Ligament-Mediated Spray Formation

E. Villermaux,[1,*] Ph. Marmottant,[2] and J. Duplat[3]

[1]*Université de Provence and Institut Universitaire de France, IRPHE, 13384 Marseille CEDEX 13, France*
[2]*University of Twente, Department of Applied Physics, 7500 AE Enschede, The Netherlands*
[3]*Université de Provence, IUSTI, 13453 Marseille CEDEX 13, France*

(Received 18 March 2003; published 19 February 2004)

The spray formed when a fast gas stream blows over a liquid volume presents a wide distribution of fragment sizes. The process involves a succession of changes of the liquid topology, the last being the elongation and capillary breakup of ligaments torn off from the liquid surface. The coalescence of the liquid volumes constitutive of a ligament at the very moment it detaches from the liquid bulk produces larger drops. This aggregation process has its counterpart on the shape of the size distribution associated with the ligament breakup, found to be very well represented by *gamma* distributions. The exponential shape of the overall distribution in the spray coincides with the large excursion wing of these elementary distributions, underlying the crucial role played by the ligament dynamics in building up the broad statistics of sprays.

DOI: 10.1103/PhysRevLett.92.074501 PACS numbers: 47.20.Dr, 47.20.Ft, 47.20.Ma

The disintegration and dispersion of a liquid volume by a gas stream is a phenomenon which embraces many natural and industrial operations. The entrainment of spume droplets by the wind over the ocean, the generation of pharmaceutical sprays, or the atomization of liquid propellants in combustion engines are among obvious examples [1,2]. In order to compute the rate of exchanges of solutes between the ocean and the atmosphere or to estimate the size of a combustion chamber, it is frequently desirable to have a precise knowledge of the liquid dispersion structure, in particular, its distribution of droplet sizes as a function of the external parameters.

The broad size statistics is a salient and fundamental feature of natural sprays formed in an uncontrolled way. Spume droplets [3,4], atmospheric aerosols [5], rain drops [6], volcanic Tephra, and fuel sprays [7,8] all display a wide, highly skewed distribution of sizes, the most probable droplet sizes being close to the smallest ones and the probability of finding a drop size 2 or 3 times larger than the mean being not vanishingly small. A generic character of these distributions is an exponential tail at large sizes. For instance, Simmons [7,8] notes that, for a large collection of industrial sprays, the distribution of sizes $p(d)$ is universal in shape and that its tail is well fitted by an exponential falloff. The existing models invoked for this fragmentation process essentially rely on cascade ideas [9], following the early suggestion of Kolmogorov [10], leading to log-normal statistics of the fragment sizes. A notable exception in this context is the work of Longuet-Higgins [11] which shows how a simple geometrical model of ligament random breakup produces broad, skewed size distributions without resorting to sequential cascade arguments, and that of Cohen [12] which shows how pure combinatory and thermodynamic arguments [13] lead to a Poisson distribution for the fragment volumes.

The flow configuration designed to address this problem consists of a round water jet surrounded by a coaxial air flow, an axisymmetric geometry convenient for visualization purposes. Each stream is potential at the exit of

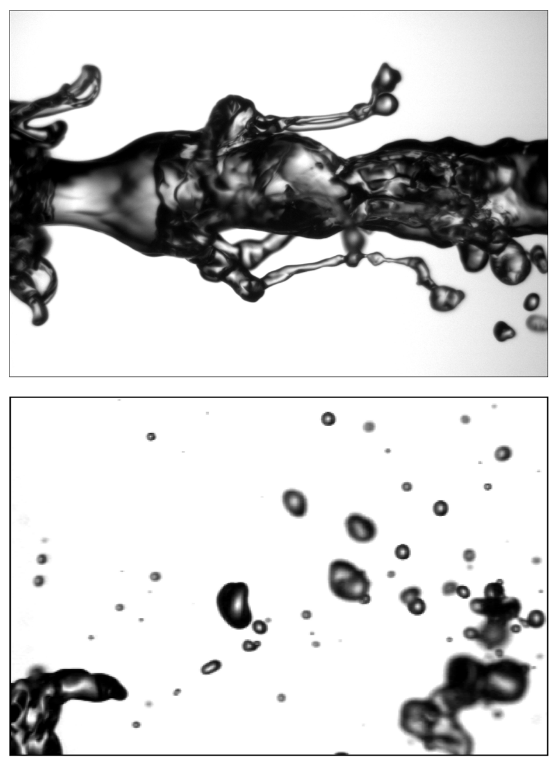

FIG. 1. Top: Instantaneous view of the destabilization of a water jet by a coaxial fast air stream showing the development of an axisymmetric shear instability, the digitations at the wave crests, and the ligament formation. The initial diameter of the liquid jet is 8 mm, its velocity 0.6 m/s, and the velocity of the air stream is 35 m/s. Bottom: Resulting dispersion of droplets in the spray.

the injector, with a very weak residual turbulence. The velocity of the water stream is of the order of 1 m/s and that of the air stream is varied up to $u = 50$ m/s. The spray was analyzed on frozen images using a short time illumination system, and high speed video. As suggested by Fig. 1, at the root of the disintegration process is a shear between the light, fast stream and the slow, dense liquid. Provided that the Weber number $\mathrm{We} = \rho u^2 \delta/\sigma$ and the Reynolds number $\mathrm{Re} = u\delta/\nu$, where ρ and ν denote the density and viscosity of the gas and σ the surface tension of the liquid, are large enough [14,15], this shear induces an instability of a Kelvin-Helmoltz type forming axisymmetric waves on the liquid jet interface. Their wavelength and growth rate are controlled by the thickness δ of the velocity profile in the gas stream [14,16,17]. When the amplitude of these primary undulations is large enough, they undergo a transverse destabilization of a Rayleigh-Taylor type [18,19] caused by the accelerations perpendicular to the liquid-gas interface imposed by the passage of the waves. The resulting modulation of the wave crests is further amplified by the air stream forming ligaments, which ultimately break by capillary instability [20–22]. Provided a ligament is stretched in the wind at a rate which overcomes the rate of the capillary instability $\sqrt{\sigma/\rho d_0^3}$ based on its initial volume $V = d_0^3$, where d_0 depends on δ and We [15], this volume V remains constant as it deforms and finally detaches from the liquid bulk with thickness ξ, a function of the operating conditions (air velocity, liquid surface tension, gas/liquid density ratio). This overall process is usually referred to as "stripping" [23]. Since the ligaments are elongated at breakup (see Figs. 2 and 3), their typical transverse thickness ξ, which is all the more thin as the air velocity is fast, is smaller than d_0.

A key observation is that, although the ligaments are very thin when they detach from the liquid bulk, they give rise to drops whose size is substantially larger (Fig. 3), and which scales similar to d_0. In other words, no matter how thin a ligament is at breakup, it will form drops of the order of the size which sets the ligament volume.

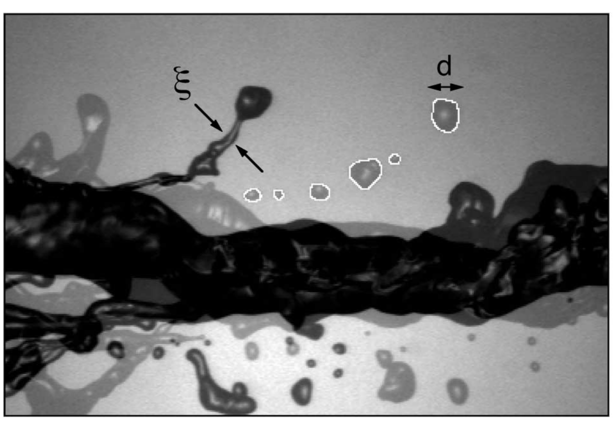

FIG. 2. Double flash exposure of a ligament just before and after breakup. The duration of a flash is 5 μs, and the interval between the two flashes is 1.6 ms. The resulting droplets are highlighted by a white rim.

The situation considered here is fundamentally different from the classical Rayleigh breakup of a liquid thread of uniform thickness in a quiescent environment: The reorganization of the liquid volume in the ligament while it stretches is a superposition of remnant motions from the liquid bulk, motions due to the transient growth and damping of capillary waves [24], motions induced by the deformation of the ligament due to perturbations in the gas stream, etc., These are so complex that they are out of reach of a microscopic analysis. However, capillary forces ultimately fragment the ligament in several blobs. When two liquid blobs of different sizes d_1 and d_2 (with, say, $d_1 < d_2$) are connected to each other, they aggregate due to the Laplace pressure difference $\propto \sigma(1/d_1 - 1/d_2)$. The time it takes for the coalescence to be completed is of order $\sqrt{\rho d_1^3/\sigma}$ which is, also, the time it takes for the neck connecting the two blobs to destabilize and break [21,22]; the confusion of these two time scales induces a nice "coalescence cascade" [25]. For this very reason, the blobs constitutive of the ligament tend, as they detach, to coalesce, thereby forming bigger and bigger blobs along

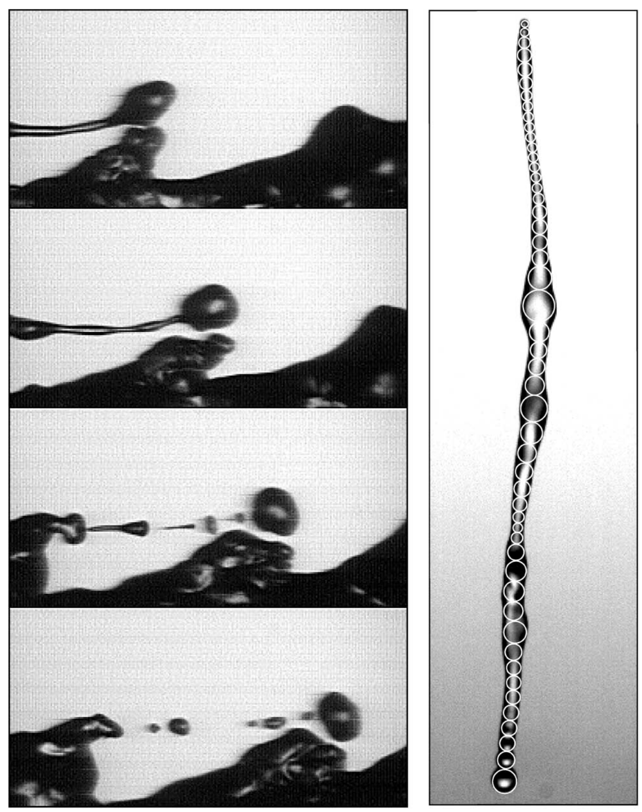

FIG. 3. Left: Time resolved series of the elongation and breakup of a ligament in the wind showing the coalescence between the blobs constitutive of the ligament as it breaks. The time interval between the pictures is 1.34 ms. Right: An isolated ligament just before breakup covered with blobs of various sizes d matching its local thickness. The evolution of the size distribution $n(d, t)$ is governed by Eq. (2).

the ligament. This is why the final mean drop size is larger than the average thickness of the ligament ξ just after it has been released from the liquid bulk. As long as the ligament is attached to the liquid bulk and is stretched in the wind, the capillary instability of its core is strongly damped. This is true for all modes whose instability rate is smaller than the stretching rate [26]. The time it takes for the ligament to detach from the liquid bulk is given by the capillary time based on its initial size $T = \sqrt{\rho d_0^3/\sigma}$. As soon as it has detached and is no more (or much less) stretched, the capillary breakup and coalescence period develop on a shorter time scale of the order of the capillary time based on its thickness ξ.

Our interest is to understand the statistics of the drop sizes in the spray and our observation is that this distribution is determined by the large excursion wing of the distribution associated to a single ligament breakup (Fig. 4). We model the dynamics of the set of traveling waves which overlap at random along the ligament by dividing it into a set of interacting nearby blobs. Let $n(d, t)\mathrm{d}d$ be the number of blobs constitutive of the ligament whose size is within d and $d + \mathrm{d}d$ at time t during the interaction period. The total number of blobs constitutive of the ligament at time t is $N(t) = \int n(d, t)\mathrm{d}d$, its length $L(t) = \int dn(d, t)\mathrm{d}d$, and its volume $V = \int d^3 n(d, t)\mathrm{d}d$ (see, e.g., Fig. 3). Let the random motions in the ligament result in ν independent layers and let

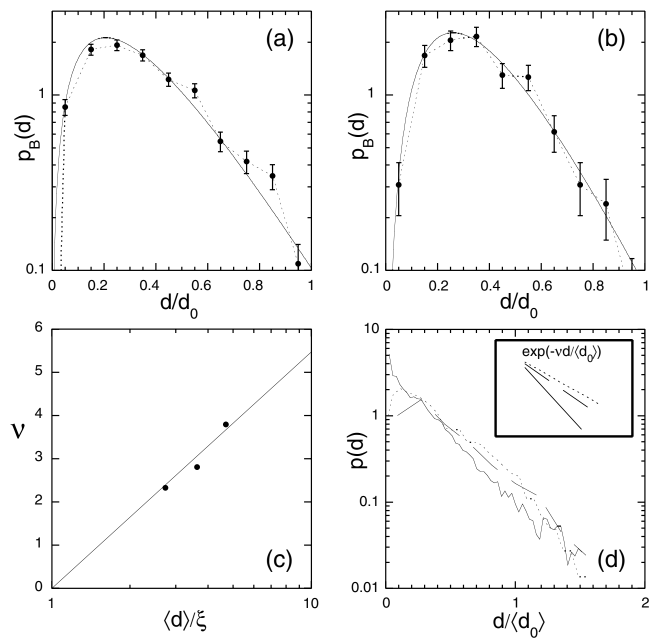

FIG. 4. Droplet size distribution after ligament breakup $p_B(d)$ for an air velocity of (a) 29 m/s and (b) 50 m/s. Lines: fit by gamma distributions. (c) Dependence of the order ν on the ratio of the average droplet size $\langle d\rangle$ to the ligament thickness at breakup ξ. (d) Distribution of droplet sizes in the spray $p(d)$. The slight increase of the exponential slopes with air velocity (inset) reflects the variation of the gamma orders ν on $\langle d\rangle/\xi$.

$q(d', t)$ be the distribution of the sub-blob sizes d' in each layer with $\int q(d', t)\mathrm{d}d' = 1$. The layers are adjacent to each other across the ligament section so that $\nu\langle d'\rangle = \langle d\rangle$ with $\langle d\rangle = \int dn(d, t)\mathrm{d}d/N(t)$. In the course of the coalescencelike process between adjacent interacting fluid particles described above, the sub-blobs overlap and the distribution of sizes in each layer an instant of time later $q(d', t + \Delta t)$ will result from the interaction of blobs of various sizes in the current distribution $q(d', t)$ at time t. The average thickness of the layers $\langle d\rangle/\nu$ is the typical mean-free path of the agitation motions in the ligament. If we conjecture that the interaction is made *at random* with no correlation between the sizes interacting, the evolution of $q(d', t)$ is then directed by a *convolution* process [27]

$$q(d', t + \Delta t) \sim \int q(d' - d_1', t)q(d_1', t)\mathrm{d}d_1' = q(d', t)^{\otimes 2}. \quad (1)$$

Since the layers are assumed independent, the distribution of the blob sizes d itself is thus $n(d, t) = N(t)q(d', t)^{\otimes\nu}$. The corresponding evolution equation for $n(d, t)$ is

$$\partial_t n(d, t) = -n(d, t)N(t)^{\gamma-1} + \frac{1}{3\gamma - 2}n(d, t)^{\otimes\gamma}, \quad (2)$$

with $\gamma = 1 + 1/\nu$. Time t is counted from the moment when the ligament detaches from the liquid bulk ($t = 0$), and is made nondimensional by $T_\xi = \sqrt{\rho\xi^3/\sigma}$, the capillary time based on the initial average blob size $\xi = \langle d\rangle_0$, where $\langle d\rangle_0 = \int dn(d, 0)\mathrm{d}d/N(0)$. The structure of Eq. (2) and prefactors are such that the net volume of the ligament V is conserved. The interaction parameter γ is determined by the compatibility of Eq. (2) with the initial distribution of the blobs along the ligament by $\gamma = \langle d^2\rangle_0/\langle d\rangle_0^2$ with $\langle d^2\rangle_0 = \int d^2 n(d, 0)\mathrm{d}d/N(0)$. A uniform thread of constant thickness (made of many, very thin independent layers) has $\gamma = 1$ and a corrugated ligament is such that $\gamma > 1$.

The asymptotic solution of Eq. (2) for $p_B = n(d, t)/N(t)$ is a *gamma* distribution of order $\nu = 1/(\gamma - 1)$. The very mechanism goes back to the discovery by von Smoluchowski that systems such as Eq. (1) evolving by self-convolution generate exponential distributions [28]. The distribution solution of Eq. (2) is indeed a convolution of ν exponentials, providing [29]

$$p_B(x = d/\langle d\rangle) = \frac{\nu^\nu}{\Gamma(\nu)}x^{\nu-1}e^{-\nu x}, \quad (3)$$

where $\langle d\rangle = \int dn(d, t)\mathrm{d}d/N(t)$ is the current average blob diameter. These gamma shapes closely fit the experimental distributions of drop sizes after ligament breakup with an average diameter $\langle d\rangle \simeq 0.4d_0$ independent of the air velocity [Figs. 4(a) and 4(b)]. The order ν increases slightly with the air velocity [Fig. 4(c)]. These facts indicate that the rearrangements and coalescence between the blobs tend to restore the average diameter $\langle d\rangle$ from ξ

to d_0, or a fraction of d_0. This is made at the expense of a reduction of the number of blobs $N(t)$ which, according to Eq. (2) decreases in time as $N(t)/N(0) = [1 + N(0)^{1/\nu} t/\nu(1 + \nu/3)]^{-\nu}$; concomitantly, the average diameter increases as $\langle d \rangle/\xi = [1 + N(0)^{1/\nu} t/\nu(1 + \nu/3)]^{\nu/3} \sim N(t)^{-1/3}$. At the end of the interaction period between the blobs along the ligament, when $t = O(1)$, the dependence of the resulting average droplet sizes $\langle d \rangle$ on ν presents two distinguished limits. For large ν that is for smooth and uniform ligaments giving rise to a narrow size distribution centered around ξ, one has $\ln(\langle d \rangle/\xi) \sim 1/\nu$. For small ν that is for corrugated ligaments inducing a broad size distribution, one has

$$\ln\left(\frac{\langle d \rangle}{\xi}\right) \simeq \ln[N(0)^{1/3}] + \frac{\nu}{3}. \tag{4}$$

The above anticipated trend is not incompatible with Fig. 4(c); this process interestingly suggests that thinner, but still corrugated, ligaments formed by faster winds, or when the capillary breakup is slowed down by an increased liquid viscosity [22], produce drops with a narrower distribution (the standard deviation of the gamma distribution is $\sim 1/\sqrt{\nu}$), within logarithmic corrections.

For given operating conditions, the diameter d_0 is itself distributed among the population of ligaments, although this distribution $p_L(d_0)$ is narrower than $p_B(d/d_0)$. The size distribution in the spray $p(d)$ is thus a mixture of the distribution of ligament size $p_L(d_0)$ and of the universal distribution of sizes after the ligament breakup $p_B(d/d_0)$ that is $p(d) = \int p_L(d_0) p_B(d/d_0) \mathfrak{d} d_0$. This composition operation stretches the large excursion wing of $p_B(d/d_0)$ over nearly the whole range of sizes d, and the size distributions in the spray shown on Fig. 4(d) thus coincide with the exponential falloff

$$p(d) \sim \exp(-\nu d/\langle d_0 \rangle). \tag{5}$$

The prefactors of the exponential slopes are about 3.5, like the orders ν of the ligament gamma distributions [Fig. 4(c)], and increase slowly with the air velocity, as does the ratio $\langle d \rangle/\xi$ [Fig. 4(c) and Eq. (4)]. The exponential shape of the global distribution and the value of their argument have thus to be understood as the large size behavior, and the order ν of the gamma distributions coming from the ligament breakup process, respectively. That step thus appears as the crucial step building up the broad statistics in the spray.

The fragmentation mechanism we have described which, somewhat surprisingly, consists of a coalescence process, is representative of situations where drops "go with the wind," as in spume or airblast sprays. It is, in fact, generic of all situations where drops come from the capillary destabilization of a strongly corrugated ligament, which was our basic ingredient. In a remote context, the disintegration of big nuclei has been suggested to obey a similar scenario in the celebrated "drop model" for nuclear fission [30]. Since the formation of smaller drops from a liquid volume implies that it shapes in ligaments for capillary forces to produce breakup [20–22], this mechanism might therefore be relevant to sprays formed by splashes or mutual droplet collisions [31]. Those occur, for instance, among drops with different terminal velocities in rain for which the gamma distribution had been identified long ago as a convenient empirical fitting distribution of the drops sizes [5,6].

*Electronic address: villerma@irphe.univ-mrs.fr

[1] L. Bayvel and Z. Orzechowski, *Liquid Atomization* (Taylor & Francis, London, 1993).
[2] A. H. Lefebvre, *Atomization and Sprays* (Hemisphere, New York, 1989).
[3] E. L. Andreas, M. Pattison, and S. E. Belcher, J. Phys. Res. **106**, 7157 (2001).
[4] M. Anguelova and R. P. Barber, J. Phys. Oceanogr. **29**, 1156 (1999).
[5] S. Twomey, *Atmospheric Aerosols* (Elsevier, Amsterdam, 1977).
[6] B. J. Mason, *The Physics of Clouds* (Clarendon, Oxford, 1971).
[7] H. Simmons, J. Eng. Power **7**, 309 (1977).
[8] H. Simmons, J. Eng. Power **7**, 315 (1977).
[9] E. A. Novikov and D. G. Dommermuth, Phys. Rev. E **56**, 5479 (1997).
[10] A. N. Kolmogorov, Dokl. Akad. Nauk SSSR **66**, 825 (1949).
[11] M. S. Longuet-Higgins, Proc. R. Soc. London, Ser. A **439**, 611 (1992).
[12] R. D. Cohen, Proc. R. Soc. London, Ser. A **485**, 483 (1991).
[13] J. Mayer and M. Mayer, *Statistical Mechanics* (Wiley, New York, 1966).
[14] E. Villermaux, J. Propul. Power **14**, 807 (1998).
[15] P. Marmottant and E. Villermaux (to be published).
[16] S. Chandrasekhar, *Hydrodynamic and Hydromagnetic Stability* (Dover, New York, 1961).
[17] L. Rayleigh, Proc. London Math. Soc. **11**, 57 (1880).
[18] L. Rayleigh, Proc. R. Soc. London **XIV**, 170 (1883).
[19] G. I. Taylor, Proc. R. Soc. London, Ser. A **CCI**, 192 (1950).
[20] J. Plateau, *Statique Expérimentale et Théorique des Liquides Soumis aux Seules Forces Moléculaires* (Gauthier Villars, Paris, 1873).
[21] L. Rayleigh, Proc. London Math. Soc. **10**, 4 (1879).
[22] J. Eggers, Rev. Mod. Phys. **69**, 865 (1997).
[23] A. Ranger and J. Nicholls, AIAA J. **7**, 285 (1969).
[24] X. Shi, M. Brenner, and S. Nagel, Science **265**, 219 (1994).
[25] S. Thoroddsen and K. Takehara, Phys. Fluids **12**, 1265 (2000).
[26] I. Frankel and D. Weihs, J. Fluid Mech. **155**, 289 (1985).
[27] S. K. Friedlander, *Smoke, Dust, and Haze* (Oxford University Press, 2000).
[28] M. von Smoluchowski, Z. Phys. Chem. **92**, 129 (1917).
[29] W. Feller, *An Introduction to Probability Theory and its Applications* (Wiley, New York, 1971).
[30] N. Bohr, Nature (London) **143**, 330 (1939).
[31] T. Low and R. List, J. Atmos. Sci. **39**, 1591 (1982).