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Series expansion for rectilinear polymers on the square lattice

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Abstract. A number of ways in which a quadratic lattice can be fully covered with given numbers of rectilinear r-mers($r \geqslant 2$) is estimated by a combinatorial method involving a series expansion.

1. Introduction

One of the simplest, yet unsolved, problems in lattice statistics is the pure r-mer problem in which each site of a lattice is singly occupied by one element of a rectilinear r-mer molecule. The pure r-mer problem is characterised by the residual entropy or 'molecular freedom' per r-mer ϕ_r, defined so that the number of arrangements of n r-mers on a lattice of $n \times r$ sites asymptotically $(\phi_r)^n$. The exact solution for the case of dimers was obtained by Fisher (1961) and Kasteleyn (1961) for the square lattice and subsequently for other two-dimensional lattices (Kasteleyn 1963). No rigorous treatments of the general r-mer problem have yet been given.

The Bethe approximation on a lattice of coordination number c (Van Craen 1970) leads to

$$\phi_r = (cr/2)[1-(2/cr)(r-1)]^{cr/2-r+1} \tag{1.1}$$

and becomes invalid as r increases (ϕ_r is less than unity already for $r > 3$ in the case of a square lattice). There are enough numerical data, provided by the matrix method of Kramers and Wannier (Van Craen 1970), the Kikuchi method (Kaye and Burley 1977), and series expansions (Van Craen and Bellemans 1972), to obtain fair estimates of the exact solution of the trimer problem.

Recently Kowalsky and Priezzhev (1978) and Gagunashvili and Priezzhev (1978) have investigated rigorously lower and upper bounds of ϕ_r for arbitrary $r \geqslant 2$. Their results are summarised in the following three inequalities:

$$\phi_r \leqslant (r/2)^{1/r} \exp(4G/\pi r) \qquad \text{for } r \text{ even} \tag{1.2}$$

$$\phi_r \leqslant \left(\frac{r-1}{2}\right)^{1/r} \exp\left\{\frac{1}{\pi r}\int_0^{\pi} \operatorname{arch}[2r/(r-1)-\cos\phi]\, d\phi\right\} \qquad \text{for } r \text{ odd} \tag{1.3}$$

$$\phi_r \geqslant \exp(4G/\pi r) \tag{1.4}$$

where $G = 0{\cdot}915\,965\ldots$ (Catalan's constant).

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In the present paper we develop a method of approaching the problem which is an extension of these works. This method, based on the combinatorial principle of inclusion and exclusion, provides a series-expansion technique for estimating the molecular freedom per r-mer for arbitrary $r \geq 2$.

2. Rectilinear polymers on the superlattice

Consider a planar quadratic $mr \times nr$ lattice to which one can attach rectilinear r-mers in such a way that every r-mer occupies r lattice points and the lattice is fully occupied by r-mers. We denote the lattice points by (x, y) and define the points of a quadratic $m \times n$ *superlattice* as points with coordinates (X, Y) which obey

$$X(\mathrm{mod}\ r) = 0, \qquad Y(\mathrm{mod}\ r) = 0.$$

To estimate ϕ_r we use the following propositions (Kowalsky and Priezzhev 1978):

Proposition 1. Let $r(mn)$ points of the lattice be occupied by mn non-overlapping r-mers, arranged on the superlattice so that each site of the superlattice accommodates one element of the r-mer. Then the rest of the points may be covered with r-mers in not more than one manner.

So any arbitrary configuration of r-mers is defined by an arrangement of mn r-mers on the superlattice. There are $2r$ different ways in which a superlattice site may be occupied and consequently there are altogether $(2r)^{mn}$ possibilities. Many of them, however, are unacceptable because of incompatibilities between arrangements of different r-mers on the superlattice.

Let us consider the reasons for which an r-mer configuration C on the superlattice may be unacceptable. The simplest of them is the overlap of r-mers covering neighbouring superlattice points. To clarify other cases we introduce some auxiliary notions.

By *reduced coordinates* of the point (x, y) we understand the pair of integers $[i, j]$ defined by

$$i = x(\mathrm{mod}\ r) \qquad i \in (0, 1, \ldots, r-1)$$

$$j = y(\mathrm{mod}\ r) \qquad j \in (0, 1, \ldots, r-1).$$

Let C be a configuration of r-mers on the superlattice and B(C) be the set of superlattice bonds containing the lattice points covered by r-mers from C. Each bond appears in B(C) 0, 1 or 2 times if there are 0, 1 or 2 r-mers covering this bond. A *graph* is defined by the collection of superlattice sites and the collection of bonds B(C). A *cycle* of this graph is a collection of bonds of the form $p_1p_2, p_2p_3, \ldots, p_{k-1}p_1$ where p_ip_j denotes the bond joining superlattice points p_i and p_j, and all points $p_1, p_2, \ldots, p_{k-1}$ are distinct. A cycle is *closed* with respect to the reduced coordinates $[i, j]$, $i, j \neq 0$ if all points of the basic $mr \times nr$ lattice belonging to its bonds and having reduced coordinates $[i, 0]$, $[0, j]$ are covered by r-mers from C. A cycle p_1p_2, p_2p_1 resulting from two neighbouring r-mers from C overlapping in one or more basic-lattice points is closed, too, with respect to the reduced coordinates of these points. We call any closed cycle a *contour* and we use $g(i_1, j_1; i_2, j_2; \ldots; i_s, j_s)$ to denote the contour closed with respect to the coordinates $[i_1, j_1], [i_2, j_2], \ldots, [i_s, j_s]$, or g if the values of these coordinates are not essential. We will say that the configuration C generates the contour g. Note that different configurations can generate the same contour and several contours can correspond to one cycle.

Proposition 2. (Gagunashvili and Priezzhev 1978.) If the r-mer configuration C on the superlattice generates at least one contour g, then a densely packed configuration on the basic lattice containing C does not exist.

Thus, in order to get an explicit expression for ϕ_r, we need to exclude, from the total number $(2r)^{nm}$ of r-mer configurations on the superlattice, those generating contours. Consider the set of all distinct contours $\{g_s\}$, $s = 1$ to k, where k is the maximum number of contours for a given lattice. Let P be the total number of r-mer configurations on the superlattice. Let P_i be the number of configurations generating the contour g_i and $P_{i_1,i_2,\ldots,i_s}$ the number generating the contours $g_{i_1}, g_{i_2}, \ldots, g_{i_s}$. Then by the principle of inclusion and exclusion the number of configurations P_0 generating none of the contours is given by

$$P_0 = P - \sum_i P_i + \sum_{i_1<i_2} P_{i_1,i_2} + \ldots + (-1)^s \sum_{i_1<i_2<\ldots<i_s} P_{i_1,\ldots,i_s} + \ldots + (-1)^k P_{1,2,\ldots,k}. \quad (2.1)$$

One may take on trust that excluding the configurations generating contours exhausts the set of all unacceptable configurations. At least we have the following statement (Priezzhev 1976):

Proposition 3. In the case $r = 2$, P_0 is the number of all possible dense and non-overlapping arrangements of dimers on the square lattice.

We conjecture that proposition 3 is valid for all $r \geqslant 2$.

If the conjecture holds, we obtain the expression for the molecular freedom of r-mers on the basic $mr \times nr$ lattice:

$$\phi_r = (P_0)^{1/rmn}. \quad (2.2)$$

In the opposite case the right-hand side of equation (2.2) is an upper bound of ϕ_r.

3. Derivation of the series expansion

Let G_s be a set of contours $g_1, g_2, \ldots, g_\nu$ generated by an r-mer configuration on the superlattice. The index s denotes the number of superlattice points belonging to contours from G_s; $\nu(G_s)$ denotes the number of contours in the set G_s. Note that one may arrange an r-mer on each of $mn - s$ superlattice points which do not belong to G_s in $2r$ independent ways. We define $W(G_s)$ by

$$\sum_C{}' (-1)^{\nu(G_s)} = (2r)^{mn-s} W(G_s) \quad (3.1)$$

where the prime denotes summation over configurations C generating the set G_s. By definition $|W(G_s)|$ is the number of arrangements of s r-mers on the s superlattice sites which lead to the set of contours G_s. According to equation (2.1) we have, in the notation introduced above,

$$P_0 = P + \sum_{G_2} \sum_C{}' (-1)^{\nu(G_2)} + \sum_{G_3} \sum_C{}' (-1)^{\nu(G_3)} + \ldots + \sum_{G_{mn}} \sum_C{}' (-1)^{\nu(G_{mn})} \quad (3.2)$$

$$= (2r)^{mn} + (2r)^{mn-2} \sum_{G_2} W(G_2) + (2r)^{mn-3} \sum_{G_3} W(G_3) + \ldots + \sum_{G_{mn}} W(G_{mn}).$$

We define the generating function

$$\Lambda_N(x) = (2r)^N\left(1 + \sum_{s=2}^{N} \omega_N(s)x^s\right) \tag{3.3}$$

where $N = mn$ and

$$\omega_N(s) = \sum_{G_s} W(G_s).$$

In the thermodynamic limit we obtain

$$\Lambda(x) = \lim_{N\to\infty} (\Lambda_N)^{1/N} = 2r\left(1 + \sum_{s=2}^{\infty} \omega(s)x^s\right) \tag{3.4}$$

where it can be shown that

$$\omega(s) = \omega_N(s)|_{N=1}.$$

From equations (2.2), (3.2), (3.3) and (3.4) it follows that

$$\phi_r = \left[\Lambda\left(\frac{1}{2r}\right)\right]^{1/r} = (2r)^{1/r}\left[1 + \sum_{s=2}^{\infty} \omega(s)\left(\frac{1}{2r}\right)^s\right]^{1/r}. \tag{3.5}$$

From the boundedness of ϕ_r for each fixed r the convergence of the series in equation (3.5) follows; so $\omega(s)/(r^s 2^s) \to 0$ for $s \to \infty$. We shall see below that the convergence of the series is rapid enough to estimate ϕ_r using the first few terms of the series.

4. Graph data

To begin the calculation of the coefficients $\omega(s)$, let us consider a few simple cases.

(i) Case $s = 2$, $r = 3$. In this case G_2 contains only one contour ($\nu(G_2) = 1$) from the collection g (1, 0), g (2, 0), g (1, 0; 2, 0), g (0, 1), g (0, 2), g (0, 1; 0, 2). The r-mer configurations corresponding to the first three contours are shown in figures 1 (*b*), (*c*) and (*d*). The remaining three contours correspond to vertical r-mers. Thus, for $r = 3$, $\omega_N(2) = -6N$ and $\omega(2) = -6$. A simple calculation shows that for arbitrary r

$$\omega_N(2) = -2[r(r-1)/2]N, \qquad \omega(2) = -r(r-1).$$

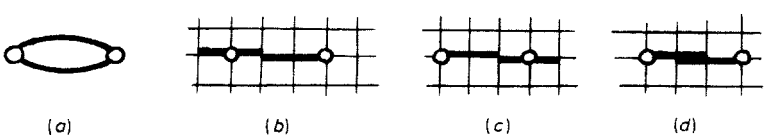
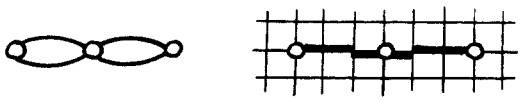


Figure 1. Case $s = 2$, $r = 3$. Open circles denote superlattice points. (*a*) A cycle; (*b*), (*c*), (*d*) r-mer configurations generating contours g(1, 0), g(2, 0), g(1, 0; 2, 0).

(ii) Case $s = 3$, $r = 3$. One of the two configurations appearing in this case is shown in figure 2. We see that $\omega_N(3) = 2N$, $\omega(3) = 2$. For arbitrary $r > 2$ we have

$$\omega_N(3) = 2\sum_{i=2}^{r-1} \frac{i(i-1)}{2} N, \qquad \omega(3) = \sum_{i=2}^{r-1} i(i-1) = 2\binom{r}{3}.$$

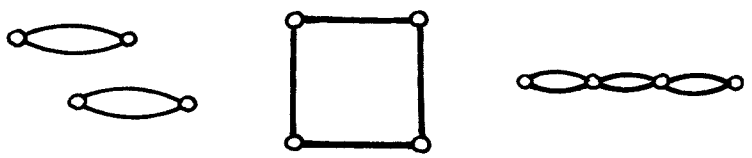


(a) (b)

Figure 2. (*a*) Two connected cycles in case $s=3$. (*b*) The r-mer configuration ($r=3$) generating contours g(1, 0) and g(2, 0).

(iii) Case $s=4$. This case is illustrated in figure 3. The enumeration of the contours corresponding to cycles of type (*a*) leads to

$$N(2N-7)[r(r-1)/2]^2.$$



(a) (b) (c)

Figure 3. Cycles contributing to the coefficient $\omega(4)$.

Similarly, for the cycles of type (*b*) we have

$$2N[r(r-1)/2]^2$$

and for those of type (*c*)

$$2N\binom{r}{4} \qquad (r>3)$$

$$0 \qquad (r=2 \text{ or } r=3).$$

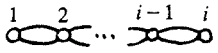
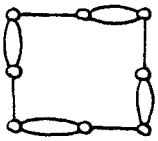
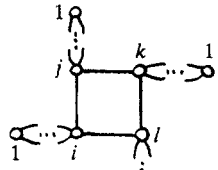
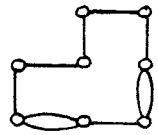
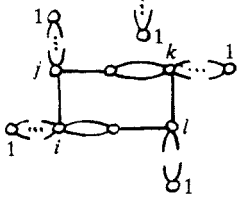
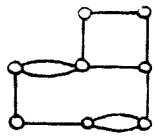
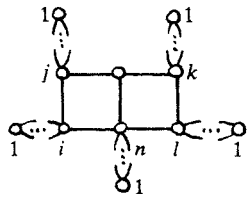
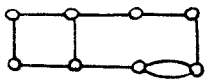
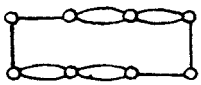
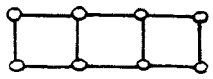
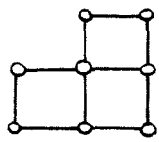
Using these expressions, we obtain

$$\omega(4)=-7\left[\frac{r(r-1)}{2}\right]^2-2\binom{r}{4}.$$

To consider more general cases, let us make first some preliminary remarks. Among the set of contours arising on a graph there can appear such pairs of contours g and g′ that for any configuration of r-mers the presence of g′ necessitates the presence of g but the inverse does not hold. In that case, following the principle of inclusion and exclusion when calculating the coefficient $\omega(s)$ we should take into account only the contribution from contour g.

Now let us derive general expressions for $\omega(s)$ up to eighth-order. To this end, consider all possible types of connected cycles entering into the eighth-order expansion (table 1) and calculate the numbers of r-mer configurations generating different contours which correspond to each of these cycles. These numbers will be denoted by $K^{(r)}$ for each value of the r-mer length; their dependence on other indices is shown in table 1. Indices i, j, k, l, n take values such that the total number of cycle vertices does not exceed eight. Formulae for calculating the number of r-mer configurations $K^{(r)}$ can

Table 1.

n	Type of cycle	$K^{(r)}$	n	Type of cycle	$K^{(r)}$
1		$K_1^{(r)}(i)$	6		$K_6^{(r)}$
2		$K_2^{(r)}(i,j,k,l)$	7		$K_7^{(r)}$
3		$K_3^{(r)}(i,j,k,l)$	8		$K_8^{(r)}$
4		$K_4^{(r)}(i,j,k,l,n)$	9		$K_9^{(r)}$
5		$K_5^{(r)}$	10		$K_{10}^{(r)}$
			11		$K_{11}^{(r)}$

be found in appendix A, where we use the convention that a sum is equal to zero if the lower index in the summation exceeds the upper one.

The formulae (A1)–(A11) are derived by using the fact that the combinatorial problem of finding sequences of overlapping r-mers on i consecutive bonds can be considered as the problem of finding a sequence of i decreasing numbers from the set $\{0, 1, \ldots, r-1\}$. Appendix B contains formulae for calculating the coefficients $\omega(s)$ expressed through $K^{(r)}$.

5. Results

The expansion coefficients $\omega(s)$, $s = 2, 3 \ldots 8$ obtained by evaluating expressions (A1)–(A11) and (B1)–(B7) for the first twenty values of r are listed in table 2. The series in equation (3.5) has been truncated after the eighth term and the resulting values of the molecular freedom

$$\phi_r = (2r)^{1/r}\left[1 + \sum_{s=2}^{8} \omega(s)\left(\frac{1}{2r}\right)^s\right]^{1/r}$$

are listed in the last column of table 2. These expansions are not long enough to lead to

Table 2. Expansion coefficients $\omega(s)$ and molecular freedom ϕ_r.

	(2)	(3)	(4)	(5)	(6)	(7)	(8)	ϕ_r
2	−2	0	−7	0	−50	0	−456	1·82
3	−6	2	−63	72	−1302	2552	−34616	1·66
4	−12	8	−254	576	−10596	40464	−575305	1·55
5	−20	20	−710	2402	−49900	280600	−4597065	1·48
6	−30	40	−1605	7212	−170702	1263200	−23904755	1·42
7	−42	70	−3157	17682	−473354	4336502	−93731071	1·38
8	−56	112	−5628	37744	−1131368	12346448	−300998182	1·34
9	−72	168	−9324	72828	−2420664	30641256	−833031414	1·32
10	−90	240	−14595	130104	−4753770	68450640	−2054905650	1·29
11	−110	330	−21835	218724	−8718974	140699460	−4624997850	1·27
12	−132	440	−31482	350064	−15124428	270315584	−9659711831	1·26
13	−156	572	−44018	537966	−25047204	491091744	−18959734183	1·24
14	−182	728	−59969	798980	−39887302	851161168	−35311572933	1·23
15	−210	910	−79905	1152606	−61426610	1417146770	−62879523305	1·22
16	−240	1120	−104440	1621536	−91892816	2279043680	−107704597660	1·21
17	−272	1360	−134232	2231896	−134028272	3555894896	−178328349436	1·20
18	−306	1632	−169983	3013488	−191163810	5402319840	−286560913644	1.19
19	−342	1938	−212439	4000032	−267297510	8015955600	−448413979212	1·18
20	−380	2280	−262390	5229408	−367178420	11645870640	−685220801205	1·17

an accurate estimate of ϕ_r by using the Padé technique. Nevertheless, to make a comparison with the results of previous papers we have estimated ϕ_2 and ϕ_3 by evaluating the Padé approximants $P(2, 2)$ and $P(4, 4)$ to the series

$$-2x^2-7x^4-50x^6-456x^8$$

at $x=\frac{1}{4}$ and

$$-6x^2+2x^3-63x^4+72x^5-1302x^6+2552x^7-35912x^8$$

at $x=\frac{1}{6}$.

The results of this calculation are listed in table 3, together with the results of Nagle (1966), Gaunt (1969) and Van Craen and Bellemans (1972). Table 3 clearly demonstrates the accuracy of the calculation of ϕ_r at $r=2$ and $r=3$. Information about the

Table 3. Values of the molecular freedom ϕ_2 per dimer and ϕ_3 per trimer for square lattice.

	Truncated series	Padé approximant
† Nagle (1966)	1·7694	1·7905
Gaunt (1969)	1·7728	1·78–1·80
this paper	1·82	1·81
‡ Van Craen and Bellemans (1972)	—	1·57
This paper	1·66	1·64

† Exact value of $\phi_2 = 1{\cdot}7916\ldots$ (Kasteleyn 1961, Fisher 1961).
‡ Estimate of $\phi_3 = 1{\cdot}60 \pm 0{\cdot}01$ obtained by matrix method (Van Craen 1975).

calculational accuracy at larger r can be obtained by comparing the calculated ϕ_r with its upper and lower bounds reported in the Introduction. For instance, at $r=20$, from equations (1.2) and (1.4), we have $1{\cdot}19 \geqslant \phi_{20} \geqslant 1{\cdot}06$, $\phi_{20} = 1{\cdot}17$, so that the accuracy is no worse than $(-10\%, +2\%)$.

Appendix A

$$K_1^{(r)}(i) = \binom{r}{i} \tag{A1}$$

$$K_2^{(r)}(i,j,k,l) = \binom{r}{i+k}\binom{r}{j+l} \tag{A2}$$

$$K_3^{(r)}(i,j,k,l) = \binom{r}{j+l}\left[\sum_{m_i=i}^{r-1}\sum_{m_{i-1}=i-1}^{m_i-1}\cdots\sum_{m_1=1}^{m_2-1}\sum_{m=m_1}^{r-1}\sum_{n=1}^{m_1}\sum_{n_1=\max(n,k)-1}^{m_1-1}\binom{n_1}{k-1}\right] \tag{A3}$$

$$K_4^{(r)}(i,j,k,l,n) = \binom{r}{i+l+1}$$
$$\times\left[\sum_{m_i=j}^{r-1}\sum_{m_{i-1}=j-1}^{m_i-1}\cdots\sum_{m_1=1}^{m_2-1}\sum_{P_k=k}^{r-1}\sum_{P_{k-1}=k-1}^{P_k-1}\cdots\sum_{P_1=1}^{P_2-1}\sum_{q_1=n-1}^{\min(m_1,P_1)-1}\binom{q_1}{n-1}\right] \tag{A4}$$

$$K_5^{(r)} = \binom{r}{2}\left[\sum_{m_3=1}^{r-1}\sum_{m_2=1}^{m_3}\sum_{m_1=1}^{m_2}\sum_{m=1}^{m_1}(m_1-m+1)(m_1-m+2)/2\right] \tag{A5}$$

$$K_6^{(r)} = \left[\sum_{m_1=1}^{r-1}\sum_{m=1}^{m_1} m(m+1)/2\right]^2 \tag{A6}$$

$$K_7^{(r)} = \left\{\sum_{m_1=1}^{r-1}\sum_{m=1}^{r-1}\min(m,m_1)[\min(m,m_1)+1]/2\right\}^2 \tag{A7}$$

$$K_8^{(r)} = \binom{r}{3}\sum_{m_2=1}^{r-1}\sum_{m_1=1}^{m_2}\sum_{m=1}^{m_1} m(2r-m-1)/2 \tag{A8}$$

$$K_9^{(r)} = \left(\sum_{n=1}^{r-1} n^2\right)\sum_{m_1=2}^{r-1}\sum_{m=1}^{m_1-1} m(r-m)(m+1)/2. \tag{A9}$$

$$K_{10}^{(r)} = \binom{r}{4}\sum_{m_1=1}^{r-1}\sum_{m=1}^{r-1}\min(m,m_1)m \tag{A10}$$

$$K_{11}^{(r)} = \left(\sum_{m_1=2}^{r-1}\sum_{m=1}^{m_1-1} m^2\right)\sum_{n_1=2}^{r-1}\sum_{n=1}^{n_1-1} n(2r-n-1)/2 \tag{A11}$$

Appendix B

$$\omega(2) = -2K_1^{(r)}(2) \tag{B1}$$

$$\omega(3) = 2K_1^{(r)}(3) \tag{B2}$$

$$\omega(4) = -7[K_1^{(r)}(2)]^2 - 2K_1^{(r)}(4) \tag{B3}$$

$$\omega(5) = 24K_1^{(r)}(2)K_1^{(r)}(3) + 2K_1^{(r)}(5) \tag{B4}$$

$$\omega(6) = -2K_1^{(r)}(6) - 46[K_1^{(r)}(2)]^3 - 20\,[K_1^{(r)}(3)]^2$$
$$-34K_1^{(r)}(2)K_1^{(r)}(4) - 4K_3^{(r)}(1,1,1,1) + 4K_4^{(r)}(1,1,1,1,1) \tag{B5}$$

$$\omega(7) = 2K_1^{(r)}(7) + 44K_1^{(r)}(2)K_1^{(r)}(5) + 56K_1^{(r)}(3)K_1^{(r)}(4)$$
$$+276[K_1^{(r)}(2)]^2K_1^{(r)}(3) + 8K_3^{(r)}(2,1,1,1) + 8K_3^{(r)}(1,2,1,1)$$
$$-8K_4^{(r)}(2,1,1,1,1) - 8K_4^{(r)}(1,2,1,1,1) - 4K_4^{(r)}(1,1,1,1,2) \tag{B6}$$

$$\omega(8) = -2K_1^{(r)}(8) - 382[K_1^{(r)}(2)]^4 - 520K_1^{(r)}(2)[K_1^{(r)}(3)]^2$$
$$-420[K_1^{(r)}(2)]^2K_1^{(r)}(4) - 39[K_1^{(r)}(4)]^2 - 54K_1^{(r)}(2)K_1^{(r)}(6) \tag{B7}$$
$$-72K_1^{(r)}(3)K_1^{(r)}(5) - 60K_1^{(r)}(2)K_3^{(r)}(1,1,1,1)$$
$$+60K_1^{(r)}(2)K_4^{(r)}(1,1,1,1,1) - 8K_3^{(r)}(3,1,1,1) - 8K_3^{(r)}(1,3,1,1)$$
$$+8K_4^{(r)}(3,1,1,1,1) + 8K_4^{(r)}(1,3,1,1,1) - 4K_3^{(r)}(2,1,2,1)$$
$$-4K_3^{(r)}(1,2,1,2) - 8K_3^{(r)}(2,1,1,2) - 8K_3^{(r)}(2,2,1,1)$$
$$+4K_4^{(r)}(2,1,1,2,1) + 4K_4^{(r)}(1,2,2,1,1) + 16K_4^{(r)}(2,2,1,1,1)$$
$$+4K_4^{(r)}(1,1,1,1,3) + 8K_4^{(r)}(2,1,1,1,2) + 8K_4^{(r)}(1,2,1,1,2)$$
$$-4K_5^{(r)} - 2K_6^{(r)} - 8K_7^{(r)} + 8K_8^{(r)} + 8K_9^{(r)} - 4K_{10}^{(r)} - 8K_{11}^{(r)}$$

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