Photon Trajectories in Incoherent Atomic Radiation Trapping as Lévy Flights

Eduardo Pereira*
Universidade do Minho, Escola de Ciências, Departamento de Física, 4710-057 Braga, Portugal

José M. G. Martinho and Mário N. Berberan-Santos
Centro de Química-Física Molecular, Instituto Superior Técnico, 1049-001 Lisboa, Portugal
(Received 19 November 2003; published 13 September 2004)

Photon trajectories in incoherent radiation trapping for Doppler, Lorentz, and Voigt line shapes under complete frequency redistribution are shown to be Lévy flights. The jump length (r) distributions display characteristic long tails. For the Lorentz line shape, the asymptotic form is a strict power law $r^{-3/2}$, while for Doppler the asymptotic is $r^{-2}(\ln r)^{-1/2}$. For the Voigt profile, the asymptotic form always has a Lorentz character, but the trajectory is a self-affine fractal with two characteristic Hausdorff scaling exponents.

DOI: 10.1103/PhysRevLett.93.120201 PACS numbers: 02.50.Ey, 32.70.–n, 32.80.–t

Interest in distributions with long tails has increased over the last years, with intensive search for such laws in real physical systems. Most of the basic work on these distributions was carried out by Lévy in the 1930s [1], but it was only recently that these distributions were shown to be applicable to the description of a number of physical, biological, and social phenomena [2–5], including particle motion in turbulent media [2,3,6], anomalous diffusion in microheterogeneous systems [7], chaotic transport in laminar fluid flow [8], the albatross flight [9], and frequency fluctuations of chromophores isolated in glassy environments [10]. The purpose of this Letter is to show that photon trajectories in incoherent radiation trapping, a basic and common phenomenon in atomic and atmospheric physics, and in astrophysics, fit in the category of Lévy (superdiffusive) flights.

Although "the general trend nowadays is to put Lévy-type anomalous (super)diffusion on a similar footing with normal, Brownian-type, diffusion" [11], the actual physical systems invoked to justify the practical importance of Lévy flights in physical space tend to be somewhat marginal [3]. This contribution aims to prove that the well-known case of classical incoherent radiation trapping is one of the simplest and best characterized Lévy flights found up to now. In addition, radiation trapping has a major role in fluorescent lamps and this could make radiation trapping the economically most relevant concretization of a Lévy flight. In retrospect, the 1990 Ott *et al.* [7] contribution on the superdiffusion in microheterogeneous systems can no longer be considered the first experimental realization of a random flight with infinite moments, given that it was preceded by many studies of radiation trapping.

Radiation trapping of energy is important in areas as diverse as stellar atmospheres [12], plasmas and atomic vapors luminescence [13], terrestrial atmosphere and ocean optics, molecular luminescence [14], infrared radiative transfer, and cold atoms [4]. In these optically thick media, the emitted radiation suffers several reabsorption and reemission events before eventually escaping to the exterior; the radiation is said to be imprisoned or trapped. Atomic *radiation trapping* is also known as *imprisonment of resonance radiation*, *line transfer*, *radiation diffusion*, or *multiple scattering* of resonance radiation.

The first quantitative theory for atomic radiation trapping was presented in the 1920s by Compton and Milne, who developed a modified diffusion equation for the (frequency) coherent spreading of excitation. It was only in 1932 that the frequency redistribution between absorption and reemission was taken into account by Kenty [15]. He considered a Doppler spectral distribution and arrived at the unexpected result that, for an infinite medium, the diffusion coefficient would be infinite. This was the first realization of the fundamental fact that all moments of the jump size distribution are infinite. Kenty's result shows that a diffusion-type equation is not valid for radiation trapping with frequency redistribution effects. Nevertheless it was only in 1947 that Holstein and Biberman independently proposed a Boltzmann-type integro-differential equation [16], which remains the starting point of the vast majority of radiation trapping models [13].

Consider the case of *inelastic scattering* where, as the result of reabsorption/reemission events there is a photon frequency redistribution in the laboratory reference frame. The frequency distribution of the emitted photons is given by the emission spectrum $\Theta(x)$. The absorption probability of a photon with frequency x at a given distance from the emission point depends on the absorption spectrum $\Phi(x)$, and is given by $p(r|x) = \Phi(x)e^{-\Phi(x)r}$ (Beer-Lambert law) where r is the *opacity* or *optical density* and is a dimensionless distance. Radiation trapping can then be envisaged as a random flight in physical space with spectral shape dependent jump size distribution. The jump size distribution takes into account the absorption probability for all possible optical emission frequencies, hence

$$p(r) = \int_{-\infty}^{+\infty} \Theta(x) p(r|x) dx. \quad (1)$$

Equation (1) fully characterizes the spatial aspects of the random flight.

The moments of this distribution are

$$\langle r^n \rangle = n! \int_{-\infty}^{+\infty} \frac{\Theta(x)}{\Phi^n(x)} dx \quad (2)$$

and can be shown to be infinite for all physical reasonable atomic emission and absorption spectral distributions as concluded by Holstein [16]. His original analysis [16] only included *classical incoherent trapping*, in which the emitting state is statistically unrelated with the absorbing one and therefore there is *complete frequency redistribution* (CFR). In this case the absorption and emission spectra are identical and Eq. (1) reduces to

$$p(r) = \int_{-\infty}^{+\infty} \Phi^2(x) e^{-\Phi(x) r} dx. \quad (3)$$

Nevertheless, it can be concluded from Eq. (2) that the case of *partial frequency redistribution* is also characterized by an infinite moments Lévy statistic. The case of partial frequency redistribution is especially important in an astrophysical context. Neither case of complete coherent nor complete incoherent scattering is achieved exactly in stellar atmospheres, and it is then necessary to consider the photon redistribution and to calculate *redistribution functions* which will give $\Theta(x)$ [12]. In the usual laboratory conditions, vapor densities are high enough for CFR to apply [13]. This will be the case considered here. In two-level CFR atomic models both absorption and emission spectra can be described by Doppler, $\Phi_D(x) = \frac{1}{\sqrt{\pi}} e^{-x^2}$, Lorentz, $\Phi_L(x) = \frac{1}{\pi} \frac{1}{1+x^2}$, or Voigt, $\Phi_V(x) = a/\pi^{3/2} \int_{-\infty}^{+\infty} e^{-u^2}/[a^2 + (x-u)^2] du$, spectral distributions. x is a normalized difference to the center of line frequency and a is the Voigt characteristic width.

We now consider the asymptotic approximations valid for large jump sizes. A random flight in which the probability density of jump lengths is given by

$$p(r) \underset{r\to\infty}{\sim} \frac{1}{r^{(1+\mu)}} \quad (4)$$

with $\mu < 2$, is a self-similar random fractal with fractal dimension μ. It is called a Lévy flight after Mandelbrot [17], and defines a broad distribution for which all the moments of order not smaller than μ are divergent. If $\Phi(x)$ is substituted for $\Theta(x)$ in Eq. (2) above, it is found that $\langle r \rangle = \infty$, whatever the spectral line shape used, as long as $\Phi(x)$ is nonzero for large $|x|$. In this way, $\mu \leq 1$ for any $\Phi(x)$. In order to find the specific value of μ for the spectral distributions mentioned, we begin by rewriting Eq. (3) as

$$p(r) = -\frac{d^2 J(r)}{dr^2} \quad (5)$$

where

$$J(r) = \int_{-\infty}^{+\infty} (1 - e^{-\Phi(x) r}) dx. \quad (6)$$

When the line shape is Gaussian (Doppler), the integrand in Eq. (6) approaches a square wave form for large r, with inflection points at $-x_0$ and x_0 with $\Phi(x_0) r \simeq 1$. Hence, $x_0 = \sqrt{\ln r}$ for large r, $J(r) \simeq 2x_0 = 2\sqrt{\ln r}$, and therefore, from Eq. (5),

$$p(r) \underset{r\to\infty}{\sim} \frac{1}{r^2 (\ln r)^{1/2}}. \quad (7)$$

Assuming a homogeneous scaling law one arrives at an effective $\mu = 1 + 1/2\ln[\ln(r)]/\ln(r)$ which goes to 1 for $r \to \infty$: although the asymptotic of Eq. (4) is only approximately valid, one can nevertheless classify Doppler trapping as a strict Lévy flight with $\mu = 1$ with all the moments of the jump distribution being infinite.

When the line shape is Cauchy-like (Lorentz), the integrand in Eq. (6) can be simplified to $1 - \exp[-\Phi(x) r] \simeq 1 - \exp(-\frac{r}{\pi x^2})$ for large r and $J(r)$ becomes $2\sqrt{r}$. Therefore,

$$p(r) \underset{r\to\infty}{\sim} \frac{1}{r^{3/2}} \quad (8)$$

and the asymptotic distribution is a Lévy flight with $\mu = 1/2$.

The Voigt distribution is asymptotically coincident with the Lorentz distribution, and therefore has also $\mu = 1/2$ for any value of its parameter a. Equations (7) and (8) were already implied in Holsteins's 1947 contribution (original equations are, up to some minor terms, integrated versions of the equations in this Letter), a fact hitherto unnoticed in the literature.

Figure 1 shows the jump probability obtained from direct numerical integration of Eq. (3). A linear fit gives for the characteristic "tail index" (Hausdorff box count-

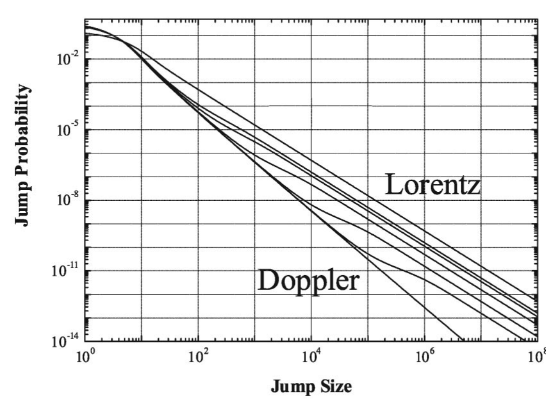

FIG. 1. Jump size distribution for CFR Doppler, Lorentz, and Voigt spectral profiles. From bottom to top: Doppler, Voigt with $a = 10^{-4}$, 10^{-3}, 0.01, 0.05, 0.1, and Lorentz.

ing), fractal dimension (μ) of the Doppler distribution $\mu = 1.07$ (jump sizes 10^3–10^4), and for the Lorentz case $\mu = 0.500$ (jump sizes 10^5–10^8), in agreement with previous assertions. Fig. 1 also shows that the continuous transformation from Doppler into Lorentz-like spectra as the Voigt a width parameter changes from zero into infinite values does not manifest itself in a continuous change of the effective, r dependent, μ values. There is an abrupt change instead at a jump distance which scales approximately as $1/a$. This is expected as the asymptotic expansion is related to the most extreme values of the wings of the spectral distribution and these change abruptly from Doppler to Lorentz-type asymptotics for a values as low as 10^{-4} (see Fig. 2). The data in Figs. 1 and 2 allow one to define the Voigt radiation trapping trajectories as a self-affine fractal with two different scaling exponents which manifest themselves at different length scales.

Consider now Fig. 3 which shows single excitation trajectories in a 3D infinite medium (compare Mandelbrot's well-known figure [17]). The Lorentz and Doppler cases display the two qualitative features characteristic of Lévy flights: (i) the longer path length jumps, although much less common, are of paramount importance to the overall spreading of excitation and, (ii) *self-similar behavior*. There is a hierarchy of clusters formed at different length scales but with similar topology. The set of visited points constitute a fractal of characteristic dimension μ for the Doppler and Lorentz distributions [2,3,6]. The Voigt case shows its *self-affine* nature. The trajectory topology changes from a Lorentz character into a Doppler one for smaller distances. The overall topology of the trajectory as a whole is of course dictated by the higher Lorentz scaling.

Up to now we have considered frequency redistribution at each scattering event. However, for high opacity two-level systems there are presumably many elastic scattering events before an inelastic scattering event occurs. It is therefore important to consider at least qualitatively the influence of *elastic scattering*. Elastic scattering events will fold up the excitation trajectories of Fig. 3 and the more folding, the higher the ratio of elastic to inelastic scattering probabilities. From this argument alone we will expect that an increase in the elastic to inelastic probabilities ratio will lead to an increase in the fractal dimension, as it will approach the 3D Brownian motion with fractal dimension 2.

The discussion thus far has been restricted to the case of *Lévy flights*. For the vapors studied in the laboratory the *time of flight* of in-transit radiation is negligible compared to the waiting time between absorption and reemission, thus rendering the Lévy flights formalism especially adequate. From the set of visited points one can obtain spatial distribution functions of the excitation density, which fully characterize the spatial aspects of radiation migration. The temporal evolution can be factorized from the spatial part (Lévy flight) and then separately handled [14,18]. However, for interstellar gases the time between scattering events might be large compared to the absorption/reemission times. Because the speed of light is finite, a *Lévy walk* modification of the flights here presented will be more appropriate in an astrophysical context [6].

The theoretical models for incoherent atomic radiation trapping are based either on the Holstein-Biberman multiexponential mode expansion or on the so-called *multiple*

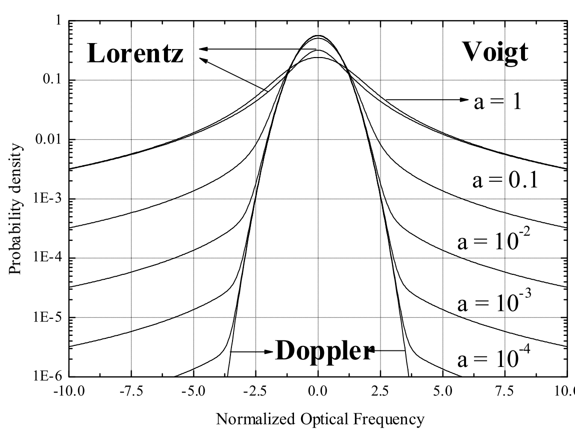

FIG. 2. Doppler, Lorentz, and Voigt spectral profiles.

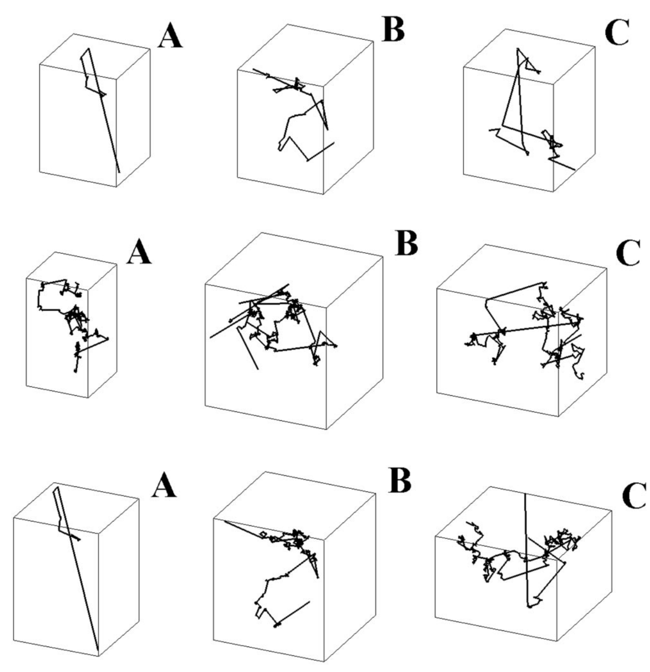

FIG. 3. Single trajectories of 50 000 jumps each for incoherent isotropic CFR radiation migration with Lorentz (top row), Doppler (middle row), and $a = 0.001$ Voigt (bottom row) profiles in infinite 3D medium. (A) shows the whole trajectory while (B) and (C) show successive details. The three trajectories were obtained with the same random number sequence.

scattering representation [13]. In the last case, the temporal evolution is known analytically and the spatial distributions can be separately obtained, either directly from Eq. (1) or from its asymptotic approximation Eq. (4) [18]. It is within this theoretical framework that the results reported in this Letter are most useful. On the other hand, the individual terms in the Holstein expansion have no direct relation with the one (or *n*th) step jump probabilities. Although both approaches are ultimately equivalent [14,19], we follow the multiple scattering approach, preferable for systems of low opacity [14]. Also, the multiple scattering representation has a simple interpretation, since each term corresponds to a specific generation of excited atoms or molecules. In a large number of practical situations the opacity is not high enough to warrant the exclusive use of Holstein's slowest exponential mode. On the other hand, the multiexponential expansion remains valid, but its fundamental mode cannot be identified with Holstein's high opacity result and should be estimated by the *stationary mode* associated with a nonchanging spatial distribution function [14,19], a point often misunderstood in the literature.

Results of Fig. 3 strictly apply to an infinite 3D medium but in a vast number of experimental situations the system is finite and the trajectory is *truncated* before the asymptotic Lévy expansion is able to manifest itself. Mantegna and Stanley [20] introduced in 1994 a class of Lévy flights, the *truncated Lévy flight*, in which the largest steps of an ordinary Lévy flight are eliminated by a sharp cutoff in its power tail. This work allows a reassessment of Kenty's pioneering contribution since he was the first to have used a truncation procedure: within the framework of the kinetic theory of gases he considered a truncated Maxwell distribution of speeds, with a maximum speed corresponding to a free path equal to the linear size of a sample cell [15]. Although *truncation of the jump size* renders the moments of the distribution finite, the convergence to a Gaussian can be extremely slow and the random walk can exhibit anomalous behavior and multiscaling properties in a wide range before convergence [20,21]. Moreover, *trajectory truncation* is more complex to handle and does not create convergence to the Gaussian in usual situations.

In this Letter, it was shown that all photon trajectories arising from incoherent two-level complete frequency redistribution trapping are superdiffusive Lévy flights with $\mu \leq 1$. In particular, for the Doppler line shape $\mu = 1$, whereas for Lorentz and Voigt $\mu = 1/2$.

E. Pereira would like to acknowledge fruitful discussions with François Bardou (CNRS, IPCMS). This work was supported by Fundação para a Ciência e Tecnologia (FCT, Portugal) within project POCTI/34836/FIS/2000.

*Electronic address: epereira@fisica.uminho.pt

[1] P. Lévy, *Théorie de l'Addition des Variables Aléatoires* (Gauthier-Villars, Paris 1954).
[2] J. P. Bouchaud and A. Georges, Phys. Rep. **195**, 127 (1990).
[3] *Lévy Flights and Related Topics in Physics* edited by M. F. Shlesinger, G. M. Zaslavsky, and U. Frisch (Springer-Verlag, Berlin, 1995).
[4] F. Bardou, J. P. Bouchaud, A. Aspect, and C. Cohen-Tannoudji, *Lévy Statistics and Laser Cooling* (Cambridge University, Cambridge, England, 2002).
[5] L. Kador, Phys. Rev. E **60**, 1441 (1999).
[6] J. Klafter, M. F. Shlesinger, and G. Zumofen, Phys. Today **49**, 33 (1996); M. F. Shlesinger, G. M. Zaslasvky, and J. Klafter, Nature (London) **363**, 31 (1993).
[7] A. Ott, J. P. Bouchaud, D. Langevin, and W. Urbach, Phys. Rev. Lett. **65**, 2201 (1990).
[8] T. H. Solomon, E. R. Weeks, and H. Swinney, Phys. Rev. Lett. **71**, 3975 (1993).
[9] G. M. Viswanathan, V. Afanasyev, S.V. Buldyrev, E. J. Murphy, P. A. Prince, and H. E. Stanley, Nature (London) **381**, 413 (1996).
[10] E. Barkai, A.V. Naumov, Yu.G. Vainer, M. Bauer, and L. Kador, Phys. Rev. Lett. **91**, 075502 (2003).
[11] C. Tsallis, S. V. F. Levy, A. M. C. Souza, and R. Maynard, Phys. Rev. Lett. **75**, 3589 (1995).
[12] D. Mihalas, *Stellar Atmospheres* (Freeman, San Francisco, 1978), 2nd ed..
[13] A. F. Molisch and B. P. Oehry, *Radiation Trapping in Atomic Vapours* (Oxford University, Oxford, 1998).
[14] M. N. Berberan-Santos, E. Pereira, and J. M. G. Martinho, in *Resonance Energy Transfer*, edited by D. L. Andrews and A. A. Demidov (John Wiley & Sons, Chichester, 1999).
[15] C. Kenty, Phys. Rev. **42**, 823 (1932).
[16] T. Holstein, Phys. Rev. **72**, 1212 (1947).
[17] B. B. Mandelbrot, *The Fractal Geometry of Nature* (Freeman, New York, 1983).
[18] M. N. Berberan-Santos, E. Pereira, and J. M. G. Martinho, J. Chem. Phys. **103**, 3022 (1995); E. Pereira, M. N. Berberan-Santos, and J. M. G. Martinho, J. Chem. Phys. **104**, 8950 (1996); E. Pereira, M. N. Berberan-Santos, A. Fedorov, M. Vincent, J. Gallay, and J. M. G. Martinho, J. Chem. Phys. **110**, 1600 (1999).
[19] R. Lai, S. Liu, and X. Ma, Z. Phys. D: At., Mol. Clusters **27**, 223 (1993); Opt. Commun. **99**, 316 (1993).
[20] R. N. Mantegna and H. E. Stanley, Phys. Rev. Lett. **73**, 2946 (1994).
[21] H. Nakao, Phys. Lett. A **266**, 282 (2000); I. Koponen, Phys. Rev. E **52**, 1197 (1995).