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A Lévy flight for light

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A random walk is a stochastic process in which particles or waves travel along random trajectories. The first application of a random walk was in the description of particle motion in a fluid (brownian motion); now it is a central concept in statistical physics, describing transport phenomena such as heat, sound and light diffusion[1]. Lévy flights are a particular class of generalized random walk in which the step lengths during the walk are described by a 'heavy-tailed' probability distribution. They can describe all stochastic processes that are scale invariant[2,3]. Lévy flights have accordingly turned out to be applicable to a diverse range of fields, describing animal foraging patterns[4], the distribution of human travel[5] and even some aspects of earthquake behaviour[6]. Transport based on Lévy flights has been extensively studied numerically[7–9], but experimental work has been limited[10,11] and, to date, it has not seemed possible to observe and study Lévy transport in actual materials. For example, experimental work on heat, sound, and light diffusion is generally limited to normal, brownian, diffusion. Here we show that it is possible to engineer an optical material in which light waves perform a Lévy flight. The key parameters that determine the transport behaviour can be easily tuned, making this an ideal experimental system in which to study Lévy flights in a controlled way. The development of a material in which the diffusive transport of light is governed by Lévy statistics might even permit the development of new optical functionalities that go beyond normal light diffusion.

In recent years, light has become a tool widely used to study transport phenomena. Various analogies between electron, light and matter-wave transport have been discovered, including weak and strong localization[12], the Hall effect[13], Bloch oscillations[14] and universal conductance fluctuations[15]. Understanding light in disordered systems is of primary importance for applications in medical imaging (for example tumour diagnostics)[16], random lasing[17] and image reconstruction[18]. Most of these studies have been limited to the simplified case in which the light performs a random walk that can be described as a diffusion process.

In a Lévy flight, the steps of the random walk process have a power-law distribution, meaning that extremely long jumps can occur[2,19,20] (Fig. 1). Consequently, the average step length diverges and the diffusion approximation breaks down for Lévy flights. Power-law distributions often appear in other physical phenomena that exhibit very large fluctuations, for instance the evolution of the stock market[21,22] and the spectral fluctuations in random lasers[23,24].

A random walk in which the step length is governed by Lévy statistics leads to superdiffusion; that is, the average squared displacement $\langle x^2 \rangle$ increases faster than linearly with time t

$$\langle x^2 \rangle = Dt^{\gamma}$$

where γ is a parameter that characterizes the superdiffusion and D is a generalized diffusion constant. For $\gamma > 1$ we have superdiffusion, whereas for $\gamma = 1$ we recover normal diffusive behaviour. Normal diffusions are therefore limiting cases of Lévy flights. In Lévy flights, superdiffusion is purely a result of the long-tailed step-length distribution. Random walks in which the step time (and thus a finite velocity) is also important are called Lévy walks[19]. A long-tailed distribution in the scattering dwell time can give rise to, for example, subdiffusion[25] ($\gamma < 1$). There is no practical difference between a Lévy walk and a Lévy flight in the experiments described in this paper, because all of the experiments are static (time independent).

We report here on the creation of an optical material in which the step-length distribution can be specifically chosen. We use this to produce a structure in which light performs a Lévy flight. In a set of experiments, we show that the optical transport in such a material is superdiffusive. To produce such a structure requires an approach that initially seems counter-intuitive. The material that we have obtained is, however, relatively easy to make and provides the first well-controlled experimental test ground for Lévy transport processes. We propose the name Lévy glass for this material.

To obtain an optical Lévy flight it might seem best to develop scattering materials with self-similar (fractal) structures. This approach turns out not to work in practice, owing to the dependence of the scattering cross-section on size. In, for instance, a fractal colloidal suspension, the larger particles would be subject to resonant (Mie) scattering, whereas the smaller particles would hardly scatter at all (Rayleigh limit). The solution is to find a way to modify the density of scatterers instead of their size. This makes it possible to obtain a scattering mean free path that depends strongly on the position inside the sample.

We have found a relatively easy, but so far unstudied, method of doing this, using high-refractive-index scattering particles (of titanium dioxide in our case) in a glass matrix. The local density of scattering particles is modified by including glass microspheres of a particular, highly non-trivial size distribution. These glass microspheres do not scatter, because they are incorporated into a glass host with the same refractive index. Their sole purpose is locally to modify the density of scattering elements.

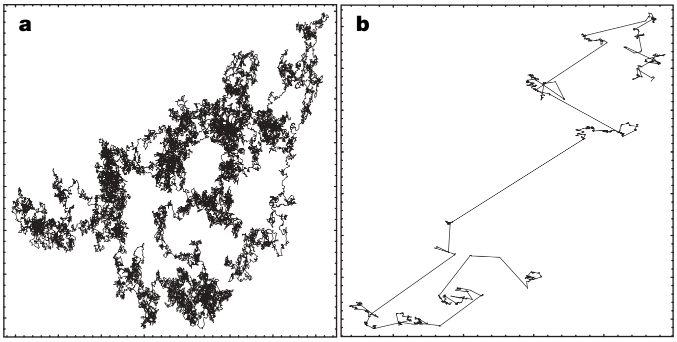

Figure 1 | Random walk trajectories. **a**, Normal diffusive random walk; **b**, Lévy random walk with $\gamma = 2$ (Lévy flight). In the normal diffusive random walk, each step contributes equally to the average transport properties. In the Lévy flight, long steps are more frequent and make the dominant contribution to the transport.

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The random walk in normal diffusive materials has a gaussian step-length distribution with average step length given by the mean free path ℓ

$$\ell = \frac{1}{\langle N\sigma \rangle} \tag{1}$$

where σ is the scattering cross-section and N is the density of scattering elements. The angle brackets indicate an average over the sample volume. To permit Lévy flights, the material should give rise to a step-length distribution with a heavy tail, decaying as[26]

$$P(z) \rightarrow \frac{1}{z^{\alpha+1}}$$

where $P(z)$ is the probability of a step of length z and α is a parameter that determines the type of Lévy flight. The parameter α can be shown to be related to the superdiffusion exponent γ by $\gamma = 3 - \alpha$, for $1 \leq \alpha < 2$ (ref. 7). The moments of this distribution diverge for $\alpha < 2$, which means that the average in equation (1) can no longer be taken over the entire sample. However, $N\sigma$ can still be interpreted as the local scattering strength of the material.

Our samples were made by suspending titanium dioxide nanoparticles in sodium silicate, together with a precisely chosen distribution $P_s(d)$ of glass microspheres of different diameters d. The total concentration of titanium dioxide nanoparticles was chosen such that, on average, about one scattering event takes place in the titanium-dioxide-filled spaces between adjacent glass microspheres. The step-length distribution is then determined by the density variations induced by the distribution $P_s(d)$ of the glass microspheres. We have calculated that a diameter distribution $P_s(d) = 1/d^{2+\alpha}$ is required to obtain a Lévy flight with parameter α, and show this experimentally below. Although with our method we can obtain a Lévy flight with any value of α, we have chosen to work with $\alpha = 1$, because this is one of the few cases in which the Lévy distribution has a simple analytical expression (namely that of the Cauchy distribution[27]). For all other details on sample preparation and the derivation of the diameter distribution for Lévy flights with parameter α, see Supplementary Information.

We made a series of samples of different thicknesses in the range 30–550 μm. This allowed us to record the thickness dependence of the total transmission. To do so, a collimated He–Ne laser beam was used incident on the sample on a spot of area 1 mm^2. The total transmitted light was then collected by means of an integrating sphere. Total transmission in normal diffusive systems is known to decay following Ohm's law, which means that the transmission depends linearly on the inverse sample thickness[12]. For superdiffusion this can be generalized as follows, where A is a constant and L is the thickness[28]:

$$T = \frac{1}{1 + AL^{\alpha/2}}$$

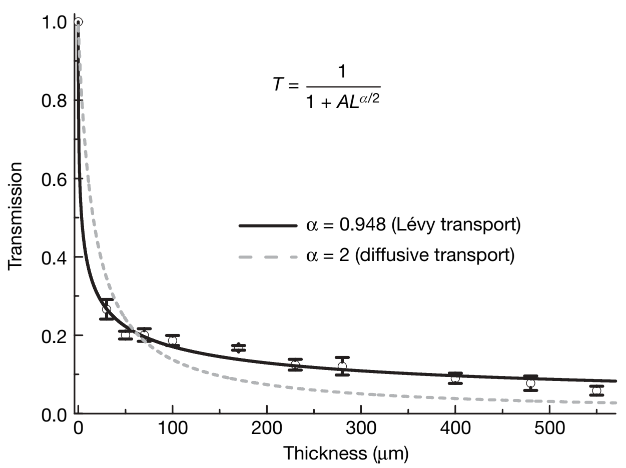

Figure 2 | Thickness dependence of the total transmission. For superdiffusion the transmission decays much more slowly than for normal diffusion, and should follow a power law with exponent $\alpha/2$. The dashed grey curve shows the normal diffusive behaviour ($\alpha = 2$), whereas the black line is a fit to the data with only α as free parameter. We obtain $\alpha = 0.948 \pm 0.09$, which is very close to the expected value, $\alpha = 1$, for a lorentzian Lévy flight. For very thick samples (550 μm), optical absorption decreases the transmission to slightly below the ideal curve. Error bars, s.d.

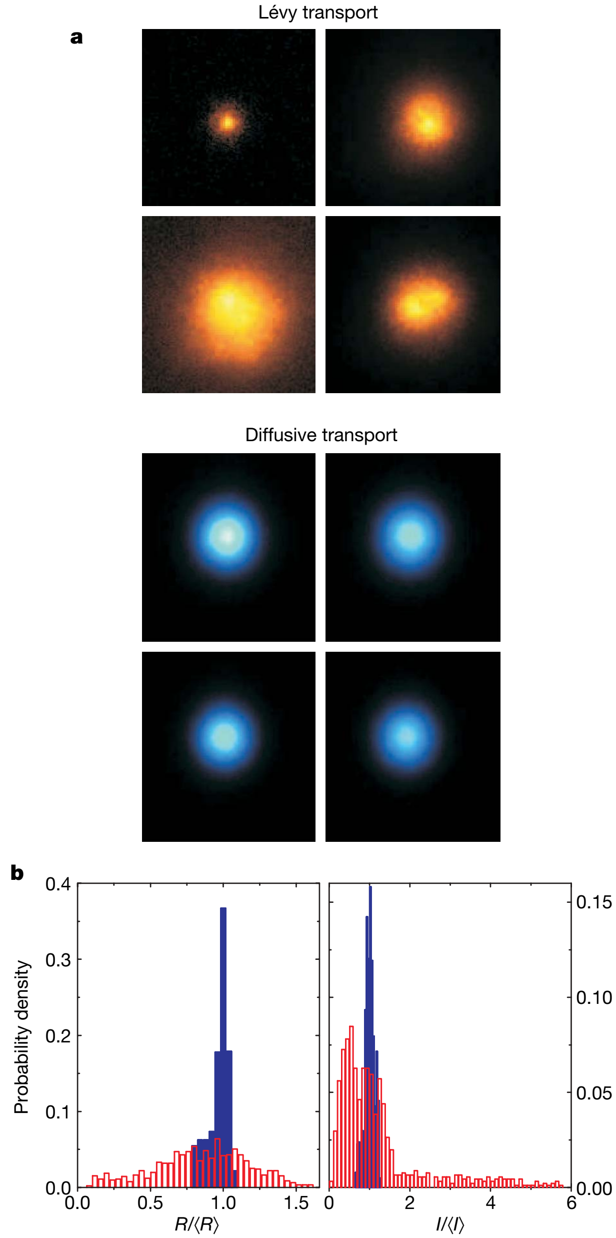

Figure 3 | Spatial dependence of the transmission on the output surface. **a**, Spatial distributions of the transmitted intensity for the Lévy samples (top) and for normal diffusive samples of the same thickness (bottom). The images were taken using a Peltier-cooled charged-coupled-device camera on the output surface of the sample, which was illuminated from the front with a focused (2μm-spot-size) He–Ne laser. The sample was placed between crossed polarizers to make sure that any residual ballistic light was blocked. The normal diffusive sample was made by using only sodium silicate and titanium dioxide powder. In the Lévy case we can see that the transmission profiles strongly fluctuate from one measurement to another, whereas in the normal diffusive case they are nearly constant. **b**, Distributions of the radius R (normalised to its average, $\langle R \rangle$)and total intensity I (normalised to its average, $\langle I \rangle$) of the transmission profiles for the normal diffusive (blue) and Lévy (red) samples. We can see that the very large fluctuations in the Lévy case correspond to a broad distribution function of both the intensity and radius of the transmission profile.

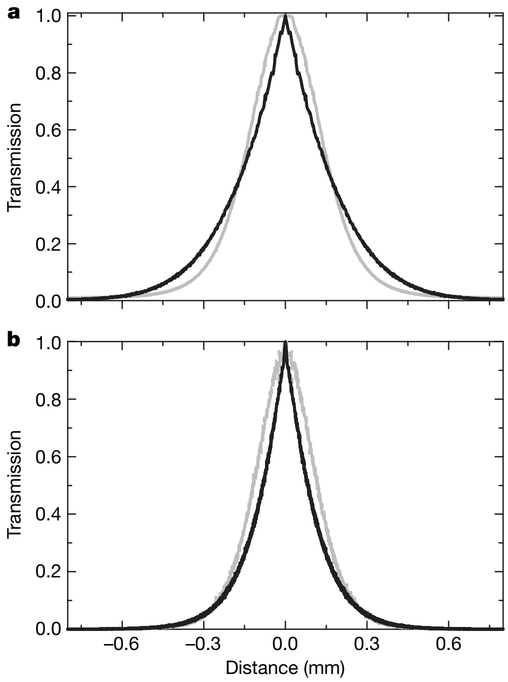

Figure 4 | Average transmission on the output surface versus radial distance from the centre. **a**, Experimental data. In the Lévy case (black) an average over 3,000 sample configurations was needed to obtain the average behaviour. The profile of the Lévy sample shows a pronounced cusp, and slowly decaying tails. The normal diffusive sample (grey) has a profile close to a gaussian lineshape: the top is rounded and long tails are absent. **b**, Result of Monte Carlo simulations of a normal diffusive random walk (grey) and a Lévy random walk (black) in a slab. The superdiffusive profile again displays a sharp cusp and decays more slowly than does the normal diffusive profile. The difference in absolute widths between experiment and simulation is due to internal reflections at the boundary of the sample, which were not taken in account in the simulations.

For the normal diffusive case in which $\alpha = 2$, we recover Ohm's law of conductance. The experimental data are shown in Fig. 2. We can see that they decay much more slowly than linearly, showing that transport in these samples is superdiffusive. In this case $\alpha = 0.948 \pm 0.09$. This result is in excellent agreement with the expected value for a lorentzian Lévy flight, without the use of any additional fit parameters.

The power-law step-length distribution of a Lévy flight is expected to give rise to strong fluctuations in the transport properties of individual samples. In the total transmission profile we should therefore observe large differences between disorder realizations. In comparison, a normal diffusive sample would show almost no fluctuations. In Fig. 3a, we present the intensity profiles taken from the output (rear) surface of a sample that is illuminated from the front with a focused He–Ne laser. Successive images were taken by moving the sample over distances much larger than the illuminated region.

We compared the behaviour of a Lévy glass with that of a normal diffusive system of the same thickness. From the Lévy glass we observed very large differences between disorder realizations, whereas the result for the normal diffusive system is nearly constant. To quantify this behaviour we calculated the distributions of the radius and the intensity of these profiles on a set of 900 disorder realizations (Fig. 3b). In the Lévy case the distributions are extremely broad, but in the normal diffusive case they are very narrow. Moreover, in the Lévy case the distributions have slowly decaying tails, which are absent in the normal diffusive case.

The characteristics of the Lévy flight also survive if we perform an average over a large number of observations. The resulting profiles of the transmitted intensity on the output surface are plotted in Fig. 4 and compared with the results of Monte Carlo simulations. Both the experimental and the simulation results show the same features. For the normal diffusive system we observe that the profile has, as expected, a bell-shaped profile, which is very close to a gaussian curve. For the Lévy sample, however, the profile exhibits a well-defined cusp and has tails that decay much more slowly than in the normal diffusive case. The agreement between the experimental and simulated profiles is very good. The small discrepancy in the overall width of the profile can be explained by the influence of internal reflections at the boundary of the sample, which were not taken into account in the Monte Carlo simulations. We have verified that in a sample made of titanium dioxide nanoparticles and just one family of (large) glass microspheres, the profile remains gaussian (Supplementary Information). This confirms that the density variations induced by the entire size distribution of glass microspheres are required to obtain a Lévy flight.

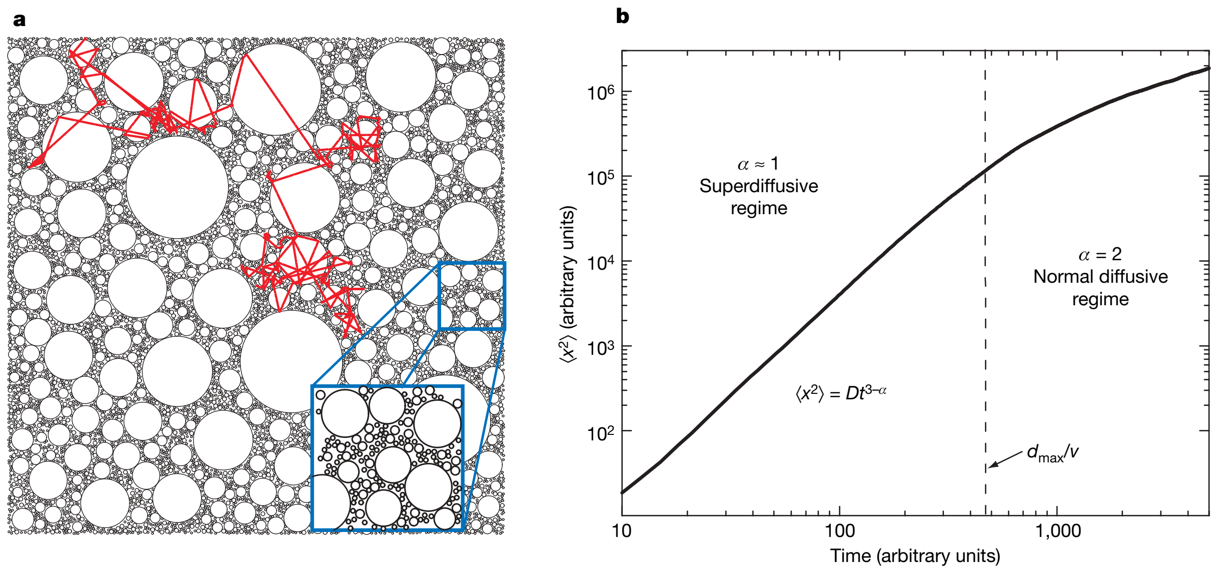

Figure 5 | Lévy walk in an inhomogeneous medium. **a**, Random walker trajectory, obtained by Monte Carlo simulation. Owing to the strong density fluctuations, the scattering material permits Lévy flights (red). Inset, magnification showing the scale invariance of the material's structure. **b**, Average squared displacement. The spreading is superdiffusive, with $\alpha = 1$. Because the sample is of finite size, the Lévy walk is truncated at $t = d_{max}/v$, where d_{max} is the maximum step length, determined by the sample thickness.

Real physical samples are intrinsically of finite size, which means that the largest step size of the Lévy flight is limited by the sample size. This introduces a cutoff in the step-length distribution and results in a so-called truncated Lévy flight. On length scales greater than this cutoff, the transport is expected to recover normal diffusive behaviour[29]. We investigated this by running a series of Monte Carlo simulations in which we studied a random walk in a two-dimensional system similar to our samples, namely a scattering medium where disk-shaped regions are introduced without scattering elements. The diameter distribution of these two-dimensional disks was chosen, following the same reasoning as above, as $P_s(d) = 1/d^2$. We simulated the evolution with time of the averaged squared displacement of light propagating in this system. The results of these simulations (Fig. 5) show superdiffusive behaviour that, on a very long timescale, develops into normal diffusive behaviour. The parameter γ of the superdiffusive expansion was found to be close to two, as expected for a lorentzian Lévy flight. The timescale of the transition from superdiffusive to diffusive transport is given by the time necessary to probe all possible step lengths: $t_{\mathrm{trans}} = d_{\mathrm{max}}/v$, where d_{max} is the greatest step length and v is the velocity of the random walker. In our samples, the thickness was equal to this cutoff length (greatest sphere diameter). As a result, the effect of the cutoff can be expected to be negligible within the signal-to-noise ratio of our experiment.

We have shown that it is possible to make disordered optical materials with controllable step-length distributions. In particular, we have made superdiffusive optical materials permitting optical Lévy flights. The physics of light transport is closely related to the transport of electrons and matter waves, and important analogies like the optical Hall effect, weak and strong localization of light, and correlations in laser speckle have been identified in recent years. The question of how these phenomena are manifest in Lévy glass is still completely open. The procedure that we have used to synthesize Lévy glass is reproducible and can be implemented on a large (industrial) scale. Our techniques could be used in the development of new opaque optical materials, such as paints with particular visual effects and lasers based on superdiffusive feedback.

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Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

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