Shattering transition in a multivariable fragmentation model

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We introduce a linear fragmentation model in which the fragments are described by two correlated variables x and y, and, for the power law breakup rate $a(x,y) = x^\alpha y^\beta$, we derive its exact solution. The large fluctuations of the additional variable prevent any mean-field reduction to a one-variable model. New features, such as shattering with power law decay of the mass, are obtained.

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Much attention has been recently given to the study of sequential fragmentation processes which are involved in a variety of physical situations like polymer degradation, aggregate fragmentation, combustion, grinding of minerals, or nuclear fragmentation. The quantity of interest in such problems is the distribution of fragments and most theoretical approaches have been formulated in terms of rate equations [1–7]. In all these studies, only one variable has been retained to describe a given fragment, e.g., its mass or size. Significant progress in understanding the kinetic behavior of the system for various forms of the breakup rate has been made thanks to the derivation of exact and scaling solutions. Additional properties like the shattering or disintegration transition [1–8], in which mass is lost to a dust phase of zero-mass particles, or intermittency of the fragment distribution [4,5] have been also discussed.

The rate equation for the concentration of fragments of mass (size) x at time t, $c(x,t)$, is usually written as

$$\frac{\partial c(x,t)}{\partial t} = -a(x)c(x,t) + \int_x^{+\infty} dx'\, a(x')f(x|x')c(x',t), \tag{1}$$

where $a(x)$ is the overall fragmentation rate, which is generally taken as a homogeneous function of x, and $f(x|x')$ is the conditional probability to produce a fragment of mass x by the breakup of a particle of mass x'. This linear equation describes a fragmentation process which is driven by an external source. Aside from polymer degradation studies [1,2], it has been, for instance, successfully applied to interpret recent experiments on desorption-induced fragmentation of colloidal aggregates [9]. A similar equation has been used to model atomic collisions cascades, where $c(x,t)$ represents the distribution function of "hot" atoms of energy x [10]. Various discrete versions of Eq. (1) have been studied as well as extensions to include discrete and continuous mass loss [3,6], binary collisions between particles [2], or inactivation mechanisms [5]. However, an obvious limitation of Eq. (1) and of its generalizations so far studied is their one-variable character: a unique variable, the mass or size x, is kept to describe the fragment distribution. All other variables, e.g., energy, charge, shape factors, or else relative composition in multicomponent fragments, have been eliminated by some (implicit) mean-field treatment. This procedure is indeed valid if the fluctuations of the additional variables for particles having the same mass are small, but this may not be taken for granted for all physical situations.

Our goal, in this paper, is to study the influence of these fluctuations on the fragmentation kinetics by introducing explicitly an additional variable y, in the rate equation. We consider a binary breakup process that is described by the following equation:

$$\frac{\partial c(x,y,t)}{\partial t} = -a(x,y)c(x,y,t) + 2\int_x^{+\infty} dx' \int_y^{+\infty} dy'\, \frac{a(x',y')}{x'y'} c(x',y',t), \tag{2}$$

where $c(x,y,t)$ is the concentration of fragments of mass x and energy y (choice of energy as the second variable is arbitrary here) and the conditional probability to produce a fragment (x,y) from a larger fragment (x',y') is uniform. Since most theoretical efforts on one-variable models have focused on homogeneous breakup rates, which seem to be relevant for most physical systems [1–3,9,10], we take for $a(x,y)$ the simplest generalization, $a(x,y) = x^\alpha y^\beta$. This breakup rate describes fragmentation processes which can be considered, in a significant range of the variables, as independent of a typical size and a typical energy (interestingly, the case $\alpha = \beta = 1$ has also been shown to describe the long-time kinetics of a process of random sequential adsorption of unoriented needles onto a line or a plane [11]). Moreover, power laws generally allow a classification among possible asymptotic behaviors of the relevant observables, which is useful to analyze experimental data. A monodisperse initial condition, $c(x,y,0) = \delta(x-L)\delta(y-L')$, is chosen. Without loss of generality, we shall set in the following $L = L' = 1$, which amounts to renormalize x as x/L, y as y/L', and t as $tL^\alpha L'^\beta$. When $\beta = 0$, one recovers that $c_o(x,t) = \int_0^{+\infty} dy\, c(x,y,t)$ follows Eq. (1), with $a(x) = x^\alpha$ and $f(x|x') = 2/x'$. This one-variable equation has been exactly solved [1,7]. For $\alpha > 0$, mass is conserved and the solution obeys (asymptotically) a scaling formula, whereas scaling breaks down and a shattering transition appears for $\alpha < 0$. In this latter regime, all existing moments of the fragment distribution including the total mass, decrease exponentially with time. Since

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our model is symmetric in x and y, a similar result is obtained for the fragment-energy distribution when $\alpha = 0$.

In what follows, we derive an exact solution for the two-variable rate equation, Eq. (2), and we show that except for $\alpha = 0$ or $\beta = 0$, *it does not reduce to an effective one-variable model.* As a result of the correlations between the two variables, new features appear in the fragmentation kinetics: even for positive α and β, no simple two-variable scaling is found; a nontrivial phase diagram is obtained for the shattering transition, and when shattering occurs for α and β of opposite sign, the competing effects associated with the two variables slow down the disintegration process and lead to power law decay of the mass.

To derive the solution, we first introduce the moments of the distributions $c(x,y,t)$,

$$M_{\lambda\mu}(t) = \int_0^1 dx \int_0^1 dy\, x^\lambda y^\mu c(x,y,t), \tag{3}$$

and, by inserting Eq. (3) in Eq. (2), we derive the following equation for $\lambda + 1 > 0$ and $\mu + 1 > 0$,

$$\frac{dM_{\lambda\mu}(t)}{dt} = -\left(1 - \frac{2}{(\lambda+1)(\mu+1)}\right) M_{(\lambda+\alpha)(\mu+\beta)}(t), \tag{4}$$

with the intial condition $M_{\lambda\mu}(0) = 1$.

When α and β are both positive, all moments ($\lambda+1 > 0$ and $\mu+1 > 0$) are defined. Following Charlesby's method [12], we expand the moments in powers of t and we obtain the solution of Eq. (4) as a generalized hypergeometric function [13],

$$M_{\lambda\mu}(t) = {}_2F_2\left(A_{\lambda\mu}, B_{\lambda\mu}; \frac{\lambda+1}{\alpha}, \frac{\mu+1}{\beta}; -t\right), \tag{5}$$

where $A_{\lambda\mu}$ and $B_{\lambda\mu}$ are given by

$$\begin{matrix} A_{\lambda\mu} \\ B_{\lambda\mu} \end{matrix} = \left[\frac{\mu+1}{2\beta} + \frac{\lambda+1}{2\alpha}\right] \pm \frac{\sqrt{[\alpha(\mu+1) - \beta(\lambda+1)]^2 + 8\alpha\beta}}{2\alpha\beta}. \tag{6}$$

The preceding formulas represent a generalization of the one-variable result in which the moment $M_\lambda(t)$ is expressed as a generalized hypergeometric function of lower order, ${}_1F_1((\lambda-1)/\alpha; (\lambda+1)/\alpha; -t)$ [1]. However, a new feature appears here; instead of a unique conserved moment in one dimension, i.e., the mass $M_1(t)$, an infinite set of moments, represented by the hyperbola $(\lambda+1)(\mu+1) = 2$, are conserved. These moments include of course the total mass, $M_{10}(t)$, and the total energy, $M_{01}(t)$, of the system. In the $(\lambda+1, \mu+1)$ plane, the hyperbola separates a region in which all moments decrease with time and a region in which all moments, including the total number of fragments, $M_{00}(t)$, increase with time [Fig. 1(a)]. The long-time behavior of the moments is obtained from the asymptotic expansion of the generalized hypergeometric function [13],

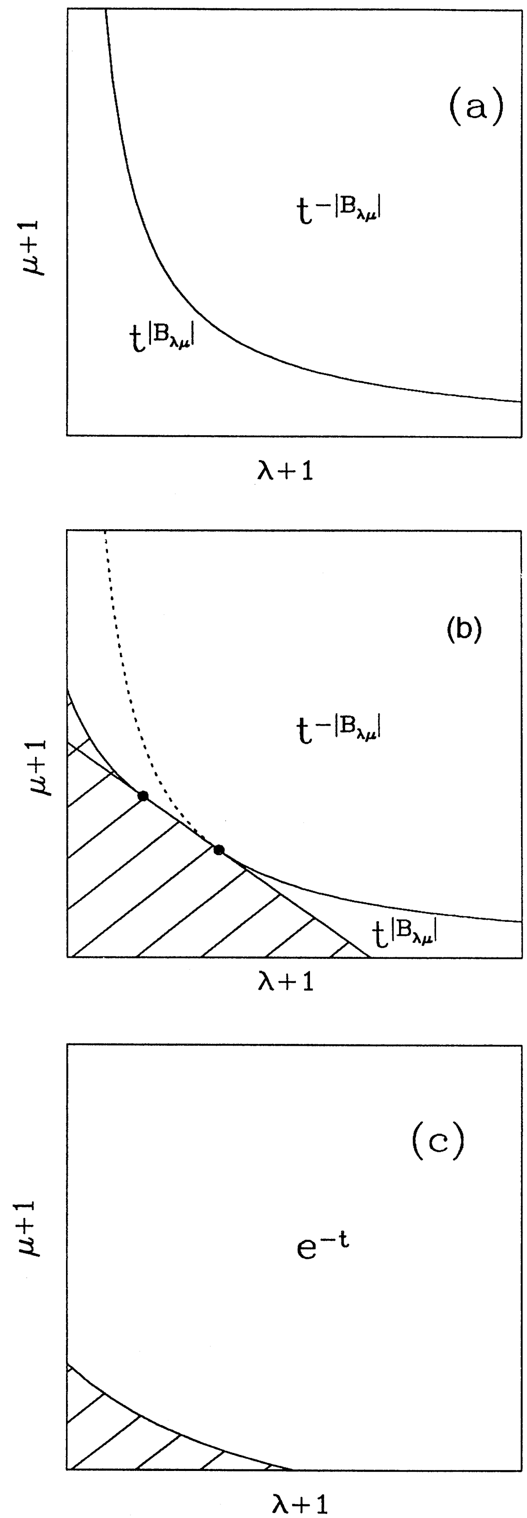

FIG. 1. Domain of definition of the moments $M_{\lambda\mu}(t)$ in the region $(\lambda+1) > 0$, $(\mu+1) > 0$: (a) $\alpha > 0$ and $\beta > 0$, (b) $\alpha < 0$ and $\beta > 0$, (c) $\alpha < 0$ and $\beta < 0$. The hatched zone denotes a region in which the moments are not defined and varies for cases (b) and (c) with the values of α and β. In (b), the dashed part of the hyperbola $(\lambda+1)(\mu+1) = 2$ corresponds to nonconserved moments.

$$M_{\lambda\mu}(t) \simeq \frac{\Gamma\left(\frac{\lambda+1}{\alpha}\right)\Gamma\left(\frac{\mu+1}{\beta}\right)\Gamma(A_{\lambda\mu}-B_{\lambda\mu})}{\Gamma(A_{\lambda\mu})\Gamma(\frac{\lambda+1}{\alpha}-B_{\lambda\mu})\Gamma(\frac{\mu+1}{\beta}-B_{\lambda\mu})} t^{-B_{\lambda\mu}}, \tag{7}$$

where $B_{\lambda\mu}$ is given by Eq. (6) and $\Gamma(z)$ denotes the Γ function. As a result of the correlations between the variables x and y, the exponent of the power law is a nontrivial function of $(\lambda+1)/\alpha$ and $(\mu+1)/\beta$. For instance, the total number of fragments increases with a power law exponent $[\sqrt{(\alpha+\beta)^2+4\alpha\beta}-\alpha-\beta](2\alpha\beta)^{-1}$.

The solution for the fragment distribution $c(x,y,t)$ is derived by taking the inverse double Mellin transform of Eq. (5). We can then express $c(x,y,t)$ as a uniformly convergent series in powers of x^α, y^β, and t, the coefficients of which are explicitly obtained with the help of the residue theorem. The formula is rather lengthy and will be presented elsewhere [14], but we give below its asymptotic form for large times:

$$c(x,y,t) \simeq \sum_{m=0}^{+\infty}\sum_{n=0}^{+\infty} \frac{(-1)^{m+n}}{m!n!} C_{mn}(\alpha,\beta) x^{m\alpha} y^{n\beta} t^{-B_{m'n'}}, \tag{8}$$

$$C_{mn}(\alpha,\beta) = \frac{\Gamma(A_{m'n'}-B_{m'n'})}{\Gamma(A_{m'n'})\Gamma(-n-B_{m'n'})\Gamma(-m-B_{m'n'})}, \tag{9}$$

where $n'=-n\alpha-1$ and $m'=-m\beta-1$. It can be checked from the preceding expressions that $c(x,y,t)$ does not obey any simple two-variable scaling law. This represents a major complication compared to the one-variable model.

When at least one of the parameters α or β is negative, Eq. (5) is no longer valid and the moments $M_{\lambda\mu}(t)$ are not everywhere defined in the region characterized by $(\lambda+1)>0$ and $(\mu+1)>0$. In this regime, one expects a phenomenon related to the shattering transition in the one-variable model to occur. In the latter case, shattering reflects the presence of a singular distribution of zero-mass particles (the dust phase) which represents the continuous limit of a macroscopic monomer contribution appearing at finite times in a discrete model [1]. However, the two-variable case presents a much richer phenomenology: in a discrete version of the model, various contributions of fragments that are both monomers in mass and k-mers in energy $(k \geq 1)$ or monomers in energy and k-mers in mass $(k \geq 1)$ would indeed appear, leading to a nontrivial singular contribution in the continuous limit [note that the singular contributions are *not* described by Eqs. (1), (2), or (4)]. The behavior will thus be different if both α and β are negative, in which case the disintegration cascade involves both mass and energy, and if α and β have opposite signs, which may lead to competing effects.

The solutions of the moment equation, Eq. (4), with the initial condition $M_{\lambda\mu}(0)=1$ and the necessary constraint $M_{\lambda\mu}(t)\geq 0$ (for all defined moments) can be built by considering the Meijer G functions which represent an extension of the generalized hypergeometric functions [13]. The proper solution of the moment equation for $\alpha<0$ and $\beta>0$ is then obtained as

$$M_{\lambda\mu}(t) = \frac{\Gamma(1-A_{\lambda\mu})\Gamma\left(\frac{\mu+1}{\beta}\right)}{\Gamma(B_{\lambda\mu})\Gamma(1-\frac{\lambda+1}{\alpha})} \times G_{23}^{21}\left(t\,\middle|\, \begin{matrix} 1-B_{\lambda\mu}, 1-A_{\lambda\mu} \\ 0, 1-\frac{\lambda+1}{\alpha}, 1-\frac{\mu+1}{\beta} \end{matrix}\right), \tag{10}$$

and for $\alpha<0$ and $\beta<0$ as

$$M_{\lambda\mu}(t) = \frac{\Gamma(1-A_{\lambda\mu})\Gamma(1-B_{\lambda\mu})}{\Gamma(1-\frac{\lambda+1}{\alpha})\Gamma(1-\frac{\mu+1}{\beta})} \times G_{23}^{30}\left(t\,\middle|\, \begin{matrix} 1-B_{\lambda\mu}, 1-A_{\lambda\mu} \\ 0, 1-\frac{\lambda+1}{\alpha}, 1-\frac{\mu+1}{\beta} \end{matrix}\right), \tag{11}$$

where $A_{\lambda\mu}$ and $B_{\lambda\mu}$ are still given by Eq. (6). The solution for $\alpha>0$ and $\beta<0$ is derived from Eq. (10) by permuting $(\lambda+1)/\alpha$ with $(\mu+1)/\beta$ and $A_{\lambda\mu}$ with $B_{\lambda\mu}$. Equations (10,11) can also be reexpressed as linear combinations of terms involving generalized hypergeometric ${}_2F_2$ functions (see Ref. [13]), but it should be stressed that they are *not* equivalent to Eq. (5). The domains of definitions of the moments for $\alpha<0$ and $\beta>0$, $\alpha<0$ and $\beta<0$ are illustrated in Figs. 1(b) and 1(c). Note that when $\beta\to 0$ and $\mu=0$, Eqs. (10) and (11) reduce to the formula recently derived by Ernst and Szamel [7] for the corresponding one-variable model.

With the exact solutions in hand, one can study the main features of the shattering transition. Consider first the case $\alpha<0$ and $\beta<0$. From the asymptotic properties of the Meijer G functions [13], it is derived that *all* defined moments including total mass and total energy, decrease exponentially with time,

$$M_{\lambda\mu}(t) \simeq \frac{\Gamma(1-A_{\lambda\mu})\Gamma(1-B_{\lambda\mu})}{\Gamma(1-\frac{\lambda+1}{\alpha})\Gamma(1-\frac{\mu+1}{\beta})} e^{-t}, \quad t\to+\infty, \tag{12}$$

which is reminiscent of the behavior in the one-variable model [1,2]. The asymptotic form of $c(x,y,t)$ is obtained by taking the inverse double Mellin transform of Eq. (12). We rather focus here on the various energy-averaged fragments mass distributions, $c_\mu(x,t) = \int_0^1 dy\, y^\mu c(x,y,t)$, which are shown to behave asymptotically, when $t\to+\infty$ and $x\to 0^+$, as

$$c_\mu(x,t) \sim x^{-\alpha-\frac{2}{\mu+1-\beta}} e^{-t}. \tag{13}$$

For $\beta=0$ and $\mu=0$, one recovers the one-variable result of Cheng and Redner [2]. When $2/|\alpha|-|\beta|\leq 0$, the above distributions go to zero in the small-mass limit. In the opposite case, due to energy fluctuations, those with $\mu+1<2/|\alpha|-|\beta|$ diverge. Equation (13) could be used, following the interpretation given by McGrady and Ziff, to define a fractal dimension of the dust [1].

Consider now the case $\alpha<0$, $\beta>0$ (the case $\alpha>0$, $\beta<0$ is obtained by suitable permutations, cf. above). A different type of behavior is observed: of all moments corresponding to the hyperbola $(\lambda+1)(\mu+1)=2$, those with $\lambda+1 \geq \sqrt{2|\alpha|/\beta}$ are conserved, whereas all others decrease with time. This can be rationalized by considering that the disintegration cascade primarily affects the variable x (since $\alpha < 0$ and $\beta > 0$). Because of the correlation between x and y, a singular distribution of fragments appears, but the fragmentation involving y makes the whole process much less effective than when α and β are both negative. There is, thus, some ambiguity in defining the shattering transition. One could choose as its definite signature either the appearance of a singular distribution of fragments or the decrease of $M_{10}(t)$, i.e., an apparent loss of mass. Retaining this latter definition, we can draw the phase diagram of the shattering transition in the (α, β) plane. This is shown in Fig. 2. Although the model is symmetric in x and y, the phase diagram is not symmetric because of the choice of $M_{10}(t)$ to characterize shattering. It is quite remarkable that an apparent loss of mass can be observed when $\alpha > 0$, provided $\beta < -\alpha/2$ (Fig. 2). The shattering transition is then triggered by the disintegration cascade primarily involving the energy y via the correlation between x and y. It represents a spectacular signature of the influence of the large fluctuations of the additional variable (here, y) in our model and has no equivalent in the one-variable case.

The asymptotic behavior of the (defined) moments is again deduced from that of the G functions. For $\alpha < 0, \beta > 0$,

$$M_{\lambda\mu}(t) \simeq \frac{\Gamma(\frac{\mu+1}{\beta})\Gamma(1-A_{\lambda\mu})\Gamma(1+B_{\lambda\mu}-\frac{\lambda+1}{\alpha})}{\Gamma(1-\frac{\lambda+1}{\alpha})\Gamma(1+B_{\lambda\mu}-A_{\lambda\mu})\Gamma(\frac{\mu+1}{\beta}-B_{\lambda\mu})}\, t^{-B_{\lambda\mu}}, \tag{14}$$

and for $\alpha > 0, \beta < 0$,

$$M_{\lambda\mu}(t) \simeq \frac{\Gamma(\frac{\lambda+1}{\alpha})\Gamma(1-B_{\lambda\mu})\Gamma(1+A_{\lambda\mu}-\frac{\mu+1}{\beta})}{\Gamma(1-\frac{\mu+1}{\beta})\Gamma(1+A_{\lambda\mu}-B_{\lambda\mu})\Gamma(\frac{\lambda+1}{\alpha}-B_{\lambda\mu})}\, t^{-A_{\lambda\mu}}. \tag{15}$$

Contrary to the one-variable model and to the present model when $\alpha < 0$ and $\beta < 0$, the moments do not decay exponentially with time. This results from competing effects generated by the opposite signs of α and β in the overall breakup rate. In the shattering region, the total mass $M_{10}(t)$ decreases as a power law of exponent $-(1/\beta + 2/\alpha)$. The asymptotic behavior of the moments, Eqs. (7), (12), (14), (15), may provide a means to check experimentally if additional variables are indeed relevant for a given fragmentation process. From Eqs. (14) and (15), one can also derive the asymptotic behavior of $c(x,y,t)$ and $c_\mu(x,t)$, but we shall present a full discussion of the fragment distribution in another paper [14].

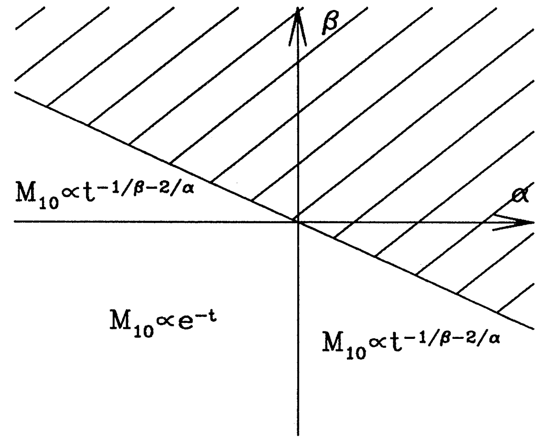

FIG. 2. Phase diagram for the shattering transition [defined by an apparent loss of the mass $M_{10}(t)$]. The hatched region above the line $\beta > -\alpha/2$ represents that part of the diagram where mass is conserved and no shattering transition occurs.

Finally, we note that the important role played by the correlations between variables in the present fragmentation process makes this model a good candidate to look for intermittency in the fragment-mass distribution.

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[1] E.D. McGrady and R.M. Ziff, Phys. Rev. Lett. **58**, 892 (1987); R.M. Ziff and E.D. McGrady, Macromolecules **19**, 2513 (1986); R.M. Ziff, J. Phys. A **25**, 2569 (1992).
[2] Z. Cheng and S. Redner, Phys. Rev. Lett. **60**, 2450 (1988); J. Phys. A **23**, 1233 (1990).
[3] B.F. Edwards, M. Cai, and H. Han, Phys. Rev. A **41**, 5755 (1990); J. Huang, B.F. Edwards, and A.D. Levine, J. Phys. A **24**, 3967 (1991).
[4] B. Elattari, J. Richert, and P. Wagner, Phys. Rev. Lett. **69**, 45 (1992).
[5] R. Botet and M. Ploszajcak, Phys. Rev. Lett. **69**, 3996 (1992).
[6] S.O. Hamphorst and J.K. Leal de Silva, Europhys. Lett. **21**, 261 (1993).
[7] M.H. Ernst and G. Szamel, J. Phys. A **26**, 6085 (1993).
[8] A.F. Filippov, Theory Probab. Its Appl. (USSR) **6**, 275 (1961).
[9] T. Narayanan, J.-M. Petit, M.L. Broide, and D. Beysens (unpublished).
[10] N.R. Corngold, Phys. Rev. A **39**, 2126 (1989).
[11] G. Tarjus and P. Viot, Phys. Rev. Lett. **67**, 1875 (1991); P. Viot, G. Tarjus, S. Ricci, and J. Talbot, Physica A **191**, 248 (1992).
[12] A. Charlesby, Proc. R. Soc. London, Ser. A **224**, 120 (1954).
[13] Y.L. Luke, *The Special Functions and Their Approximations* (Academic Press, New York, 1969), Vol. I.
[14] D. Boyer, G. Tarjus, and P. Viot (unpublished).