chaos on a molecular level quite plausible, the observed macroscopic disorder cannot be taken as direct evidence of microscopic chaos. The effectively infinite number of molecules in a fluid can generate the same macroscopic disorder without any intrinsic instability, so brownian motion can be derived for systems that would usually be called non-chaotic, such as a tracer particle in a non-interacting ideal gas. All that is needed for diffusion is 'molecular chaos' in the sense of Boltzmann, that is, the absence of observable correlations in the motion of single molecules.

Part of the confusion is due to the lack of a unique definition of microscopic chaos for systems with an infinite number of degrees of freedom. Gaspard *et al.*[1] introduced the term by extrapolating from finite dimensional dynamical systems for which chaos is well defined: on average, initially close states separate exponentially when time tends to infinity. The Lyapunov exponent, or rate of separation, is independent of the particular method used to measure 'closeness'. However, the ideas of diffusion and brownian motion involve infinitely many degrees of freedom. In this thermodynamic limit, Lyapunov exponents are no longer independent of the metric. The large system limit of a finite non-chaotic system will therefore remain non-chaotic with one particular metric and become chaotic with another.

We can illustrate this point by using an example[6] introduced in the context of cellular automata. Consider two states $\mathbf{x}=(\ldots x_{-2}, x_{-1}, x_0, x_1, x_2\ldots)$ and $\mathbf{y}=(\ldots y_{-2}, y_{-1}, y_0, y_1, y_2\ldots)$ of a one-dimensional bi-infinite lattice system. If the distance between $\mathbf{x}$ and $\mathbf{y}$ is defined by $d_{\max}(\mathbf{x},\mathbf{y})=\max_i|x_i-y_i|$, it can grow exponentially only if the local differences do. This is what is usually meant by 'chaos', and what Gaspard *et al.*[1] meant by 'microscopic chaos'. This mechanism is absent in the thermodynamic limit of finite non-chaotic systems, so the limit could also be said to be non-chaotic[7–9]. However, the distance $d_{\exp}(\mathbf{x},\mathbf{y})=\sum_i|x_i-y_i|e^{-|i|}$ can also show exponential divergence if an initially distant perturbation moves towards the origin without growing[6]. When observing a localized tracer, as Gaspard *et al.*[1] did, the latter choice may be the more appropriate.

In finite dimensional dynamical systems, chaos arises as a result of the defocusing microscopic dynamics. Positive entropy is generated by the initially insignificant digits of the initial condition brought forth by the dynamics. At the thermodynamic limit, a different mechanism also exists: perturbations coming from distant regions kick the tracer particle once and move away again to infinity. The entropy is positive because of information stored in remote parts of the initial condition. Suitable Lyapunov exponents[6] can be defined for this case as well.

To resolve the confusion, we propose letting the system size tend to infinity first, before the observation time[10]. A system observed in a particular metric μ is then described as μ-chaotic when we find a positive Lyapunov exponent using this metric. Gaspard *et al.*[1] had in mind the type of chaos that is detectable with the metric $d_{\max}$ and arises from a local instability. But they fall short of providing experimental evidence for this specific type of chaos.

Peter Grassberger, Thomas Schreiber
Department of Physics, University of Wuppertal, 42097 Wuppertal, Germany

1. Gaspard, P. *et al. Nature* **394**, 865–868 (1998).
2. Gaspard, P. & Wang, X.-J. *Phys. Rep.* **235**, 291–345 (1993).
3. Dürr, D. & Spohn, H. *Nature* **394**, 831–833 (1998).
4. Schewe, F. & Stein, B. *Physics News Update* <http://www.aip.org/enews/physnews/1998/split/pnu389-3.htm> (1998).
5. Rauner, M. *Physik. Blätt.* **54**, 1001 (1998).
6. Wolfram, S. *Physica D* **10**, 1–35 (1984).
7. Crutchfield, J. & Kaneko, K. *Phys. Rev. Lett.* **60**, 2715–2718 (1988).
8. Ershov, S. V. & Potapov, A. B. *Phys. Lett. A* **167**, 60–64 (1992).
9. Politi, A., Livi, R., Oppo, G.-L. & Kapral, R. *Europhys. Lett.* **22**, 571–576 (1993).
10. Sinai, Y. G. *Int. J. Bifurcat. Chaos* **6**, 1137–1142 (1996).

Gaspard et al. *reply* — We presented experimental evidence for the existence of microscopic chaos in the mesoscopic motion of a brownian particle in solution. We used standard techniques to analyse long trajectories of a brownian particle and inferred a dynamical entropy from this analysis. We showed that this dynamical entropy can be accessed experimentally by measuring multiple-time correlation functions for the particle. The dynamical entropy was then used to provide a positive lower bound for the sum of the Lyapunov exponents for the underlying deterministic dynamical system composed of the fluid and brownian particles. We concluded that the positive dynamical entropy, obtained experimentally, was evidence for the existence of positive Lyapunov exponents in the underlying dynamics, and hence for the existence of microscopic chaos generated by a dynamical instability.

Dettmann *et al.* and Grassberger and Schreiber argue that there may be other explanations. There is no doubt that a positive dynamical entropy could be generated by mechanisms other than a local intrinsic, dynamical instability. Illustrative models include the Rayleigh flight of a tracer particle in a non-interacting ideal gas, the motion of an impurity in a harmonic crystal, and the motion of the wind particle in the wind-tree model. In these models, an external pseudorandom generator must be involved at some stage of the simulation of the dynamics, leading to a positive dynamical entropy without a local dynamical instability.

In the Rayleigh flight and the harmonic crystal, randomness is generated repetitively by the new particles or waves continuously coming from infinity. In the wind-tree model, the scatterers are randomly located by using a pseudorandom generator before the simulation of the time evolution. As the wind particle collides with a new scatterer, it picks up new information on the location of this scatterer and the information recorded on this trajectory grows as a result. However, if the square scatterers formed a regular lattice, this growth would stop and the dynamical entropy would vanish. In contrast, for circular disks, as in the Lorentz gas, the dynamical entropy is always positive whether the lattice is regular or not. The apparent dynamical randomness of the wind-tree model therefore has its origin in the structural disorder of the model.

The models mentioned by Dettmann *et al.* and Grassberger and Schreiber fail to capture an essential feature of brownian motion: that the diffusion coefficient is inversely proportional to the viscosity of the fluid surrounding the brownian particle. The viscosity of the fluid is absent in models in which the surrounding particles either do not move or have no interaction or only harmonic interactions. These models are therefore not plausible for the interpretation of our experiment[1].

However, if the interactions between surrounding particles are generic and nonlinear, the viscosity can be positive and the dependence of the diffusion coefficient on the viscosity can be explained. In such systems with nonlinearly interacting particles, numerical and analytical studies have shown that intrinsic local instability is the dominant mechanism for generating dynamical randomness, and no external pseudorandom generator is needed[2,3]. Standard models of brownian motion are of this kind and develop chaotic dynamics with a full spectrum of positive Lyapunov exponents.

For these reasons, we believe that our explanation in terms of a randomness self-generated by the dynamical instability is the most plausible one for this experiment. We hope that further experiments will be able to distinguish more directly between different regimes of random behaviour in systems with a large number of particles, and so provide further evidence of microscopic chaos.

P. Gaspard*, M. E. Briggs†, M. K. Francis‡, J. V. Sengers‡, R. W. Gammon‡, J. R. Dorfman‡, R. V. Calabrese‡
**Faculté des Sciences, Université Libre de Bruxelles, Boulevard du Triomphe, 1050 Brussels, Belgium*
†University of Utah, Salt Lake City, Utah 84112, USA
‡University of Maryland, College Park, Maryland 20742, USA

1. Gaspard, P. *et al. Nature* **394**, 865–868 (1998).
2. Posch, H. A. & Hoover, W. G. *Phys. Rev. A* **38**, 473–482 (1988).
3. van Beijeren, H., Dorfman, J. R., Posch, H. A. & Dellago, C. *Phys. Rev. E* **56**, 5272–5277 (1997).