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Meloy's law of morphological coefficients as a scaling condition for the Fourier representation of particle shape: a stochastic renormalization approach

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Abstract. A new theoretical model for the description of particle shape distributions is suggested. Within its framework a complex Fourier representation of particle shape is used. A frequency distribution expressing the contributions of the different complex harmonics to the dispersion of a position vector is defined. We assume that particles with different shapes are generated during a scale-invariant fragmentation process. Starting from an arbitrary shape distribution the fragmentation leads to a succession of different shape distributions. The asymptotic behaviour of this sequence is analysed by means of a stochastic renormalization approach. Two different scaling laws are found. One scaling law describes the behaviour of the moments of amplitudes attached to the Fourier representation of particle shape, whereas the other corresponds to the renormalized size distribution. In particular, we give a theoretical derivation of the empirical law of morphological coefficients discovered by Meloy. A physical interpretation of the scaling conditions is given based on a many-particle branched-chain approach. We suggest that the scaling behaviour is mainly due to the time-homogeneous Markovian structure of the many-particle evolution equations.

1. Formulation of the problem

A common approach to the morphological analysis of particle shape consists of tracing the contour of one of its sections using a ray originating from a fixed point and by recording the length r of the ray as a function of the angle θ relative to a fixed position:

$$r = r(\theta). \quad (1a)$$

The contour is a closed curve and thus the function $r(\theta)$ has a period equal to 2π . We can represent $r(\theta)$ as a Fourier series:

$$r(\theta) = A_0 + \sum_{m=1}^{\infty} A_m \cos(m\theta + \chi_m). \quad (2)$$

We should outline that such a representation of a contour is always possible. For instance, by displacing the origin with a length r_0 along a direction characterized by

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the angle θ_0 the shape profile may be represented as

$$r_1(\theta) = [r_0^2 + r^2(\theta) - 2r_0r(\theta) \cos(\theta - \theta_0)]^{1/2}. \quad (1b)$$

The dependence $r_1 = r_1(\theta)$ is also periodic. Thus, a Fourier representation is still possible, with the difference that the corresponding Fourier coefficients have different numerical values.

Meloy showed that a graphical representation of $\ln A_n$ versus $\ln n$ approximates to a straight line (Meloy 1978):

$$\ln A_n = g \ln n + h. \quad (3)$$

Equation (3) is known as 'Meloy's law of morphological coefficients'. It is satisfied by a relatively large class of particle shapes encountered in nature. Its range of validity is extended when we consider ensembles of particles having the same provenance (Beddow and Meloy 1980, Carmichael 1982, Clark 1981, Bandemer *et al* 1985). In this case the corresponding amplitudes are replaced by average values. The average coefficients g and h determined from a sample of 'representative' particles may be used to characterize the whole particle collective. This method is commonly used in applied stereology.

In spite of its practical utility, a satisfactory theoretical explanation of Meloy's law is still missing. Vlad (1988) tried to obtain equation (3) as an extremal law which optimizes a certain informational entropy function. Unfortunately, equation (3) corresponds to a less-common isoperimetric condition which has no clear physical significance. The purpose of this paper is to give a plausible physical interpretation of Meloy's law. Our main assumption is that this law expresses in fact a scaling condition for an ensemble of particles having different individual shapes but which are the result of the same type of multifragmentation processes. In developing such an approach a clear distinction should be made between the measurement process aimed at by the Fourier method and the fragmentation process itself. In this context the fragmentation theory is not used to illustrate, but is merely a fundamental tool.

Fragmentation processes have long been studied in specialized areas of science and technology, but quite recently a more general interest has appeared, with new approaches of potentially wider applicability being proposed (see Cheng and Redner 1990, Cai *et al* 1991, Mekjian 1990, Lee and Mekjian 1990, and references therein). The mathematical description of multifragmentation processes is not easy. We distinguish the following aspects. When we are interested in the number of particles generated by a succession of multifragmentation events we can use the theory of branched-chain processes (Athreya and Ney 1972, Vere-Jones 1977). However, this theory cannot be used to evaluate the statistical properties of particle shape. A possible solution would be to combine the theory of branched chains with the theory of multiplicative random walks. This leads to a very complicated model which cannot be treated in the general case. Fortunately, we are not interested here in the evaluation of the statistical properties of the number of particles. This fact allows for a simplified description. By considering the offspring generated by an initial particle we shall analyse only a branch of the corresponding 'genealogical tree'. We mention that a similar simplified description has been used in connection with the problem of turbulent flow (Eggers and Grossmann 1991). The behaviour of this branch will be described in terms of a multiplicative random process. The main advantage of the theory of multiplicative processes is that it allows a simple description of self-similarity. Rather than trying to develop a detailed specific model, we shall assume the validity of self-similarity as the main feature of

many fragmentation theories (Cheng and Redner 1990, Cai *et al* 1991, Mekjian 1990, Lee and Mekjian 1990).

The plan of the paper is as follows. First we shall introduce a complex Fourier representation of particle shape and discuss the behaviour of complex Fourier amplitudes. A stochastic renormalization approach will be introduced by assuming that the number of fragmentation events is distributed according to a geometrical law. Further, we shall analyse the particle size scaling. We shall also try to make a connection between our approach and many-particle fragmentation dynamics; a full branched-chain description will be used. By generalizing the fragmentation theory of Vlad (1991) to include the particle shape we shall recover the one-particle theory as a particular case. On this basis a physical interpretation of the scaling behaviour will be given. As a final topic we shall discuss some drawbacks and open problems related to our approach.

2. A complex Fourier representation of particle shape

We shall represent the shape of a given particle by a complex Fourier series:

$$r(\theta) = \sum_{-\infty}^{+\infty} C_n \exp(in\theta) \quad C_n = (2\pi)^{-1} \int_0^{2\pi} r(\theta) \exp(-in\theta) d\theta. \quad (4)$$

The amplitudes A_n from equation (2) are equal simply to $A_n = 2|C_n|$. By using equation (4) we can evaluate the mean and the dispersion of the ray corresponding to a given shape. We get

$$\tilde{r} = (2\pi)^{-1} \int_0^{2\pi} r(\theta) d\theta = C_0 \quad (5)$$

and

$$\sigma^2 = \overbrace{(r(\theta) - \tilde{r})^2} = (2\pi)^{-1} \int_0^{2\pi} (r(\theta) - \tilde{r})^2 d\theta = 2 \sum_{n=1}^{\infty} |C_n|^2. \quad (6)$$

Taking equation (6) into account we can introduce the frequency distribution

$$P_n = 2|C_n|^2/\sigma^2 \quad n = 1, 2, \dots \quad (7a)$$

which obviously fulfils the conditions

$$P_n \geq 0 \quad \sum_{n=1}^{\infty} P_n = 1. \quad (7b)$$

P_n expresses the contribution of the n th harmonic $C_{-n} \exp(-in\theta) + C_n \exp(in\theta)$ to σ^2 . We note that we can ascribe a probabilistic interpretation similar to the interpretation of wavefunctions in quantum mechanics to the square modulus of amplitude $|C_n|^2$.

We shall assume that each fragmentation step is scale-invariant. The shape $r_q(\theta)$ of a particle resulting from the q th fragmentation process is random. It depends on the shape $r_{q-1}(\theta)$ of its ancestor as well as on other random factors which fulfil the condition of scale invariance. A simple choice would be to consider that $r(\theta)$ is a superposition of self-affine transformations of the shape of the ancestor,

$$r_1(\theta) \sim \beta_1 r_{q-1}(N_1\theta), \beta_2 r_{q-1}(N_2\theta), \dots \quad (8)$$

where β_i and N_i are random variables selected from a certain probability law. The different realizations β_i, N_i corresponding to different steps may differ. However, the scale invariance requires that the probability law should have the same form for all steps. Although β_i and N_i are random, they are not completely arbitrary. As the size of a fragment is less than the size of its ancestor, β should be less than unity. In contrast, as each particle contour is closed we have $\beta r_{q-1}(N\theta) = \beta r_{q-1}[N(\theta + 2\pi)]$. But r_q and r_{q-1} have the period 2π and therefore N should be an integer. The value $N = 0$ is excluded, as it leads to a circular shape irrespective of the shape of ancestors. As the shapes corresponding to $-N$ and $+N$ are identical, we shall consider only the positive values of N . By inserting the Fourier representations of $r_{q-1}(\theta)$ and $r_q(\theta)$ into equation (8) we get a recurrence equation for the values of the Fourier coefficients

$$C_n^{(q)} = \sum_{n'} \beta \delta_{(n'N)(n)} C_{n'}^{(q-1)}. \tag{9}$$

Equation (9) may be used to derive a chain of equations for the moments of the square modulus $\langle |C_n^{(q)}|^{2m} \rangle$. Denoting by $\xi_N(\beta)$ the probability density of the fragmentation parameters N and β , raising equation (9) and its complex conjugate to the m th power and averaging over all values of $\beta, N, C_n^{(q)}$ and $C_n^{(q-1)}$ we arrive at

$$\begin{aligned} \langle |C_n^{(q)}|^{2m} \rangle &= \sum_{n_1^*} \sum_{n_1} \dots \sum_{n_m^*} \sum_{n_m} \sum_N \delta_{(Nn_1^*)(-n)} \delta_{(Nn_1)(n)} \dots \delta_{(Nn_m^*)(-n)} \delta_{(Nn_m)(n)} \\ &\quad \times \langle C_{n_1}^{(q-1)} C_{n_1^*}^{(q-1)} \dots C_{n_m}^{(q-1)} C_{n_m^*}^{(q-1)} \rangle \int_0^1 \beta^{2m} \xi_N(\beta) d\beta \\ &= \sum_{n'} \sum_N \langle |C_{n'}^{(q-1)}|^{2m} \rangle \delta_{(Nn')(n)} \int_0^1 \beta^{2m} \xi_N(\beta) d\beta. \end{aligned} \tag{10}$$

3. A stochastic renormalization approach

In order to avoid the occurrence of an infinity of particles of size 0, the fragmentation process should be limited in some way. It follows that for each step there is a finite probability λ that the fragmentation process terminates. For scale-invariant systems λ is the same for all steps. In this case the probability that the fragmentation consists of q steps is equal to

$$\phi_q = (1 - \lambda)^q \lambda. \tag{11}$$

The final values of the moments of the Fourier coefficients may be derived by averaging over ϕ_q :

$$\langle |\tilde{C}_n|^{2m} \rangle = \sum_{q=0}^{\infty} (1 - \lambda)^q \lambda \langle |C_n^{(q)}|^{2m} \rangle. \tag{12}$$

By combining equations (10) and (12) we get a renormalization group relationship:

$$\langle |\tilde{C}_n|^{2m} \rangle = \lambda \langle |C_n^{(0)}|^{2m} \rangle + (1 - \lambda) \sum_{N;n'} \langle |\tilde{C}_{n'}|^{2m} \rangle_N \delta_{(n'N)(n)} \int_0^1 \beta^{2m} \xi_N(\beta) d\beta. \tag{13}$$

Equation (13) is a discrete analogue of the equations derived by Novikov (1966), Shlesinger and Hughes (1981), West and Shlesinger (1989) and West (1990a, b) in other physical contexts. If the mean number of fragmentation steps $\langle q \rangle = \sum q \phi_q = (1/\lambda) - 1$ is very large, i.e. if $1 - \lambda$ is near unity, the first term on the RHS of equation

(13) is negligible and equation (13) becomes a 'true' scaling equation which has the non-analytic solution

$$\langle |\tilde{C}_n|^{2m} \rangle \sim n^{-(1+H_m)} \tag{14}$$

where H_m is the real root of the transcendental equation

$$(1-\lambda) \sum_{N \geq 1} N^{H_m} \int_0^1 \beta^{2m} \xi_N(\beta) d\beta = 1. \tag{15}$$

This equation has a single real solution H_m . The sign of H_m is the same as the sign of the difference $(1-\lambda)^{-1} - \langle \beta^{2m} \rangle$. In particular, for $(1-\lambda)^{-1} = \langle \beta^{2m} \rangle$, we have $H_m = 0$.

Putting in equation (14) $m = 2$ and introducing the notation $\tilde{A}_n = 2(\langle |\tilde{C}_n|^2 \rangle)^{1/2}$ we get a relationship similar to Meloy's law:

$$\ln \tilde{A}_n \sim \text{const} - (1 + H_1) \ln n. \tag{16}$$

If the dispersion $\langle |\tilde{C}_n|^2 \rangle - \langle \tilde{C}_n \rangle^2$ is sufficiently small, i.e. the fluctuation of the particle shape is not too high, then equation (16) is equivalent to Meloy's law applied to an ensemble of particles.

We note that for $m = 2$ and σ^2 finite, the probabilistic interpretation of $\langle |C_n|^2 \rangle$ requires that $H_1 > 0$; otherwise the relationship (7b) is violated and the probability P_n (equation (7a)) is not normalized.

4. Size scaling

The scale invariance also generates a scaling behaviour for the distribution of particle size. Since, as usual, we have considered only the bidimensional representation of the particle shape we shall measure the size by the area of a given section:

$$S = \int_0^{2\pi} r^2(\theta) d\theta. \tag{17}$$

We start from an initial probability density $B_0(S)$ which corresponds to the distribution of initial shapes. By using equation (8) it is easy to show that two successive sizes S_q and S_{q-1} are related to each other through the relationship

$$S_q = \beta^2 \int_0^{2\pi} r_{q-1}^2(N\theta) d\theta = \beta^2 S_{q-1}. \tag{18}$$

The probability density $B_q(S)$ corresponding to the q th step results by averaging the factor $\delta(S - \beta^2 S_{q-1}) B_{q-1}(S_{q-1})$ over all possible values of β and S_{q-1} :

$$B_q(S_q) = \sum_N \int_0^1 \xi_N(\beta) B_{q-1}(S_q/\beta^2) \beta^{-2} d\beta. \tag{19}$$

The final size distribution results by averaging over q :

$$\tilde{B}(S) = \sum_q \phi_q B_q(S). \tag{20}$$

By combining equations (11), (19) and (20) we get a renormalization equation similar to equation (13):

$$\tilde{B}(S) = \lambda B_0(S) + (1-\lambda) \sum_N \int_0^1 \xi_N(\beta) \tilde{B}(S/\beta^2) \beta^{-2} d\beta. \tag{21}$$

This integral equation can be solved by means of a Mellin transformation: the solution corresponds to the roots of the transcendental equation

$$1 = (1 - \lambda) \sum_N \int_0^1 \xi_N(\beta) \beta^{2(X-1)} d\beta. \tag{22}$$

This equation has a single real solution $X = X_0$ which fulfils the condition $1 > X_0 > 0$. Unlike the case of equation (13) the complex roots may also have a significant contribution to $\tilde{B}(S)$. However, as the real parts of the complex roots are less than $X = X_0$, in the limit $1 - \lambda \rightarrow 1$ and $S \rightarrow 0$ these contributions are negligible. We get

$$\tilde{B}(S) \sim S^{-X_0} \quad \text{as } S \rightarrow 0, 1 - \lambda \rightarrow 1. \tag{23}$$

Thus $\tilde{B}(S)$ has a singularity at $S = 0$. Equation (23) implies that the probability density of the reciprocal value of size $Y = S^{-1}$ behaves as a statistical fractal as $Y \rightarrow \infty$. The probability density of Y has the asymptotic behaviour $Y^{-(1+H^*)}$, with $H^* = 1 - X_0$.

5. A branched-chain approach

In this section we address a very difficult problem: in what sense is scaling and renormalization as proposed a novel result? Once scale invariance is assumed it is not surprising that a power law (equation (16)) comes out of the calculation. Indeed, renormalization always implies scaling; hence, it seems that as such finding scaling is no result. To answer this question we shall consider a particular fragmentation mechanism and try to recover the scaling equations derived above. We shall assume that a full description of the fragmentation dynamics can be done in terms of a branched-chain process. By generalizing the theory of Vlad (1991) the dynamics of the process can be described, in terms of the probability density functional

$$\Gamma_M^{(q)}[r_1(\theta), \dots, r_M(\theta)] \mathcal{D}r_1(\theta) \dots \mathcal{D}r_M(\theta), \tag{24a}$$

that at the q th fragmentation step there are M particles having sizes between $r_1(\theta)$ and $r_1(\theta) + \delta r_1(\theta), \dots, r_M(\theta)$ and $r_M(\theta) + \delta r_M(\theta)$. We have

$$\sum_M \int \dots \int \Gamma_M^{(q)}[r_1(\theta), \dots, r_M(\theta)] \mathcal{D}r_1(\theta) \dots \mathcal{D}r_M(\theta) = 1. \tag{24b}$$

In terms of these functionals we can write the evolution equations

$$\begin{aligned} \Gamma_{M_q}^{(q)}[r_1^{(q)}(\theta), \dots, r_{M_q}^{(q)}(\theta)] &= \sum_{M_{q-1}} \sum_{\nu_1} \dots \sum_{\nu_{M_{q-1}}} \int \dots \int \Gamma_{M_{q-1}}^{(q-1)}[r_1^{(q-1)}(\theta), \dots, r_{M_{q-1}}^{(q-1)}(\theta)] \\ &\times f_{\nu_1}^{(q)}[r_1^{(q-1)}(\theta) \rightarrow r_1^{(q)}(\theta), \dots, r_{\nu_1}^{(q)}(\theta)] \dots f_{\nu_{q-1}}^{(q)}[r_{M_{q-1}}^{(q-1)}(\theta) \\ &\rightarrow r_{M_q - \nu_{M_{q-1} + 1}}^{(q)}(\theta), \dots, r_{M_q}^{(q)}(\theta)] \mathcal{D}r_1^{(q-1)}(\theta) \dots \mathcal{D}r_{M_{q-1}}^{(q-1)}(\theta) \\ M_q &= \nu_1 + \dots + \nu_{M_{q-1}} \end{aligned} \tag{25}$$

where

$$f_{\nu}^{(q)}[r^{(q-1)}(\theta) \rightarrow r_1^{(q)}(\theta), \dots, r_{\nu}^{(q)}(\theta)] \mathcal{D}r_1^{(q)}(\theta) \dots \mathcal{D}r_{\nu}^{(q)}(\theta) \tag{26}$$

is the rate of fragmentation at the q th step of a particle having the shape $r^{(q-1)}(\theta)$ into ν particles having the shapes $r_1^{(q)}(\theta), \dots, r_{\nu}^{(q)}(\theta)$, respectively.

The rate of fragmentation may be evaluated by combining the theory of Vlad (1991) with the considerations developed in section 2. We note by $\varphi^{(q)}(\nu)$ the probability that at the q th step a particle breaks into ν pieces. Taking into account equation (8) we can express $f_\nu^{(q)}$ as an average of a product of δ -functionals:

$$f_\nu^{(q)}[r^{(q-1)}(\theta) \rightarrow r_1^{(q)}(\theta), \dots, r_\nu^{(q)}(\theta)] = \sum_{N_1} \dots \sum_{N_\nu} \varphi^{(q)}(\nu) \int_0^1 \dots \int_0^1 \xi_{N_1}^{(q)}(\beta_1) \dots \xi_{N_\nu}^{(q)}(\beta_\nu) \times \prod_{l=1}^\nu \{\delta[r^{(q)}(\theta) - \beta_l r^{(q-1)}(N_l \theta)]\} d\beta_1 \dots d\beta_\nu \tag{27}$$

where we have assumed that the probability densities $\xi_N^{(q)}(\beta)$ are generally q -dependent.

In order to make a connection with the one-particle description introduced above we shall define the one-particle density functional

$$\Xi^{(q)}[\mathcal{R}(\theta)] \mathcal{D}\mathcal{R}(\theta) = \sum_{M=1}^\infty M^{-1} \sum_{l=1}^M \overbrace{\int \dots \int}^M \delta[r_l(\theta) - \mathcal{R}(\theta)] \Gamma_M^{(q)}[r_1(\theta), \dots, r_M(\theta)] \times \mathcal{D}r_1(\theta) \dots \mathcal{D}r_M(\theta) \mathcal{D}\mathcal{R}(\theta). \tag{28}$$

By using this functional density we can evaluate the moments of the Fourier coefficients through the relationship

$$\langle |C_n^{(q)}|^{2m} \rangle = \int \int \Xi^{(q)}[\mathcal{R}(\theta)] (2\pi)^{-2m} \times \int_0^{2\pi} \dots \int_0^{2\pi} \prod_{l=1}^m \{ \mathcal{R}(\theta_l) \mathcal{R}(-\theta_{-l}) \exp[in(\theta_l - \theta_{-l})] d\theta_l d\theta_{-l} \} \mathcal{D}\mathcal{R}(\theta). \tag{29}$$

By combining equations (25)–(29) we can derive a chain of equations for the moments of the square modulus $\langle |C_n^{(q)}|^{2m} \rangle$. The calculations are standard but cumbersome. By performing the computations we recover equation (10) only if

$$\begin{aligned} \varphi^{(q)}(\nu) &= \varphi(\nu) = \text{independent of } q \\ \xi_N^{(q)}(\beta) &= \xi_N(\beta) = \text{independent of } q \end{aligned} \tag{30a}$$

which corresponds to

$$f_\nu^{(q)} = f_\nu = \text{independent of } q. \tag{30b}$$

The self-similar form of equation (10) was essential for the application of the renormalized approach. For fragmentation models defined by equations (25)–(27) this equation is a consequence of the restrictions (30a) and (30b). The physical significance of these restrictions is very clear: they express a condition of homogeneity with respect to the discrete time q , i.e. that the fragmentation of a particle into other particles obeys the same rules at all times. We have no explanation why the branched chain fragmentation processes described by equations (25)–(27) should be time homogeneous; however, we note that the condition of time homogeneity is commonly used in Markovian dynamics (Van Kampen 1981).

6. Discussion

As follows we shall outline some drawbacks and limitations of the above approach.

A first problem is related to the choice of the origin in writing $r = r(\theta)$. Although this description is commonly used in the literature, and presumably all the results are independent of the location of the origin, we were unable to convince ourselves of this.

On the other hand, the calculations seem to indicate that the higher moments of the distribution, obtained from $\langle |\tilde{C}_n|^{2m} \rangle$, are governed by different exponents for different values of m . We do not know whether this is a genuine effect with observable consequences or not. If this is the case, it would be more appropriate to compute $\langle \ln A_n \rangle$, rather than \tilde{A}_n , for comparison with the empirical Meloy's law.

The analogy to quantum mechanics was quite useful. It allowed us to ascribe a probabilistic interpretation to the square modulus of the complex amplitudes $|C_n|^2$. However, it seems that this analogy is too vague and limited. Indeed, the amplitudes C_n themselves are random variables. This is due to the fact that the initial state may correspond to particles having different shapes; moreover, each fragmentation event is a source of randomness. Thus, our problem is rather related to quantum statistics, which generally corresponds to mixed states. In this context a stochastic analogue of the density matrix formalism would be appropriate.

Other questions concern the significance of the time homogeneity condition introduced in section 5. In the particular case considered, this is a necessary condition for the success of the renormalization approach. We do not know whether this is a general property or if it is true only for equations (25)–(27).

Another problem is related to the particle shape. It is not clear whether the above scaling laws are related to a geometrical fractal or not. The idea that the fragmentation phenomena could generate fractal contours seems to be physically plausible.

To answer these questions further investigations are necessary.

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