.1)$$

Defining the ratios ρ_n as in the previous section, we calculate the successive linear intercepts ρ_n' given by

$$\rho_n' = n\rho_n - (n-1)\rho_{n-1}. \quad (4.2)$$

(The quantities $1/\rho_n$ are plotted in Fig. 1. It is seen that their behavior is again linear but with a different slope.) This yields

$$\begin{aligned} \rho_4' &= 8.42\ 706, \\ \rho_5' &= 8.38\ 583, \\ \rho_6' &= 8.38\ 246, \end{aligned} \quad (4.3)$$

and we take $\rho = 8.38$ as a first approximation to the limit. Using this value we obtain as estimates for the slope

$$\begin{aligned} j_4 &= 0.7040, \\ j_5 &= 0.7047, \\ j_6 &= 0.7050. \end{aligned} \quad (4.4)$$

These estimates are of course dependent on our choice of ρ , but if they are compared with the corresponding estimates for the triangular lattice in the second column of Table I it appears that the j value for the face-centered cubic is almost exactly a half that found in two dimensions. We therefore make the hypothesis that j is $11/16$ for the three-dimensional problem and give in Tables IV and V the corresponding β_n for the bond and site problems on the diamond, face-centered cubic,

body-centered cubic, and simple cubic lattice. It will be seen that our choice of j has effectively removed the "slope" from all these eight series of estimates and this confirms the over-all correctness of the hypothesis that this is primarily dimensionality dependent. The extrapolated limits are consistently lower than the Monte Carlo estimates of Frisch *et al.* by an amount of the order of the standard error quoted by these authors.

5. GENERAL CONCLUSIONS

We have found that exact series expansions for the mean size of finite clusters can be used to estimate the critical percolation probability of an infinite lattice and we have proposed a formal procedure for this purpose. In two dimensions the extrapolated values are in good agreement with the exact values where known and are very slightly higher than Monte Carlo estimates. It seems that published Monte-Carlo estimates for two-dimensional lattices are consistently *too low* by about their standard error. In three dimensions the series estimates are consistently *below* the Monte Carlo estimates by an order of magnitude of about the standard error. We suggest that the different critical behavior of two- and three-dimensional structures introduces a bias into the Monte Carlo estimates. In the series method the difference in j should compensate any bias properly.

We have made the hypothesis that in two and three dimensions the appropriate index (j) is $11/8$ and $11/16$, respectively, and have adopted these values in our extrapolation procedure. We stress that we have adopted these appealingly simple fractions as a working rule only, and that we have not found the evidence conclusive. We think the evidence presented supports the view that percolation problems in two and three dimensions give rise to a dimensionality-dependent index j and that

$$\begin{aligned} j &= 1.375 \pm 0.03 \quad \text{for } D=2, \\ j &= 0.6875 \pm 0.05 \quad \text{for } D=3. \end{aligned} \quad (5.1)$$

Near $p = p_c$ the mean size of clusters will thus exhibit a singularity of the form

$$1/(p_c - p)^{i+1}, \quad (5.2)$$

TABLE V. Site problem. Three-dimensional lattices: successive estimates for the critical percolation probability $\beta_n = (n+j)/n\rho_n$, $j=11/16$.

n	fcc	bcc	sc	Diamond
3	0.1897	0.2208	0.2820	0.4097
4	0.1957	0.2498	0.3067	0.3906
5	0.1950	0.2375	0.3093	0.4200
6		0.2397	0.3161	0.4353
7		0.2458	0.3082	0.4369
8			0.3071	0.4290
9				0.4206
10				0.4270
Limit	0.195±0.005	0.243±0.010	0.307±0.010	0.425±0.015
Monte Carlo*	0.199±0.008		0.325±0.023	0.436±0.012

* See Ref. 3.

or $1/(p_c - p)^{2.375}$ in two dimensions and $1/(p_c - p)^{1.6875}$ in three dimensions. There is, therefore, a much sharper growth of large clusters in two dimensions than in three dimensions as the critical concentration is approached from below. This latter conclusion is unaffected by relatively small uncertainties in the exact value of j appropriate to any individual case.

The mean size of finite clusters for $p > p_c$, and also the probability of a site or bond being a member of an infinite cluster, can also be expanded as power series and we shall treat this problem in a subsequent paper.

ACKNOWLEDGMENTS

The authors are indebted to Professor C. Domb for suggesting the problem and first explaining to them how suitable power series could be derived. We are also grateful to Dr. M. E. Fisher for useful criticism and discussions and to D. S. Gaunt for his assistance in deriving data.

APPENDIX

Coefficients for expansion of $S(p)$ for bond and site problems. We quote the successive a_r in tabular form [$S(p) = 1 + \sum_r a_r p^r$].

Two-dimensional bond problems.

r	Triangular	Plane square	Honeycomb
1	10	6	4
2	46	18	8
3	186	48	16
4	706	126	32
5	2568	300	54
6	9004	762	100
7	30 894	1668	182
8	103 832	4216	328
9	343 006	8668	494
10		21 988	984
11		43 058	1572
12			2656
13			4212
14			8162

Two-dimensional site problems

r	Honeycomb matching lattice	Plane square matching lattice	Triangular	Plane square	Honeycomb
1	12	8	6	4	3
2	66	32	18	12	6
3	312	108	48	24	12
4	1368	348	126	52	24
5	5685	1068	300	108	33
6		3180	750	224	60
7		9216	1686	412	99
8			4074	844	156
9			8868	1528	276
10				3152	438
11					597

Three-dimensional bond problems

r	Face-centered cubic	Body-centered cubic	Simple cubic	Diamond
1	22	14	10	6
2	234	98	50	18
3	2348	650	238	54
4	22 726	4202	1114	162
5	214 642	26162	4998	456
6	1 993 002	163 154	22 562	1302
7		984 104	98 174	3630
8		6 015 512	434 894	10 158
9			1 855 346	27 648
10				77 022
11				206 508

Three-dimensional site problems

r	Face-centered cubic	Body-centered cubic	Simple cubic	Diamond
1	12	8	6	4
2	84	56	30	12
3	504	248	114	36
4	3012	1232	438	108
5	17 142	5690	1542	264
6		26 636	5754	708
7		113 552	19 574	1668
8			71 958	4536
9				10 926
10				28 416