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Lévy flights of photons in hot atomic vapours

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Gaussian distributions are often used to study properties of random and fluctuating systems. However, in a number of situations, rare events have drastic consequences, which can not be explained by Gaussian statistics. Considerable efforts have thus been devoted to the study of non Gaussian fluctuations such as Lévy statistics, generalizing the standard description of random walks. Unfortunately only macroscopic signatures, obtained by averaging over many random steps, are usually observed in physical systems. We present experimental results investigating the elementary process of anomalous diffusion of photons in hot atomic vapours. We measure the step size distribution of the random walk and shows that it follows a power law characteristic of Lévy flights.

Systems that exhibit power law behavior often present counterintuitive features on macroscopic scales. In probability theory, power laws correspond to situations where rare events can have drastic consequences and relevant average values, defined by the moments of the distribution, can even be infinite. The broad range of applications of power law distributions includes biology, economics, finance, catastrophe management and resonance fluorescence in astrophysical systems and atomic vapours [1, 2, 3, 4]. Large (non Gaussian) fluctuations also play a fundamental role in many physical situations, in particular around phase transitions, triggering considerable efforts to understand universal features of such phenomena [5, 6]. One example is Anderson localization, a disorder-induced phase transition of non interacting waves in three-dimensional disordered media [7, 8]. In this case, around the localization threshold, a non diffusive behavior of the random walk appears in a multiple scattering regime. The experimental characterization of this peculiar behavior is elusive, because observations correspond to macroscopic quantities, obtained by averaging over many scattering events [9, 10, 11, 12]. As for theoretical descriptions, even though large fluctuations are expected close to the threshold [13], they often consider a mean free path and a diffusion coefficient. The assumption that these average quantities can still be defined and are finite is thus often implicitly made.

Even far from any phase transition, the existence of a mean free path and a diffusion coefficient can in some cases be questioned. For instance, radiation trapping in hot atomic vapours can indeed exhibit anomalous random walk. Because this phenomenon occurs in many different systems, ranging from stars [14] to dense atomic vapours [15] such as gas lasers, discharges and hot plasmas, this field has been subject to intense studies for many decades, including seminal work by Holstein [16]. It has been realized very early [17] that frequency redistribution has a profound impact on the multiple scat-

tering features of photons. Whereas elastic scattering, which occurs in laser-cooled dilute atomic vapours [18, 19, 20], leads to normal diffusion with well defined scattering mean free path and diffusion coefficient, inelastic scattering as in hot vapours can lead to situations where the central limit theorem no longer applies. Gaussian statistics is then no longer the appropriate tool and various moments of the particle distribution can become infinite. The essential microscopic ingredient to describe the random walk of light in large samples is the photon step size distribution $P(x)$. If this distribution has an asymptotic power law dependence ($P(x) \sim 1/x^\alpha$), then the average step size ℓ (mean free path) or the diffusion coefficient D depends critically on the value of α . For $\alpha < 3$, the variance σ_x^2 of the step size distribution diverges and one can no longer define a usual diffusion coefficient. In this case, rare but large steps can dominate the average spreading of the photon distribution. According to the generalized central limit theorem [21], photon trajectories in this case are Lévy flights. Unfortunately, in most systems where multiple scattering of photons occurs, it is difficult to have direct experimental access to the single step size distribution at the origin of this random walk and anomalous diffusion is usually inferred from macroscopic observations [22].

In this Article we present experimental results investigating the microscopic ingredient leading to a regime of superdiffusion in multiple scattering of light in hot vapours of rubidium atoms. We have used a specific geometrical arrangement to isolate a single step in the multiple scattering sequence. We measure the single step size distribution $P(x)$, which follows a power law $P(x) \propto 1/x^\alpha$, with $\alpha < 3$. Therefore the photon trajectories are Lévy flights, with an infinite variance of $P(x)$.

The random walk of photons in atomic vapours is usually characterized by the various moments of the step size distribution $P(x)$. One can define a mean free path by $\ell = \langle x \rangle = \int_0^\infty xP(x)dx$ and a diffusion coefficient from the variance σ_x^2 of the distribution. For photons at frequency ω , this step size distribution $P(x, \omega)$ can be obtained from $P(x, \omega) = -\frac{\partial T(x, \omega)}{\partial x}$, where T is the forward transmission given by the Beer-Lambert's

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law $T(x, \omega) = e^{-x/\ell(\omega)}$, with the frequency dependant mean free path $\ell(\omega)$. While these considerations give a good description for atoms close to zero temperature [18, 19, 20], most samples in and outside laboratories present a Doppler broadening $\Delta\omega_D$ much larger than the natural linewidth Γ of the optical transition. The normalized spectrum $\Theta(\omega)$ of the light in the multiple scattering regime then influences the properties of its random walk [17], and the step size distribution $P(x)$ is obtained by

$$P(x) = \int_0^{+\infty} \Theta(\omega) P(x, \omega) d\omega. \quad (1)$$

An analytical expression of the single step size distribution of photons in an atomic vapour can be obtained in some limits. Assuming that the emission and absorption spectra are purely Gaussian (Doppler broadening), then the single step size distribution asymptotically follows [16, 23]:

$$P(x) \sim \frac{1}{x^2 \sqrt{\ln(x/\ell_0)}}, \quad (2)$$

where $\ell_0^{-1} = n_{\text{at}}\sigma_0$ with n_{at} the atomic density and σ_0 the scattering cross section at the atomic resonance. This heavy tail distribution leads to Lévy flights of photons.

To measure this step size distribution, we have used a specific multicell arrangement as shown in Fig. 1. We image on a cooled charge coupled camera (CCD) the fluorescence of a natural isotopic mixture of rubidium atoms in a long cylindrical observation cell, illuminated along its axis. We thus measure the probability of a photon to be scattered after a distance x along the cell axis, *i.e.* the step size distribution $P(x)$.

As a reference, we first measure $P(x)$ in the case when the observation cell is illuminated by a monochromatic incident laser, locked to the $F = 3 \rightarrow F' = 4$ transition of the D2 line of rubidium 85. From the corresponding image [Fig. 2(a)] we extract an exponential step size distribution as expected from Beer-Lambert's law [Fig. 3]. This preliminary measurement allows the calibration of the atomic density and of the mean free path for resonant photons, which can be varied by adjusting the temperature of the observation cell from 20°C to about 47°C. The atomic density thus varies from 9×10^{15} to $2 \times 10^{17} \text{ m}^{-3}$, and the mean free path changes accordingly from 50 mm to 5 mm.

To measure Lévy flights of photons we use a double cell configuration [Fig. 1]. Our 11 mW, 2 mm-waist laser beam is incident on a first, small cylindrical cell of rubidium of optical thickness 0.2, where photons undergo at most one scattering event with a well defined position. A 2 mm-diameter pencil of diffused light propagating in a direction orthogonal to the initial laser beam is then selected by two 12 cm-spaced diaphragms. This scheme produces photons with a frequency spectrum $\Theta(\omega)$ given by the Doppler broadening. If we neglect the finite but small width of the atomic transition, the absorption spectrum is also purely Doppler, and Eq. (2) holds. The scattered light then goes through the 7 cm-long observation

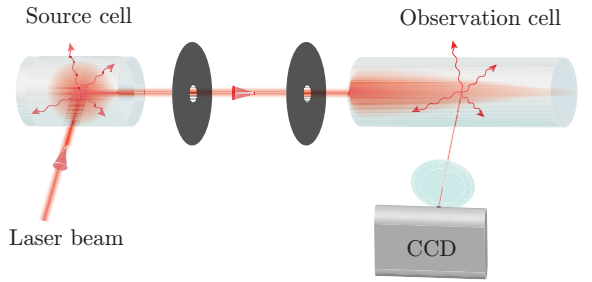


FIG. 1: Scheme of the experimental setup. A laser beam is incident on a so-called source cell filled with rubidium vapour. Scattered light propagating at orthogonal direction is selected with two diaphragms and illuminates a second, observation cell. The light scattered in this second cell is imaged on a cooled CCD camera. This fluorescence signal is proportional to the step size distribution function.

cell, with an angle of about 10° from the cell axis to avoid stray reflections at the center of the image. Raw images of the fluorescence signal are obtained after a 30 minutes exposure. Reproducible noise is then eliminated by subtracting a dark frame. The resulting image is shown in Fig. 2(b). We extract the corresponding step size distribution $P(x)$ (shown on a log-log scale in Fig. 3) by taking longitudinal slices along the incident direction of propagation. This signal is integrated over 30 lines of the CCD matrix (corresponding to 1.6 mm in the cell), then smoothed over 30 pixels along the x direction to increase the signal to noise ratio. In order to obtain the correct $P(x)$ distribution, we need to correct the effect of multiple scattering on the signal. We thus subtract the intensity measured along a slice slightly off the line of sight of the diaphragms, which is due only to multiple scattering, from the intensity measured on the central slice. A Monte Carlo simulation, which can track the position of a photon emerging from the observation cell and register the number of scattering it has performed, confirmed that this procedure efficiently filters single scat-

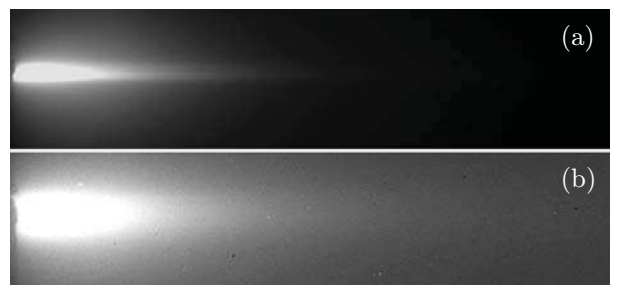


FIG. 2: Fluorescence images obtained after an exposure time of 30 minutes and dark frame subtraction, for (a) an incident laser beam at the atomic resonance frequency and (b) incident light provided by a first scattering cell. The temperature of the observation cell is 41°C. The step size distribution is extracted from the intensity along the axis.

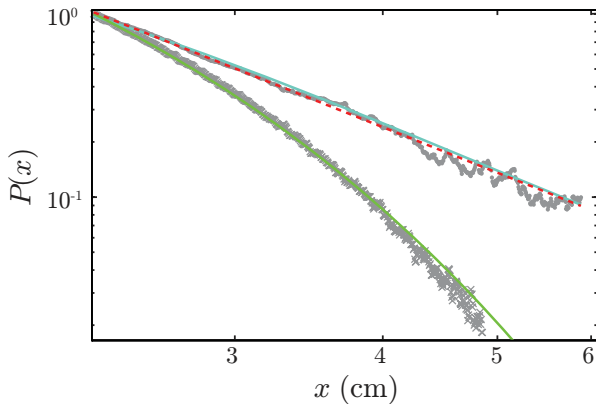


FIG. 3: Single step size distributions $P(x)$ plotted in log-log scale. For an incident monochromatic laser beam of frequency ω (crosses), $P(x)$ shows an exponential decrease, as shown by the green continuous fitting line. For an incident Doppler-broadened field originated from a first scattering cell (dots), $P(x)$ has a power law decrease, well fitted by $P(x) \sim \frac{1}{x^\alpha}$ with $\alpha = 2.41 \pm 0.12$ (red dashed line), characteristic of Lévy flights. A microscopic model, taking into account all hyperfine levels and Raman transitions, is shown as the blue continuous line. Without any free parameter except the vertical intensity scale, the agreement with the experimental data is very satisfactory.

tering events in the observation cell. We clearly identify a power law, which can be fitted by:

$$P(x) \sim \frac{1}{x^\alpha}, \quad \text{with } \alpha = 2.41 \pm 0.12. \quad (3)$$

This decay is in clear contrast to the exponential decay for the monochromatic incident field. Note that within our range of parameters a fit with $1/(x^\alpha \sqrt{\ln(x/\ell_0)})$ is not noticeably different from the pure power law fit of Eq. (3). We have also checked that varying the atomic density in the observation cell from 7×10^{16} to $2 \times 10^{17} \text{ m}^{-3}$ has no impact on the measured value of α .

This result calls for two important comments. First, the exponent α is smaller than 3. We can thus not ignore the heavy tails of the step size distribution. The variance of $P(x)$ is infinite, making this distribution characteristic of Lévy flights. Second, we measure an important difference compared to the prediction $\alpha = 2$ expected when the natural width Γ of the atomic transition is neglected [Eq. (2)]. Even though this width $\Gamma/2\pi = 6 \text{ MHz}$ is much smaller than the Doppler broadening $\Delta\omega_D/2\pi \sim 220 \text{ MHz}$, one cannot neglect the effect of the natural Lorentzian line shape on the atomic absorption profile. In the opposite limit of negligible Doppler broadening ($\Delta\omega_D \ll \Gamma$) one expects to recover the cold-atom limit, where all momenta of the step size distribution are finite. It is thus not surprising that the actual case of a finite natural width does increase the absolute value of the exponent from the ideal Doppler limit, where $\alpha = 2$. A power law fit to the step size distribution $P(x)$ obtained by including this finite natural width

in a numerical integration [Eq. (1)] yields an exponent of 2.3, close to the experimental value. We have also implemented a more refined model, taking into account all hyperfine levels of rubidium atoms including the hyperfine Raman transitions. The result of this numerical calculation is in excellent agreement with the experimental result as shown in Fig. 3. Although the finite natural line width is only a few percents of the Doppler width, it surprisingly changes the power law exponent by 15%. The measured difference to $\alpha = 2$ has a particular significance, as for $\alpha \leq 2$, even the mean free path is no longer defined, in contrast to $\alpha > 2$ where $\langle x \rangle$ is still finite.

The knowledge of the single step size distribution for photons which have been scattered only once by hot atoms is however not sufficient to describe the multiple scattering regime. Indeed, the spectral characteristics of the photons depend on their previous history. For instance, at angles close to forward scattering, the Doppler broadening is very small and the scattering can be considered to be almost elastic. This feature is described by a partial frequency redistribution of the photons during their multiple scattering process [15]. In order to have experimental access to the step size distribution (and therefore to its exponent in case of power laws) in a multiple scattering regime, it is important to extract the single step size distribution of a photon that has been scattered several times by hot atoms.

To determine numerically the shape of the step size distribution P_n for photons after n scattering events, we compute the evolution of the spectrum Θ_n of scattered light, taking into account Doppler broadening and averaging over the scattering angles [24]. P_n is then obtained by using Eq. (1) with Θ_n . The numerics show that P_n quickly converges towards a power law, which is independent of the frequency of the initial photon, and hence, that the step size distribution of photons in the multiple scattering regime is well defined. The corresponding power law exponent $\alpha(n)$ is reported on Fig. 4.

In order to approach experimentally the situation of multiple scattering, we have implemented a triple cell geometry. In a first cell, with an on-resonance optical thickness on the order of 2, we prepare photons which have been scattered several times ($n_{sc} \sim 4$) when they leave the cell. Those photons have thus no memory of their initial direction and their properties are well described by an angular average of the fluorescence spectrum. These photons are then sent towards the “source cell” of Fig. 1, which still has a low optical thickness. Photons thus undergo one more scattering event with a well defined position before they can arrive onto the observation cell. This experimental protocol allows us to produce photons which have done multiple scattering and forgotten their initial direction and frequency, as required for a steady state situation. We then record the image of the fluorescence on the CCD. The signal in this geometry is much weaker than in the previous double cell geometry and is limited by photon shot noise and cosmic rays. From the median averaging of 6 images corresponding to an ex-

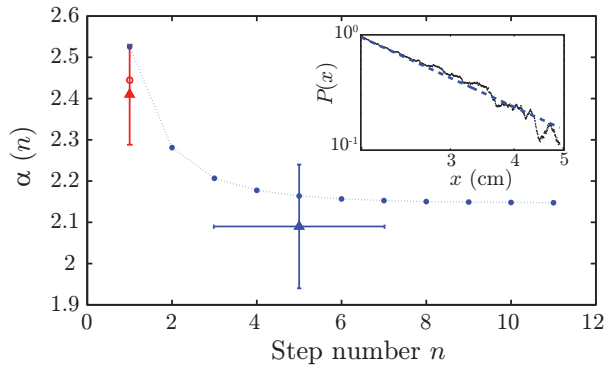


FIG. 4: Power law exponent α of the step size distribution as a function of the number n of scattering events (steps). The double cell configuration ($n = 1$) [Fig. 1] yields at 90° a pure Doppler-broadened spectrum and the measured α (red triangle) is $\alpha = 2.41 \pm 0.12$. For the triple cell configuration, photons are scattered about 5 times (in average) before entering the observation cell. The measured exponent is then $\alpha = 2.09 \pm 0.15$ (blue triangle). The data and fit for this last configuration are shown in the inset. Vertical error bars represent the uncertainty of the power law fit. The blue dots are numerically computed by taking into account all the hyperfine levels of the rubidium atoms and an angular average of the Doppler-broadened frequency spectrum. The open red circle is computed without angular average, so that it can be compared with the corresponding experimental data.

posure time of 5 hours each, we are still able to extract the step size distribution $P(x)$ after multiple scattering and obtain a value $\alpha = 2.09 \pm 0.15$ [Fig. 4]. This value, clearly below 3, excludes a diffusion approach to be used for multiple scattering of photons in hot atomic vapours.

This experimental result is in good agreement with our numerical estimation of the power law, taking into account all details of the atomic transition [Fig. 4].

We have shown by direct experimental observation of the elementary ingredient of the random walk, that multiple scattering of photons in hot atomic vapours are Lévy flights. Corrections due to the finite width of the excited state vanish for large scattering orders and the power law approaches the one expected from a pure-Doppler broadened system.

As all the experiments described in this Article correspond to steady state situations (with continuous laser sources) no velocity consideration is required to describe the experimental results. In a pulsed experiment however one might be able to address questions related to the time needed to cover a distance x . In resonant systems such as two level atoms, a trapping time on the scatterers can be defined, which has to be combined to the propagation time [19] and one might have to distinguish Lévy flights from Lévy walks [25]. Another extension of this work is the possibility of including further broadening mechanisms, such as pressure broadening or inelastic scattering at large intensities. Even if the absorption cross sections then have Lorentzian wings, as in the natural broadening case, the emission spectra are very different as one no longer has coherent emission processes, and one expects in that case that $P(x)$ follows a power law with an exponent 1.5 leading to an infinite mean free path [23]. Finally, truncated Lévy flights [26] could be considered to deal with finite-size samples or, on the other hand, with very large systems, if the step size distribution exhibits a cutoff at long distance [24].

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