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Rotating-liquid-drop model limit tested on macroscopic drops

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The collisions of $m = 0.3$–2.0 g, $v = 5$–50 cm/s mercury drops are studied experimentally. A transition between a fusion and a nonfusion outcome is observed, and found to be influenced by angular momentum. The mass dependence measured for the limiting angular momentum L_c(expt) is compared with predictions L_c(theor) of a surface-potential model widely used in nuclear physics. A systematic $L_c(\text{expt}) \leq L_c(\text{theor})$ discrepancy is found in these and in other drop collision data. Dynamical considerations and the use of more elaborate surface shapes than those assumed by the model are found to reduce the disagreement.

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The behavior of liquid drops represents a classical source of scientific inspiration. This interest is partly due to the fact that the scaling properties of fluid systems allow a generalization of certain laws from the largest astronomical objects to the nuclear [1], and even subnuclear [2], level. One such law states that there is a limiting (critical) angular momentum L_c that a rotating drop can stand before it disintegrates, typically, into two smaller pieces [3]. Assuming that the conditions leading to such *fissioning* result from the dominance of the repulsive centrifugal (plus Coulomb for charged drops or atomic nuclei) force over the attraction due to surface tension (plus gravitation for astronomical objects), Cohen, Plasil, and Swiatecki [4] (CPS), developed a scheme to estimate L_c, and its dependence on the drops' mass. To test this *rotating-liquid-drop model* (RLDM) limit it would be necessary to induce a variable angular momentum on isolated drops of different masses. These experimental conditions, difficult to meet for macroscopic drops in an earthly laboratory [5], are characteristic of atomic nuclei. Thus, since the nucleus behaves collectively as a fluid, it is not surprising that the RLDM has been most extensively used in nuclear physics where, among other applications, it has helped in understanding the difficulties encountered in the synthesis of heavy elements [6]. In general terms, however, the need to include a number of corrections characteristic of nuclear systems (finite range [7] and diffuse [8] potentials, detailed structure effects [6], etc.) indicate that the nucleus may not be the best ground to test the *bare* RLDM and, in particular, its prediction for the mass dependence of L_c.

Concerning macroscopic systems, the Spacelab experiments [5] on the behavior of drops spinning in a microgravity environment demonstrated the existence of a critical rotational velocity. As in nuclear reactions, angular-momentum limitations also affect the probability for coalescence of small drops. Indeed, the existence of rotational instabilities has been established while studying the collisions of small drops [9, 10]. However, none of these studies [5, 9, 10] has been directly concerned with determining L_c values.

Here we report an experiment designed to study the mass dependence of the limiting angular momentum for the coalescence, or *fusion*, of liquid drops.

The measurements were carried out with the aid of a liquid-drop collider in which we observe the interactions of equal-size mercury drops moving along a flat, horizontal glass surface, specially treated [11] to minimize the drag induced by wetting. Two drops, each of mass m, are *accelerated* to equal and opposite velocities v, with the aid of plastic ramps fixed on two extremes of the glass surface. A groove on each ramp surface guides the drops down the slopes and smoothly into parallel trajectories separated by an impact parameter b. In this way, the outcome of the drop collisions can be studied as a function of $|v|$, b, and m. The position-vs-time information, needed to determine v and b, is obtained by recording the action with a fast-shutter-speed (1/4000 s) video system having a 30 frames/s recording frequency. The drops' masses m are measured with a 0.1-mg precision analytic scale. The action of every drop collision experiment lasts, typically, 1 s (i.e., 30 frames). The image information on each frame consists on 620 000 color pixels. For simplicity, in the present study this volume of information was reduced to the drop contours on each frame, using standard image processing techniques. Figure 1 shows a typical sequence of 8 frames taken during a drop collision. By studying the time evolution of the kinetic energies of both drops in *binary* collisions (two equal drops in the final channel, as in Fig. 1), we find that the rate of kinetic energy loss during the collision is, at least, an order of magnitude greater than before, or after, the drop-drop contact.

With this experimental setup, we have investigated the influence of angular momentun L on the probability for fusion for 8 different symmetric systems of total masses $M = 2m = 0.6, 1.0, 1.6, 2.0, 2.6, 3.0, 3.6,$ and 4.0 g. For each M value, 24 collisions were recorded for a set of pre-

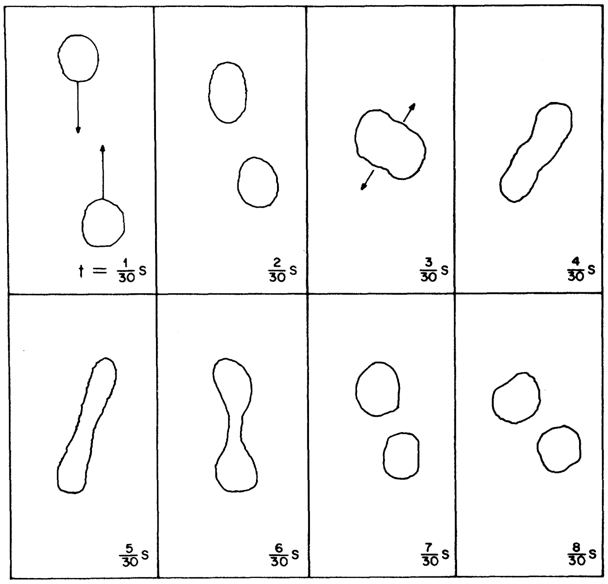

FIG. 1. Time evolution of a *binary* drop-drop collision measured for a symmetric $m = 1$ g system. The arrows in the first frame (top left) indicate the original direction of motion, while in the third frame are drawn to point at the *side splash* of matter in the contact region.

established values of $|v|$ and b based on our calibration of the apparatus. However, to calculate L, the actual values of v and b that the drops have upon contact were extracted from a frame-by-frame analysis of the position of each drop. In this way, a binary variable $N(L)$ was assigned to the outcome of every collision, which distinguished between a *fusion*, $N = 1$, event (only one drop in the final channel) from a *nonfusion*, $N = 2$ (two or more drops) event. Figure 2 shows an example of the results, obtained for $M = 2$ g, in which a fusion-nonfusion transition is observed. A *critical* value L_c was extracted by a $N(L) = 2 - \{1+\exp[(L-L_c)/R)]\}^{-1}$ fit, leaving L_c and R as free parameters. The width of the transition region, measured by the parameter R, is partly due to oscillations induced on the drops by the acceleration procedure (see the initial frames in Fig. 1). Since these shape perturbations have an arbitrary phase relative to the contact time, they broaden the L distribution, increasing the uncertainty in the determination of L_c. The error bar associated with each L_c, which reflects this effect, was taken to be $4.4R$, which is the width of the $1.1 \leq N \leq 1.9$ region. As can be appreciated in Fig. 2, outside this relatively narrow transition zone, L is a determining factor for the outcome of the collisions. The resulting L_c values are given in Table I and plotted as a function of M in Fig. 3.

The observed near-linear dependence of L_c on the drops' mass is reminiscent of the CPS predictions (see Fig. 15 of Ref. [4]) for light nuclei, in the region where Coulomb effects are small. In these [4] calculations, for a given angular momentum, the shape of the two-drop system is assumed to depend on the separation (ρ) between the two drops. Blocki and Swiatecki [12] (BS) have added two new degrees of freedom: a *neck* (λ) variable, and an *asymmetry* (Δ) variable. The former measures the dimensions of the neck connecting the two drops, represented by a quadratic surface of revolution. When dealing with symmetric systems, $\Delta = 0$. Thus, in our case, the value of L_c can be estimated in a gyrostatic approximation [4], by searching in the ρ-λ-Δ ($= 0$) potential-energy surfaces the L values for which the potential-energy pocket disappears. Using this criterium, the L_c values predicted by the BS code [12] for $A \leq 100$ nuclei are very similar to those of the original CPS paper [4]. Hence, we have used the BS code

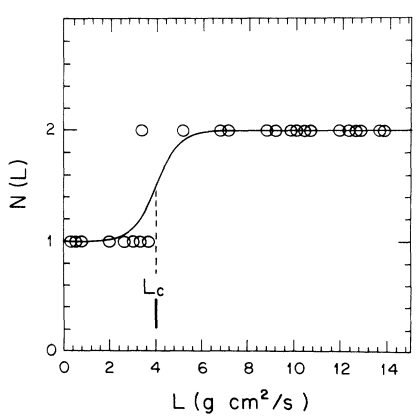

FIG. 2. The outcome $N(L)$ observed for symmetric $m = 2$ g drops as a function of angular momentum L. The $N = 1, 2$ circles represent fusion and nonfusion events, respectively. The curve represents a fit (see text) used to determine the limiting angular momentum L_c.

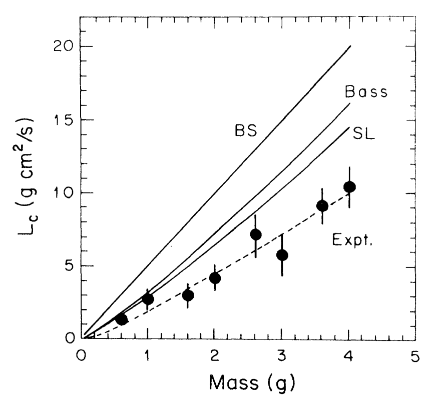

FIG. 3. Measured limit of angular momentum for the fusion of mercury drops as a function of mass (circles). The dashed curve was drawn to guide the eye. The BS curve represents the gyrostatic RLDM calculations obtained with the BS code [12] for spherical drops; our estimate for the effect of an oblate deformation yields results which are indistiguishable from this curve. The Bass curve represents the predictions of the Bass model [18]. The SL curve results from imposing the condition proposed by Schmidt and Lutz [20] on the potential surface predicted by the BS [12].

TABLE I. Limiting angular momenta extracted from the present mercury-drop data, as well as from the available water [9] and propanol-2 [10] data. The liquid densities (ρ_l, in g/cm^3) and surface tensions (σ in dyn/cm) assumed in these calculations were: $\rho_l = 13.0$, $\sigma = 435.0$ for Hg; $\rho_l = 1.0$, $\sigma = 73.5$ for H_2O; and $\rho_l = 0.78$, $\sigma = 21.4$ for propanol-2.

Liquid	M (g)	V_r (cm/s)	L_c(expt) (g cm^2/s)	L_c(theor) (g cm^2/s)	L_c(expt)/L_c(theor)
Hg	0.6	5–50	1.3 ± 19%	2.23	0.58 ± 0.11
Hg	1.0	5–50	2.7 ± 29%	4.05	0.67 ± 0.19
Hg	1.6	5–50	3.0 ± 22%	7.01	0.43 ± 0.09
Hg	2.0	5–50	4.2 ± 24%	9.09	0.46 ± 0.11
Hg	2.6	5–50	7.2 ± 21%	13.35	0.58 ± 0.12
Hg	3.0	5–50	5.8 ± 28%	14.59	0.40 ± 0.11
Hg	3.6	5–50	9.2 ± 14%	18.50	0.51 ± 0.07
Hg	4.0	5–50	10.5 ± 14%	20.42	0.51 ± 0.07
Water	1.8×10^{-6}	220–800	9.5×10^{-7} ± 31%	1.8×10^{-6}	0.53 ± 0.16
Water	2.2×10^{-4}	120–740	3.6×10^{-4} ± 13%	5.0×10^{-4}	0.72 ± 0.09
Propanol-2	3.1×10^{-7}	270–905	1.0×10^{-7} ± 30%	1.5×10^{-7}	0.70 ± 0.21
Propanol-2	8.2×10^{-7}	340–845	2.4×10^{-7} ± 34%	4.7×10^{-7}	0.51 ± 0.17
Propanol-2	3.4×10^{-6}	425–910	1.0×10^{-6} ± 40%	2.2×10^{-6}	0.47 ± 0.19
Propanol-2	6.3×10^{-6}	400–500	2.5×10^{-6} ± 18%	5.3×10^{-6}	0.48 ± 0.09

[12] to obtain the RLDM estimates of $L_c(M)$ for equal, spherical, electrically neutral mercury drops (BS curve in Fig. 3). As can be seen, these RLDM calculations reproduce the near-linear mass dependence but overestimate L_c by a factor of ≈ 2. We now investigate some of the possible reasons for this large discrepancy.

First, an experimental aspect to be considered is the fact that mercury drops lying on a horizontal glass surface assume shapes which are closer to oblate spheroids than to spheres. The systematics of the BS calculations have been used to estimate the influence of an oblate-spheroidal deformation on $L_c(M)$ as follows. Within the L-value range of interest here (Fig. 3), and for $\Delta = 0$, the BS model predicts that the saddle-point in the ρ–λ potential surface (which determines the fusion-nonfusion transition) remains at an approximately fixed position (ρ_c, λ_c), lying close to the two-separate-drop shape limit. Using the observed [11] experimental shapes, a unidimentional model has been built in which the horizontal projections of the drops coincide with those of the corresponding spherical case (i.e., adopting a shape characterized by ρ_c, λ_c in the same region), while the vertical projections are approximated by two ellipses joined by a quadratic surface, thus generating elliptical cross section necks. The L_c values estimated in this way are only $\approx 1\%$ smaller than those predicted by the BS for spherical drops. This insensitivity of the predictions to the shape changes is due to the near cancellation between two opposite effects. First, compared with the spherical case, the deformed drops are subject to a less attractive surface tension potential, which has the effect of decreasing L_c. However, the oblate deformation implies an increment in the drop dimensions along the contact plane. Hence, relative to the spherical case, the drops touch at a larger distance, where the centrifugal force has smaller values, having the effect of increasing L_c. Thus, we are led to conclude that this type of deformation may not be the main source of the large discrepancy shown in Fig. 3 between theory and experiment.

Independent evidence for a systematic overestimation of L_c by the gyrostatic RLDM estimates [12] may be found in the data of Adam, Lindblad, and Hendricks [9] for spherical, electrically charged, water drops of $m = 0.9$ and 113 μg, and in the data of Brenn and Frohn [10] for spherical, uncharged, propanol-2 drops of $m = 0.15, 0.4, 1.7$, and 3.3 μg. These authors [9,10] studied the fusion-nonfusion transition on collisions of equal-size drops, as a function of the relative velocity v_r and impact parameter b. According to Natowitz and Namboodiri [13], and to Griffin and Wong [14], that transition is due to two independent effects: rotational instabilities, presumably related to L_c, and vibrational instabilities. The latter were introduced to explain a back-bend in the fusion-nonfusion transition observed in the water-drop data [9] for collisions at large v_r and small b values (see Figs. 4 and 6 of Ref. [9]). Thus, excluding the b, v_r pairs in the back-bend region, we have used the water data [9] and the propanol-2 data [10] to extract average L_c values by fitting a $b = \frac{2L_c}{mv_r}$ relationship. These experimental results [L_c(expt)] are included in Table I, together with the corresponding predictions L_c(theor) from the BS code [12] for spherical (and, when appropriate [9], charged) drops. The quoted uncertainties are equal to the standard deviation of the L_c's, obtained from each b, v_r pair, relative to the mean. The values of surface tensions and densities used are indicated in the table caption. The last column shows the ratio between the experimental and the theoretical L_c values, illustrating that the magnitude of systematic deviation is similar to what we found for mercury drops. A common feature of the data shown in the Table I values is that the corresponding experiments were carried out under the action of a small external (drop-glass and/or aerodynamic) retarding force. However, this implies that, for a fixed impact parameter, the initial velocities need to be incremented accordingly to obtain the same outcome, indicating that the discrepancy with the RLDM estimates may be even larger.

There are several theoretical aspects which could help explain the discrepancy between the RLDM estimates and the drop-collision measurements. One of them is the need for dynamical (viscous friction, shape evolution, etc.) effects. This deficiency of gyrostatic RLDM calculations [4] is well known in nuclear physics [15]. A macroscopic approach [16] to treat this problem has been to assume a viscous friction mechanism and then to solve the dynamical equations following the time evolution of the nuclear collisions. This procedure systematically yields lower fission barriers [15] and, thus, predicts smaller L_c values. However, these calculations [16] would be difficult to adapt to macroscopic fluids as their dynamics are based on a *one-body* [16] dissipation mechanism characteristic of nuclear reactions at low incident energies [17]. Still, the extent of the reduction of L_c due to dynamics can be illustrated (Bass curve in Fig. 3) by the use of a simple model proposed by Bass [18] in which a unidimensional potential is combined with a sharply localized friction approximation.

We also find that, when applied to drops, the two-dimensional shapes assumed by the potential-energy surface calculations [4, 12] are inadequate to describe the evolution of the collisions, particularly in their initial stages. This is illustrated in Fig. 1 (see the third frame) showing a side splash, characteristic of colliding hydrodynamical systems [19], occurring in the contact region. Note that the intersecting-spheres configurations, which would contain the splash shapes, are specifically excluded in the ρ-λ-Δ potential-energy surfaces in the BS calculations [12]. In more general terms, the complex shapes adopted by colliding drops imply the use of a multidimensional surface potential. Schmidt and Lutz [20] (SL) have recently proposed that the effect of those complex deformations on L_c can be estimated using a *shallow potential* approximation, in which the limiting L would be reached when the fission barrier measured at the saddle point equals the total collective energy of the spherical complex. This calculation [20] yields L_c estimates (SL curve in Fig. 3) which are systematically 30% smaller than those predicted by the pocket disappearing condition in surface potential calculations [4, 12].

The above arguments indicate that both the dynamical aspects and the more complex surface shapes are expected to have important reducing effects on the L_c predictions. Considering nuclear collisions, since the splashlike deformations are external signs of a two-body fluid incompressibility, we believe that more complicated shapes in dynamical potential-energy surface calculations [16] would be necessary, at least in the incident energy range where this type of friction dominates [17].

In summary, we have studied the mass dependence of the limiting angular momentum L_c for the fusion of 0.3 g$\leq m \leq$ 2.0 g mercury drops moving on a rough glass surface which minimizes the effect of wetting. When compared with the predictions of gyrostatic calculations using the model of Blocki and Swiatecki [12], the experimental L_c values are found to be systematically smaller that the predictions. This discrepancy also holds for L_c values extracted from available data on water and propanol-2 drop collisions. As in nuclear physics, this overestimation of L_c is an indication of the need for dynamical considerations and, in the particular case of drops, of the need to consider more complex surface shapes.

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