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1985 J. Phys. A: Math. Gen. 18 847

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Clustering of clusters processes above their upper critical dimensionalities

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Received 23 October 1984, in final form 28 November 1984

Abstract. In contrast with the diffusion limited particle-cluster aggregation (P-CI), the cluster-cluster aggregation model (CI-CI) has a finite upper critical dimensionality above which the fractal dimension of the clusters is trivial. This exponent is derived analytically for a whole set of models, including P-CI and CI-CI. The results clarify the physical differences between these two models.

1. Introduction

In view of the profusion of aggregation models (particle-cluster aggregation (P-CI) (Witten and Sander 1981, Meakin 1983a), clustering of clusters (CI-CI) (Meakin 1983b, Kolb *et al* 1983), hierarchical models (Sutherland and Goodarz-Nia 1971, Botet *et al* 1984a), chemically limited cluster-cluster aggregation (Kolb and Jullien 1984, Jullien and Kolb 1984), ballistic aggregation (Vold 1963, Sutherland 1966, Bensimon *et al* 1983), 'Sutherland's ghost' model (Ball and Witten 1984, Kolb 1984), Rikvold's screened model (Rikvold 1982, Meakin 1983c) . . .) it seems important to search for some physical (relevant) parameters which could aid in classifying all of them. The fractal (Hausdorff) dimension D of a cluster (Hausdorff 1919, Mandelbrot 1982), which characterises the increase of the radius with the number of particles, is the only available parameter which allows some rough classification. Unfortunately, there is at present no rigorous proof that clusters grown by the P-CI or CI-CI process are self-similar, and if we assume that this is the case, there are only numerical (Monte Carlo) or approximate values for D (Muthukumar 1983, Tokuyama and Kawasaki 1983, Kawasaki and Tokuyama 1983, Hentschel 1984, Ball and Witten 1983, Ball *et al* 1984, for the P-CI process; Hentschel and Deutch 1984, for the CI-CI process).

Here we derive the fractal exponent D_c of the clusters above the upper critical dimensionality, when it exists, for a continuous class of models of aggregation. The results yield some fundamental difficulties in classifying unambiguously the clustering of clusters model among the others.

According to the nature of the kinetics, the clustering of clusters process can lead either to the hierarchical CI-CI or to the P-CI model (Botet *et al* 1984b, c, Jullien *et al* 1984). To be more precise, let us consider the irreversible aggregation between an i -mer and a j -mer:

$$(i) + (j) \rightarrow (i+j). \quad (1)$$

The probability of such an event is proportional to $K_{ij}c_i c_j$ where c_i and c_j are the concentrations of i - and j -mers and K_{ij} is the kinetic kernel. The normalised probability of the reaction (1) is just

$$K_{ij}c_i c_j / \sum_{i'+j'=i+j} K_{i'j'}c_{i'}c_{j'}. \quad (2)$$

As a basic hypothesis for this kind of process, we assume the following homogeneity (scaling) condition for the coagulation kernel K :

$$K_{\lambda i, \lambda j} = \lambda^{2\omega} K_{ij} \quad (3)$$

(Botet and Jullien 1984, Kolb 1984, Jullien *et al* 1984). With this type of kernel, the C1-C1 process leads to the hierarchical C1-C1 model when $\omega \rightarrow -\infty$, and to the P-C1 model when $\omega \rightarrow +\infty$. Numerical simulations in $d = 2$ dimensions show that $D \approx D_{\text{C1-C1}} \approx 1.42$ for all $\omega \leq \frac{1}{2}$, and $D \approx D_{\text{P-C1}} \approx 1.67$ for all $\omega \geq \frac{1}{2}$. This result seems independent of the form of the K_{ij} 's provided that (3) is satisfied.

2. The transparency argument

The derivation of D_c of these models is based on an extension of the 'transparency argument' (Witten and Sander 1983, Ball and Witten 1984, Herrmann 1984) which assumes that above the upper critical dimension d_c , clusters become 'transparent' in the sense that, when they collide, they can stick on every point of each cluster with the same probability (because of their very tenuous structure, there is a finite probability that one goes through the other without sticking). Moreover, the following relation holds between d_c and D_c ,

$$d_c = 2D_c + d_w, \quad (4)$$

where d_w is the dimensionality of their relative trajectory ($d_w = 0$ in chemically limited aggregation, $d_w = 1$ in ballistic aggregation, $d_w = 2$ for Brownian random walks, ...).

Relation (4) expresses that one cluster (fractal dimension: D_c) and another one with relative motion of dimensionality d_w (fractal dimension of its trail: $D_c + d_w$) have a finite probability, even for very large clusters, never to meet all along their relative motion.

The derivation of D_c in high-dimensional space, for the hierarchical C1-C1 model ($\omega \rightarrow -\infty$), has been done independently by Ball and Witten (1984) and Obukhov (1984). Here, we generalise these calculations when varying ω .

3. Recursion formula for the radius of gyration

In the spirit of Ball and Witten's derivation (Ball and Witten 1984, Ball and Jullien 1984, Ball 1984), we start from the following definition of the radius of gyration of a cluster of size N :

$$N^2 R_g^2(N) = \frac{1}{2} \sum_{\substack{i \in (N) \\ j \in (N)}} (\mathbf{r}_i - \mathbf{r}_j)^2. \quad (5)$$

If the N cluster has been built by aggregation of an M cluster and an $N - M$ cluster

we can write

$$N^2 R_g^2(N) = M^2 R_g^2(M) + (N - M)^2 R_g^2(N - M) + \sum_{\substack{i \in (M) \\ j \in (N - M)}} (r_i - r_j)^2 \quad (6)$$

because two points of the cluster (N) can belong to (M), or to ($N - M$), or one to (M) and the other to ($N - M$). Now if $\delta = r_{i_0} - r_{j_0}$, $\delta^2 = 1$, is one vector bonding (M) and ($N - M$), we can write

$$N^2 R_g^2(N) = M^2 R_g^2(M) + (N - M)^2 R_g^2(N - M) + (N - M) \sum_{i \in (M)} (r_i - r_{i_0})^2 + M \sum_{j \in (N - M)} (r_j - r_{j_0})^2 + M(N - M)\delta^2 \quad (7)$$

and the transparency and the isotropy hypothesis imply for large clusters

$$\sum_{\substack{i \in (M) \\ j \in (N - M)}} (r_i - r_{i_0})(r_j - r_{j_0}) = \sum_{i \in (M)} (r_i - r_{i_0})\delta = \sum_{j \in (N - M)} (r_j - r_{j_0})\delta = 0.$$

Assuming that a fractal structure exists for the cluster, we use for large N

$$R_g^2(N) \sim N^{2\nu} \quad (8)$$

where ν is the inverse of the fractal exponent.

For large N and positive ν , formula (7) is

$$N^{2+2\nu} = M^{2+2\nu} + (N - M)^{2+2\nu} + 2M(N - M)[M^{2\nu} + (N - M)^{2\nu}] \quad (9)$$

giving the mean radius of gyration of an N cluster in terms of the radii of gyration of its parents.

4. Recursion formula for the 2nth moment

In this section, we show how this formula can also be derived in the framework of Obukhov's formulation. Let us define the two-point correlation function by

$$G_N(\mathbf{r}) = \frac{1}{N} \sum_{\mathbf{r}'} \rho(\mathbf{r}') \rho(\mathbf{r} + \mathbf{r}') \quad (10)$$

where $\rho(\mathbf{r}) = 1$ if the point located at \mathbf{r} belongs to the N cluster, and $\rho(\mathbf{r}) = 0$ otherwise. The quantity $NG_N(\mathbf{r})$ is just the number of pairs of points separated by a distance \mathbf{r} belonging to the N cluster. If the parents of (N) are (M) and ($N - M$), each of these pairs can be a pair of points of (M), or of ($N - M$), or one of the points belongs to (M) and the other to ($N - M$), so that

$$NG_N(\mathbf{r}) = MG_M(\mathbf{r}) + (N - M)G_{N-M}(\mathbf{r}) + \sum_{\mathbf{r}'} G_M(\mathbf{r}') G_{N-M}(\mathbf{r} + \delta - \mathbf{r}') + \sum_{\mathbf{r}'} G_{N-M}(\mathbf{r}') G_M(\mathbf{r} - \delta - \mathbf{r}') \quad (11)$$

where δ is one bond connecting (M) and ($N - M$).

Now defining the Fourier transform of G_N by

$$\tilde{G}_N(k) = N^{-1} \int G_N(\mathbf{r}) e^{i\mathbf{k}\mathbf{r}} d\mathbf{r}, \quad (12)$$

(11) becomes

$$N^2 \tilde{G}_N(\mathbf{k}) = M^2 \tilde{G}_M(\mathbf{k}) + (N - M)^2 \tilde{G}_{N-M}(\mathbf{k}) + 2M(N - M) \tilde{G}_M(\mathbf{k}) \tilde{G}_{N-M}(\mathbf{k}) \cos(\mathbf{k}\boldsymbol{\delta}) \tag{13}$$

which is explicitly given in Obukhov's paper in the case $M = N - M = N/2$ (hierarchical) and $\boldsymbol{\delta} = \mathbf{0}$ (thermodynamic limit).

Therefore, if we perform a formal expansion of $e^{i\mathbf{k}\mathbf{r}}$ in terms of $\mathbf{k}\mathbf{r}$, we have (using the isotropy hypothesis)

$$\tilde{G}_N(\mathbf{k}) = 1 - \frac{1}{2!} k^2 R_g^2(N) + \frac{1}{4!} k^4 \langle r^4(N) \rangle \dots \tag{14}$$

Since $\boldsymbol{\delta}^2 = 1$, for large N and positive ν , we get a generalisation of formula (7) (case $n = 1$):

$$N^2 \langle r^{2n}(N) \rangle = M^2 \langle r^{2n}(M) \rangle + (N - M)^2 \langle r^{2n}(N - M) \rangle + 2M(N - M) \sum_{m=0}^n \binom{2n}{2m} \langle r^{2m}(M) \rangle \langle r^{2n-2m}(N - M) \rangle \tag{15}$$

where we define

$$\langle r^{2n}(N) \rangle = \frac{1}{N} \int G_N(\mathbf{r}) r^{2n} d\mathbf{r} = \frac{1}{2N^2} \sum_{\substack{i \in (N) \\ j \in (N)}} (\mathbf{r}_i - \mathbf{r}_j)^{2n}.$$

5. Derivation of D_c

For each ω appearing in (3), there exists a well defined size-distribution of clusters which is a function of time. More precisely, it can be shown that there is a useful scaling form for the concentration c_k :

$$c_k(t) \sim k^{-\tau} \phi(kt^{1/\sigma}) \tag{16}$$

for $\omega < \frac{1}{2}$, with

$$\tau = 2\omega, \quad \sigma = 2\omega - 1, \quad \text{for } \omega < 0 \tag{17}$$

(Lushnikov 1973, Botet and Jullien 1984, Ernst *et al* 1984) and

$$\tau = 2\omega + 1, \quad \sigma = 2\omega - 1, \quad \text{for } 0 < \omega < \frac{1}{2} \tag{17'}$$

(Leyvraz 1984).

For $\frac{1}{2} < \omega < 1$, there exists a finite gelation time t_g (e.g. Hendriks 1983) and (16) becomes:

$$c_k(t) \sim k^{-\tau} \phi(k|t - t_g|^{1/\sigma}) \tag{16'}$$

near the gel point, with

$$\tau = \omega + \frac{3}{2}, \quad \sigma = \frac{1}{2} - \omega, \quad \text{for } \frac{1}{2} < \omega < 1. \tag{17''}$$

We define now a value $\nu(\omega, N)$ by the implicit equation

$$N^{2+2\nu} = \langle M^{2+2\nu} + (N - M)^{2+2\nu} + 2M(N - M)[M^{2\nu} + (N - M)^{2\nu}] \rangle_{\text{Smol}}. \tag{18}$$

where the Smoluchowski average is taken over all possible parents (M) , $(N - M)$ with

the probability (2), that is

$$\langle \phi \rangle_{\text{Smol}} = \frac{\sum_{M=1}^{N-1} K_{M,N-M} C_M C_{N-M} \phi(M, N-M)}{\sum_{M=1}^{N-1} K_{M,N-M} C_M C_{N-M}} \tag{19}$$

for any function ϕ .

As in numerical simulations, ν , defined by (18), depends on N , but if there exists a limiting value of ν for large clusters, we can say that this value $\nu(\omega) = \nu(\omega, \infty)$ is the characteristic exponent ν of the model. Once we know the asymptotic behaviour of sums like $\sum_{M=1}^{N-1} M^\alpha (N-M)^\beta$ for large N and every choice of exponents α and β (see appendix), the derivation of the limiting value of ν is easy.

The results are the following, and are reported in figure 1.

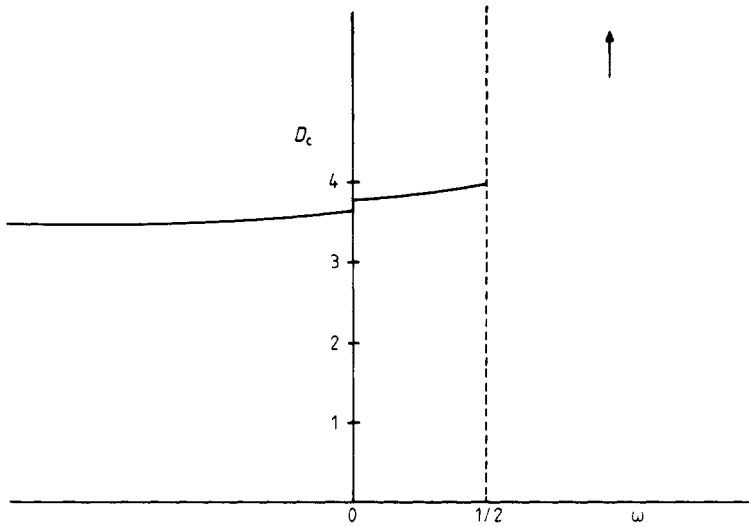


Figure 1. Fractal dimension above the upper critical dimension as a function of ω .

For $\omega < 0$, ν is given by

$$\frac{\Gamma^2(1-\omega)}{\Gamma(2-2\omega)} = (4\nu + 8 - 2\omega) \frac{\Gamma(2\nu + 2 - \omega)}{\Gamma(2\nu + 4 - 2\omega)} \tag{20}$$

As special cases, one has $\nu(-\infty) = \log \frac{3}{2} / \log 4 = 0.2924$ ($D_c(-\infty) \approx 3.42$), found by Ball and Witten (1984) and Obukhov (1984) for the hierarchical C1-C1 model, and $\nu(0^-) = (\sqrt{17} - 3) / 4 \approx 0.2808$ ($D_c(0^-) \approx 3.56$) (Ball 1984).

For $0 < \omega < \frac{1}{2}$, ν is given by

$$\frac{\Gamma(1-\omega)}{\Gamma(1-2\omega)} = (2\nu - 3\omega + 1) \frac{\Gamma(2\nu - \omega + 1)}{\Gamma(2\nu - 2\omega + 2)} \tag{20'}$$

As particular cases, one has $\nu(0^+) \approx 0.2696$ ($D_c(0^+) \approx 3.7$, ν is solution of $\psi(2\nu + 1) - (2\nu + 1)^{-1} = \psi(1)$), and $\nu(\frac{1}{2}^-) = \frac{1}{4}$ ($D_c(\frac{1}{2}^-) = 4$).

For $\frac{1}{2} < \omega$, we have $\nu = 0$, which is consistent with an 'infinite' D_c (i.e. there is no upper critical dimension).

Let us point out some interesting features of these results. First, the change at $\omega = \frac{1}{2}$, from a finite value of D_c to an infinite one, is consistent with the proposal (Botet *et al* 1984a, b, c) that at $\omega = \frac{1}{2}$ one goes from C1-C1 to P-C1, the latter having no upper critical dimensionality (Witten and Sander 1983). Also, we find a (slight) unexpected variation of D_c with ω , for $\omega < \frac{1}{2}$. Moreover for $\omega = 0$, because of the occurrence of a peculiar large-time behaviour which looks like a pregelation stage for $0 < \omega < \frac{1}{2}$ (Leyvraz 1984), there is a slight discontinuity of D_c .

6. Relevance of the relative sizes of colliding clusters

An additional calculation permits us to shed light on these two last points. Let us denote by r the size ratio $M/(N - M)$ during a collision process,

$$(M) + (N - M) \rightarrow (N)$$

(where we consider $M \leq N - M$ to ensure $0 < r \leq 1$). We can derive, using (9), a value for $\nu(r)$ as a function of r (it is N independent). Explicitly, ν is given by

$$(1 + r)^{2+2\nu} = r^{2+2\nu} + 2r^{1+2\nu} + 2r + 1. \tag{21}$$

Figure 2 shows $\nu(r)$ against r . Note that $\nu(r) \sim -1/2 \log r$ for small values of r .

Is it possible to find a connection between this $\nu(r)$ and $\nu(\omega)$ given in figure 1? In other terms, is $\nu(\omega)$ coming from a characteristic ratio $r(\omega)$? For the P-C1 process, we have $r = 0$ (when $N \rightarrow \infty$) and so $\nu(\omega = +\infty) = \nu(r = 0) = 0$. At the other limit, in the hierarchical C1-C1 process, we have $r = 1$ by construction, and $\nu(\omega = -\infty) = \nu(r = 1) = \log \frac{3}{2} / \log 4$. Between these two limits, let us define a parameter $r(\omega, N)$ by

$$r(\omega, N) = \frac{\sum_{M=1}^{N-1} K_{M,N-M} C_M C_{N-M} r(M, N - M)}{\sum_{M=1}^{N-1} K_{M,N-M} C_M C_{N-M}} = \langle r(N) \rangle_{\text{Smol}}. \tag{22}$$

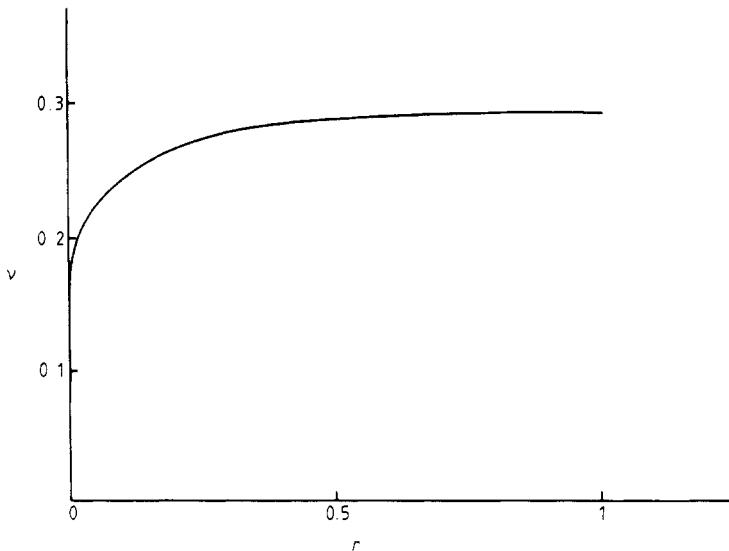


Figure 2. Inverse of the fractal dimension for fixed size-ratio process as a function of the size ratio r .

A derivation similar to that for $\nu(\omega)$, implies the following.

For $\omega < 0$:

$$r(\omega, N) \sim \frac{1-\omega}{-\omega} \left(1 - \frac{\Gamma(2-2\omega)}{\Gamma^2(1-\omega)} \frac{2^{2\omega}}{1-\omega} \right) \tag{23}$$

for large N . We remark that there exists a limiting value independent of the size N such that we can define unambiguously $r(\omega) = r(\omega, \infty)$. Now, if we compare $\nu(r(\omega))$ (as given by (21) and (23) and $\nu(\omega)$ (as given by (20) or (20')), we find a very good agreement (but not perfect, because of the different ways of taking the averages). The two exponents $\nu(\omega = -\infty)$ and $\nu(r(-\infty))$ are equal, and $\nu(r = 2 \log 2 - 1) \sim 0.2824$ instead of $\nu(\omega = 0^-) \approx 0.2808$, so we have deviations of less than 0.5% on the values of ν .

For $0 < \omega < \frac{1}{2}$:

$$r(\omega, N) \sim \frac{2}{(1+\omega)\zeta(1+\omega)} \left(2^{2\omega} - (1-2\omega) \frac{\Gamma^2(1-\omega)}{\Gamma(2-2\omega)} \right) N^{-\omega}. \tag{23'}$$

In this case r is a decreasing function of N , leading to 0 when $N \rightarrow \infty$. A value of r is very difficult to deduce since it depends on the whole 'genealogy' of a cluster. We cannot say that we are in the CI-CI regime, since $\lim_{N \rightarrow \infty} r(\omega, N) = 0$, but we cannot say either that we are in the P-CI regime, since big clusters always collide (we nearly have $(N - N^{1-\omega}) + (N^{1-\omega}) \rightarrow (N)$, and $1 - \omega$ is positive).

For $\frac{1}{2} < \omega < 1$:

$$r(\omega, N) = 0 \text{ above the gel time.} \tag{23''}$$

This last result is consistent with a P-CI process, where microscopic particles stick on a macroscopic cluster (this is also consistent with the above result: $\nu = 0$).

7. Conclusion

With the transparency assumption, we have been able to derive analytically a value of D_c above the upper critical dimensionality, for any model of clustering of clusters with homogeneous kinetic coagulation coefficient K_{ij} . If 2ω is this degree of homogeneity, we have a pure CI-CI process for negative values of ω , with a well defined (time and size independent) size ratio of the colliding clusters. In this case, D_c is slightly increasing with ω . This can be qualitatively explained by the fact that for a given couple of clusters, the smaller one is, the deeper it can penetrate into the other, so that D_c increases if the size ratio decreases (nevertheless, note that this is a very small effect: if Δ is the mean distance between the centres of gravity of the two colliding clusters ('hitching radius' of Botet *et al* (1984)), we have $\Delta/R_g \sim (1+r^{2\nu})/(1+r)^{2\nu}$ which varies from $\frac{4}{3}$ for $\omega = -\infty$ to 1.32 for $\omega = 0$). So there is not a unique value of D_c characterising the CI-CI model but a (small) range of values (3.42-3.56) depending on the kinetics.

For $\omega > \frac{1}{2}$, all the results are consistent with a P-CI process (the size ratio is 0, there is no upper critical dimensionality).

Between 0 and $\frac{1}{2}$, the situation is not so clear because we have to deal with a process where the size ratio is a decreasing function of time from a finite value to 0, so it does not look like a CI-CI process, and there is a finite upper critical dimensionality and a

well defined value of D_c , so that it does not look like a P-Cl process either. This might be a third class of process. The limiting case $\omega = \frac{1}{2}$, with $D_c = 4$, and located on the boundary between non-gelling and gelling systems, seems very interesting to study in more detail.

Acknowledgments

I acknowledge R Jullien for many fruitful comments, R Ball for discussions on this problem during his stay at Orsay, M Kolb and M Herrmann for more general comments.

Appendix

We tabulate here the asymptotic behaviours of

$$\sum_{M=1}^{N-1} M^\alpha (N - M)^\beta$$

for large N and exponents α and β greater than -2 .

Case 1: $\alpha > -1$ and $\beta > -1$.

$$\sum_{M=1}^{N-1} M^\alpha (N - M)^\beta = N^{\alpha+\beta+1} \frac{\Gamma(\alpha + 1)\Gamma(\beta + 1)}{\Gamma(\alpha + \beta + 2)} + O(N^{\alpha+\beta}).$$

Case 2: $\alpha < -1$ and $\beta > 0$.

We have the identity (Abel's summation technique):

$$\sum_{M=1}^{N-1} M^\alpha (N - M)^\beta = \sum_{M'=1}^{N-2} \left(\sum_{M=1}^{M'} M^\alpha \right) [(N - M')^\beta - (N - M' - 1)^\beta] + \sum_{M=1}^{N-1} M^\alpha$$

so

$$\sum_{M=1}^{N-1} M^\alpha (N - M)^\beta = N^\beta \zeta(-\alpha) + N^{\alpha+\beta+1} \frac{\Gamma(\alpha + 1)\Gamma(\beta + 1)}{\Gamma(\alpha + \beta + 2)} + O(N^{\beta-1}).$$

Case 3: $-2 < \alpha < -1$ and $-1 < \beta < 0$.

By the same kind of derivation as for case 2, we have

$$\begin{aligned} \sum_{M=1}^{N-1} M^\alpha (N - M)^\beta &= N^\beta \zeta(-\alpha) + \frac{\beta}{\alpha + 1} \zeta(1 - \beta) N^{\alpha+1} \\ &+ N^{\alpha+\beta+1} \frac{\Gamma(\alpha + 1)\Gamma(\beta + 1)}{\Gamma(\alpha + \beta + 2)} + O(N^{\max\{\alpha, \beta-1\}}). \end{aligned}$$

Case 4: $-2 < \alpha < -1$ and $-2 < \beta < -1$.

Writing twice that $1 = N^{-1}(M + N - M)$, we have

$$\begin{aligned} \sum_{M=1}^{N-1} M^\alpha (N - M)^\beta &= \frac{2}{N^2} \sum_{M=1}^{N-1} M^{\alpha+1} (N - M)^{\beta+1} \\ &+ \frac{1}{N^2} \sum_{M=1}^{N-1} M^\alpha (N - M)^{\beta+2} + \frac{1}{N^2} \sum_{M=1}^{N-1} M^{\alpha+2} (N - M)^\beta. \end{aligned}$$

On the right-hand side, the first sum is a case-1 term, and the others case-2 terms, so that

$$\sum_{M=1}^{N-1} M^\alpha (N-M)^\beta = \zeta(-\alpha) N^\beta + \zeta(-\beta) N^\alpha + N^{\alpha+\beta+1} \frac{\Gamma(\alpha+1)\Gamma(\beta+1)}{\Gamma(\alpha+\beta+2)} + O(N^{\max(\alpha-1, \beta-1)}).$$

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