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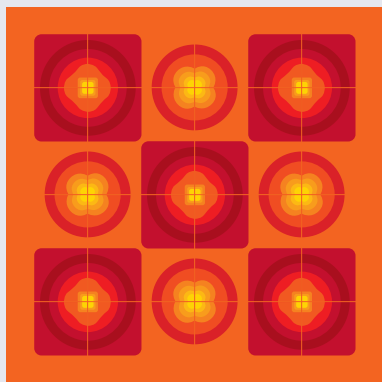
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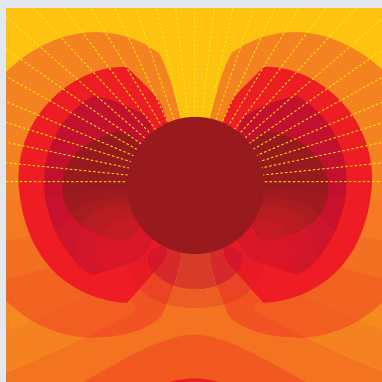
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Image: Ornamental multiplication of space-time figures of temperature transformation rules (adapted from T. S. Bíró and P. Ván 2010 EPL **89** 30001; artistic impression by Frédérique Swist).

A test-tube model for rainfall

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Abstract – If the temperature of a cell containing two partially miscible liquids is changed very slowly, so that the miscibility is decreased, microscopic droplets nucleate, grow and migrate to the interface due to their buoyancy. The system may show an approximately periodic variation of the turbidity of the mixture, as the mean droplet size fluctuates. These precipitation events are analogous to rainfall. This paper considers a theoretical model for these experiments. After nucleation the initial growth is by Ostwald ripening, followed by a finite-time runaway growth of droplet sizes due to larger droplets sweeping up smaller ones. The model predicts that the period Δt and the temperature sweep rate ξ are related by $\Delta t \sim C\xi^{-3/7}$, and is in good agreement with experiments. The coefficient C has a power-law divergence approaching the critical point of the miscibility transition: $C \sim (T - T_{\rm c})^{-\eta}$, and the critical exponent η is determined. It is argued that while the mechanism does not provide a quantitative description of terrestrial rainfall, it may be a faithful model for precipitation on other planets.

Introduction. – Rainfall is a cyclic process, in which water vapour builds up in the atmosphere, reaches supersaturation, and then forms droplets of visible moisture which appear as clouds. The microscopic droplets can grow by collisions and coalescence until they are large enough to fall as rain, which removes the moisture from the atmosphere. The same mechanisms will be applicable on other planets, but the role of water could be taken by other small molecules. Many different processes may be involved [1,2], and a quantitative understanding of the rainfall cycle has been lacking. In terrestrial rainfall, the growth of droplets through the size range 15–$50\,\mu$m is considered to be a "bottleneck" in the kinetics of rain initiation [2], for which the mechanism is not yet fully understood. It is valuable to investigate simplified models which can serve as a benchmark against which complex phenomena can be compared. As well as being important for terrestrial meteorology, such models will be particularly important in analysing weather phenomena on other planets, where the composition and physical parameters of the atmosphere are relatively uncertain [3,4].

This paper discusses a laboratory model which includes the essential features of the rain cycle. Two partially miscible liquids are placed in a small cell, where they form two layers. The cell is placed in a computer-controlled thermostat, and its temperature is varied smoothly away from the phase coalescence temperature $T_{\rm c}$, at a rate described by a variable ξ which has dimensions of inverse time (defined by eq. (4) below). Several examples of this type of experiment have been reported: see, for example, refs. [5–9]. The experiments show similar qualitative results: both layers show a variation in turbidity, which is approximately periodic, with period Δt. Periods of high turbidity end with microscopic droplets of fluid floating up or down towards the interface. In general the upper and lower layers have different periods, $\Delta t_{\rm t}$ and $\Delta t_{\rm b}$, respectively. These periodic precipitation events are analogous to rainfall, in that they result from a runaway growth of droplets mediated by collisions resulting from a size-dependent sedimentation rate. It is argued that while the initial mechanism of droplet growth in these experiments (Ostwald ripening) is too slow to account for terrestrial rainfall, the mechanism is likely to be a good description of rainfall in other planetary systems.

The experimental results reported in these earlier works do not span a sufficiently wide range of parameters to definitively test quantitative predictions about how the period Δt depends upon the heating rate ξ. However, a recent series of experiments reported in [10] (see also [11])

explores a very wide range of values of the experimental parameters, and is consistent with

$$\Delta t = C \xi^{-3/7}, \tag{1}$$

where the multiplier C is a function of the mean temperature T during the period, and is predicted to become independent of the height h of the layer when this is larger than some characteristic scale, h_0. There is a slowing of the dynamics at the critical point, with C having a power-law divergence approaching the miscibility transition at temperature T_c: the data in [10] are consistent with

$$C \sim (T - T_c)^{-\eta}, \qquad \eta \approx 0.50. \tag{2}$$

Here I present the theoretical model leading to eq. (1), and discuss how C is determined from the thermodynamic and kinetic coefficients of the mixture. The value of the exponent η in eq. (2) is determined from standard critical exponents.

The theory uses the Lifshitz-Slezov [12,13] theory of Ostwald ripening to describe the initial growth of droplets after they are nucleated. When the droplets reach a certain size, their motion due to the difference in density from the surrounding fluid becomes significant. Buoyancy drives droplets towards the interface, with larger droplets catching up and coalescing with smaller drops. As droplets grow, their velocity towards the interface increases, which further increases the rate at which they sweep up smaller droplets. The model predicts a divergence of the droplet size in finite time, similar to that which occurs in the evolution of rain droplets [14]. The macroscopic droplets sweep up all of the microscopic droplets in their path, leaving a clear solution which is in equilibrium, so that the cycle can be repeated.

The experimental system. – The system is a small transparent cell containing two liquids (water and isobutoxyethanol were used in [10,11]), which are only partially miscible above a critical temperature T_c. The phase equilibrium line for the two phases is a line in the Φ-T plane, where $0 \le \Phi \le 1$ is the volume fraction of the denser component. Above T_c the coexistence curve has two branches, denoted $\Phi_b(T)$ and $\Phi_t(T)$ for the bottom (denser) and top phases, respectively.

As the system is heated, the miscibility decreases, implying that the minority component will come out of solution in each phase. If the temperature change is extremely slow, it may diffuse across the boundary, but as the system size is increased the material must precipitate out, no matter how slow the temperature change. It is assumed that, immediately after a precipitation event, the two layers are in equilibrium at points on the phase diagram with compositions Φ_b and Φ_t and with volumes V_b and V_t. As the system is slowly heated, the condition for equilibrium is no longer satisfied. The resulting supersaturation is relieved by the nucleation of microscopic droplets. A temperature difference δT causes a change in the equilibrium composition by $\delta\Phi_b < 0$ and $\delta\Phi_t > 0$. Because the change is too

rapid to allow significant diffusion across the interface, the change in the volume of the lighter component in the lower layer is $\delta v_b = 0$. This change has contributions from the amount of the lighter component in solution, and from the formation of droplets of total volume δV_d, so that

$$\delta v_b = 0 = (V_b - \delta V_d)\Phi_b + \delta V_d \Phi_t + V_b \delta\Phi_b - V_b\Phi_b. \tag{3}$$

The most convenient measure of the rate of temperature change is the rate of change of the volume fraction occupied by the droplets. For the lower phase, this is

$$\xi_b \equiv \frac{1}{V_b}\frac{\delta V_d}{\delta T}\frac{dT}{dt} = \frac{1}{\Phi_b - \Phi_t}\frac{d\Phi_b}{dT}\frac{dT}{dt}. \tag{4}$$

A model. – Now consider a model for the cyclic fluctuations of turbidity. Assume that at time t_0 a "rain" event has occured and that this leaves a solution which is well-mixed and very close equilibrium, with negligible supersaturation and with no visible particles in suspension. It is assumed that there are always sufficient nucleation centres that material can come out of equilibrium as the temperature increases and reduce the supersaturation of the solution so that it is always small. The resulting droplets of denser fluid will cause turbidity of the suspension if they are sufficiently large, but while they are small compared to the wavelength of light they might not be evident in optical observations. The sub-microscopic droplets will grow as the apparent supersatuation increases, so that at $t = t_0 + \delta t$ there is an excess volume fraction $\xi\delta t$ in the form of sub-microscopic droplets.

Consider how to model the evolution of the radius of a droplet, $a(t)$. After the droplets are nucleated they grow by diffusion of supersaturated material onto their surface. As the droplets grow, the level of supersaturation diminishes and the smallest droplets become unstable, because surface tension increases their Laplace pressure. They are re-absorbed into the solution and the material they contained diffuses onto the surface of the larger droplets. This process is an example of "Ostwald ripening", and it was first given a satisfactory treatment by Lifshitz and Slezov [12]. Their account treats a quenched solution, in which the total amount of excess material in the supersaturated solution (having a volume fraction V_d) is a constant. In our problem the amount of excess material increases linearly with time: $V_d = \xi(t - t_0)$. This will result in a minor modification to the Lifshitz-Slezov theory when it is applied to the experiment.

The essential features of the Lifshitz-Slezov approach can be summarised as follows. For the sake of simplicity, a dilute solution approximation is used, which is analytically tractable. The interior of a droplet of radius a has a pressure which is higher than the ambient pressure by $\Delta p = 2\sigma/a$, where σ is the surface tension at the phase boundary. This increased pressure implies that the minority component must have a higher concentration in order to be in equilibrium: the chemical potential must be increased by $\Delta\mu = \Delta p v$, where v is the molecular volume of the minority component. Using a dilute solution

approximation, this increase in chemical potential is created by an increase in the volume fraction, $\Delta\Phi$, satisfying $kT\Delta\Phi/\Phi_e = \Delta\mu$, where Φ_e is the equilibrium concentration. The surface of a droplet of radius a is therefore in contact with a layer of solvent with a volume fraction

$$\Phi(a) = \Phi_e + \frac{\Lambda}{a}, \qquad (5)$$

where Λ is a Kelvin length, defined by

$$\Lambda = \frac{2\sigma v \Phi_e}{kT} = \frac{2\sigma V_m}{RT}\Phi_e \qquad (6)$$

(here V_m is the molar volume of the minority component, and R is the universal gas constant). The concentration in the vicinity of each droplet is in quasi-equilibrium and may be approximated by a solution of Laplace's equation. The concentration far from a droplet is $\Phi(\infty) = \Phi_e + s$, where s is the supersaturation of the solution. The concentration at a distance r from the nearest droplet is approximated by

$$\Phi(r) = \Phi_e + s + \frac{\Lambda - sa}{r}. \qquad (7)$$

There is a diffusive flux of material onto the droplet surface, which causes its radius to change at a rate

$$\frac{da}{dt} = D\frac{\partial\Phi}{\partial r}\bigg|_{r=a} = \frac{D\Lambda}{a}\left(\frac{1}{a_0} - \frac{1}{a}\right) \qquad (8)$$

with $a_0 = \Lambda/s$, where D is the interdiffusion coefficient of the two components. The interpretation is that droplets smaller then a_0 shrink under the effects of the Laplace pressure, and those larger than a_0 grow by absorption of material evaporating from the smaller droplets. The supersaturation must decrease as a function of time so that the largest droplets can continue to grow. It follows that a_0 is comparable to the typical droplet size (and in fact the Lifshitz-Slezov theory predicts that $\langle a \rangle = a_0$ [12]). For the growth of the largest droplets, the a_0 term in (8) can be neglected, implying that $a^3 \sim D\Lambda t$. In fact the Lifshitz-Slezov theory predicts that the mean droplet size is [12]

$$\langle a(t)\rangle = \left[\frac{4}{9}D\Lambda(t - t_0)\right]^{1/3}. \qquad (9)$$

Here the dynamics is a little different from the standard Ostwald ripening process, where the system is quenched at a fixed temperature. In that case the amount of material which comes out of solution is independent of time, and the number density of droplets decreases due to evaporation as the mean droplet size increases. In the case treated here, however, the amount of material which comes out of solution is proportional to time. Together with a growth law of the form $a \sim t^{1/3}$, this implies that the droplet number remains approximately constant in the long-time limit, until coalescence driven by gravitational settling becomes important. The droplet density created by the nucleation process is set by competitive growth in the early stages, but at large times droplets do not evaporate and their number density approaches a constant n_0. Equating the volume fraction of the droplets, $\xi(t - t_0)$, to $4\pi n_0\langle a^3\rangle/3$, eq. (9) indicates that the number density of nucleated droplets is $n_0 \sim \xi/(D\Lambda)$.

The diffusive growth of droplets continues until other effects become significant, creating macroscopic motion. The first such effect to become relevant is when droplets start to settle towards the interface due to their density contrast. The sinking velocity in the upper layer, u_t, may be estimated by balancing gravitational forces and viscous drag. Using the Stokes formula for the drag on a small sphere gives $\frac{4\pi}{3}(\rho_b - \rho_t)ga^3 = 6\pi\rho_t\nu_t au_t$, where ν_t is the kinematic viscosity of the lighter fluid, and where the densities of the lighter and heavier phases are ρ_t and ρ_b, respectively. In the following $\Delta\rho = \rho_b - \rho_t$. The settling speed is

$$u_t = \frac{2}{9}\frac{\Delta\rho g}{\rho_t \nu_t}a^2 \equiv \kappa_t a^2. \qquad (10)$$

The gravitational settling allows the rate of accumulation of material on a droplet to increase because the larger droplets, which fall at a faster rate, can overtake the smaller droplets. If droplets collide, they can merge, and the merged droplet will fall at an even larger rate. This sweeping process will be modelled by assuming that a droplet has already grown to a size $a(t)$ where it is much larger than the other droplets in its path. The settling velocity of the smaller droplets may be neglected. The larger drop moves through a "gas" of smaller droplets which occupy a volume fraction $\xi(t - t_0)$, and they cause the lower surface to sweep up a volume per unit time equal to $\dot{v}_d = \pi a^2 u\epsilon\xi(t - t_0)$, where ϵ is a collision efficiency. This rate of growth of the volume implies a rate of growth of the droplet radius given by $\dot{v}_d = 4\pi a^2\dot{a}$, so that the droplet radius grows by particle accretion at a rate $\dot{a} = \xi t\epsilon u/4$, where u is the settling speed (given by (10) for the droplets sinking in the upper layer).

Now consider a model for the time evolution of the radius of the largest droplets. Noting that $a_0 \sim \langle a\rangle$, growth of the largest droplets may be described by the relation $\dot{a} \sim D\Lambda/a^2$. Combining this with the relation for growth by sweeping gives the model equation

$$\frac{da}{dt} = \frac{D\Lambda}{a^2} + \frac{\epsilon}{4}\kappa a^2\xi t. \qquad (11)$$

This equation suggests that the evolution of the droplets can be divided into two stages, depending upon which of the two terms in the expression for $\dot{a}$ is dominant. The first stage, involving growth by Ostwald ripening and diffusional accretion, lasts for a time t_1, which is determined by the condition that the two terms in (11) become equal. This condition is satisfied when the droplets reach a size a_1. From this point on it will be assumed that the collision efficiency is of order unity, and other dimensionless constants will be dropped. The condition for the

crossover is

$$\frac{D\Lambda}{\kappa\xi} \sim a_1^4 t_1. \tag{12}$$

However, from (9) it is found that $a_1^3 \sim D\Lambda t_1$, so that solving for t_1 gives $\frac{D\Lambda}{\kappa\xi} \sim (D\Lambda t_1)^{4/3} t_1$, and hence

$$t_1 \sim \left(\frac{1}{D\Lambda(\kappa\xi)^3}\right)^{1/7}. \tag{13}$$

In the second stage, eq. (11) for droplet growth may be approximated by $\dot{a}/a^2 = \kappa\xi(t - t_0)$. This has a solution (with initial condition $a = a_1$ at $t = t_1 + t_0$)

$$\frac{1}{a_1} - \frac{1}{a} = \frac{\kappa\xi}{2}\left[(t - t_0)^2 - t_1^2\right]. \tag{14}$$

According to this solution, $a(t)$ diverges in a finite time, so that $a \to \infty$ as $t \to t_1 + t_2$, with t_2 having the same scaling as t_1. In summary, the model predicts that the period $\Delta t = t_1 + t_2$ is

$$\Delta t \approx \alpha \left(\frac{1}{D\Lambda\kappa^3}\right)^{1/7} \xi^{-3/7} \tag{15}$$

where α is a dimensionless prefactor. Because of the finite-time divergence of the droplet sizes, the timescale for the oscillations is not predicted to depend upon the size of the cell. But the cell does need to be large eonugh for the finite-time runaway growth to happen.

In the analysis leading to (15) it was assumed that the collision efficiency is close to unity. This may appear surprising, because the low collision efficiency of small water droplets in air is acknowledged to be a barrier to the initiation of rainfall [15]. The low collision efficiency arises because colliding water droplets slide over each other, lubricated by a thin film of air trapped between them. In the test-tube precipitation experiments [5–11], however, the physics of the coalescence process is very different. This is because the binary mixture is always close to its critical point, where the correlation length diverges and the interfacial surface tension vanishes. Because the interface in these experiments is diffuse on the molecular scale, efficient coalescence of droplets can occur by evaporation of the trapped layer into the droplets.

Equation (15) was compared with experimental data in the PhD Thesis of Lapp [10], where it was shown to give a very good description of the experimental data for water/isobutoxyethanol. The rate of change of order parameter ξ was varied over nearly three decades and the temperature relative to the critical point, $T - T_c$, by nearly three decades. Another observation which supports the theory is that the periods of high and low turbidity are of a comparable extent, which is consistent with the theoretical prediction that $t_1 \sim t_2$.

The experimental data in [10] show that the period Δt diverges as we approach the critical temperature, T_c, in accordance with eq. (2), with the exponent η taking the same value for both layers. This divergence is a consequence of the fact that the coefficient C contains three factors which vanish at the critical point. The density difference between the phases, $\Delta\rho$, has the critical behaviour of an order parameter, with exponent β. The critical exponent for the interfacial surface tension, σ, can be related to the correlation length exponent ν [16]. The van Hove theory of phase transitions indicates that the critical exponent of the interdiffusion coefficient is the susceptibility exponent γ [17] (a different value has been proposed [18], but γ appears closer to experimental measurements [19] on the system used in [10]). For a three-dimensional system with a one-component conserved order parameter, the exponents are

$$\Delta\rho \sim |T - T_c|^\beta, \qquad \beta \approx 0.327,$$
$$\sigma \sim |T - T_c|^{2\nu}, \qquad \nu \approx 0.630,$$
$$D \sim |T - T_c|^\gamma, \qquad \gamma \approx 1.237. \tag{16}$$

From eqs. (6), (10) and (15), the critical exponent for the coefficient C is

$$\eta = \frac{3\beta + 2\nu + \gamma}{7} \approx 0.498 \tag{17}$$

which is also in satisfactory agreement with the experimental results [10].

Discussion. – This letter has presented a theory to account for periodic precipitation in binary mixtures, as observed in [5–11]. The theory gives very good agreement with observations of the most thoroughly explored system, described in [10].

It has been argued that the periodic precipitation phenomenon seen in these experiments is analogous to a precipiation cycle in a stable atmosphere. Consider whether the same mechanisms are relevant to the growth of real rain droplets. It has been argued [1] that there is a "bottleneck" in the growth kinetics of raindrops at radii in the range $a \approx 15$–$50\,\mu$m, between smaller particle sizes where growth by condensation is efficient, and larger particle sizes where collisions due to gravitational settling become important. The Ostwald ripening process may allow this gap to be bridged, because it allows droplets to grow by diffusive transfer of material from smaller droplets, without the necessity for collisions. Note that the growth law, given by eq. (9), is independent of the density of the droplets. The physical parameters which are required to estimate the growth law are, at $10\,°$C, diffusion constant for water vapour in air, $D = 2.4 \times 10^{-5}\,\mathrm{m^2\,s^{-1}}$, surface tension $\sigma = 7.4 \times 10^{-2}\,\mathrm{Nm^{-1}}$, molar volume $V_m = 1.8 \times 10^{-5}\,\mathrm{m^3}$, saturation volume fraction $\Phi_0 = 1.2 \times 10^{-5}$. These give a Kelvin length $\Lambda = 1.4 \times 10^{-14}\,$m and consequently, using (9), the growth law is $a(t) \approx 6 \times 10^{-9}(t/\mathrm{s})^{1/3}$. For water droplets in the air, losing latent heat by conduction is another significant effect, and D must be replaced by an effective diffusion coefficient $D_{\mathrm{eff}} < D$ [15]. These estimates imply that growth to a radius of $25\,\mu$m by Ostwald ripening requires several days,

so that Ostwald ripening is too slow to be important for terrestrial clouds. Clement [20] reached the same conclusion about the relevance of Ostwald ripening to terrestrial rainfall. It is acknowledged to be difficult to explain the initiation of rainfall from cumulus clouds because of the low collision rates of the smallest droplets (below $30\,\mu m$ radius). This implies that non-collisional processes may be very important. These estimates indicate, however, that Ostwald ripening is not the relevant non-collisional process for rainfall from warm cumulus clouds.

Finally, a speculative remark. Extra-solar planets are being discovered at a prodigious rate [21], and techniques are being developed to identify their atmospheric composition [3,4]. The sizes and orbital parameters of these planets are highly variable, and it seems likely that many of them will exhibit exotic weather phenomena. On Earth, rainfall is chaotic and unpredictable. Other planets, however, may have temporally periodic rainfall events driven by Ostwald ripening, analogous to the test-tube model considered here.

$$* * *$$

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