Experimental Evidence of the Role of Compound Counting Processes in Random Walk Approaches to Fractional Dynamics

Justyna Trzmiel\textsuperscript{*} and Karina Weron\textsuperscript{†}

Institute of Physics, Wrocław University of Technology
Wyb. Wyspiański 27, 50–370 Wrocław, Poland

Aleksander Stanislavsky\textsuperscript{‡}

Institute of Radio Astronomy, 4 Chervonopraporna St., 61002 Kharkov, Ukraine

Agnieszka Jurlewicz\textsuperscript{§}

Hugo Steinhaus Center, Institute of Mathematics and Computer Science, Wrocław University of Technology
Wyb. Wyspiański 27, 50–370 Wrocław, Poland

We present dielectric spectroscopy data obtained for gallium-doped Cd\textsubscript{0.99}Mn\textsubscript{0.01}Te:Ga mixed crystals which exhibit a very special case of the two-power-law relaxation pattern with the high-frequency power-law exponent equal to 1. We explain this behavior, which cannot be fitted by none of the well-known empirical relaxation functions, in a subordinated diffusive framework. We propose diffusion scenario based on a renormalized clustering of random number of spatio-temporal steps in the continuous time random walk. Such a construction substitutes the renewal counting process, used in the classical continuous time random walk methodology, by a compound counting one. As a result, we obtain a novel relaxation function governing the observed non-standard pattern, and we show the importance of the compound counting processes in studying fractional dynamics of complex systems.

PACS numbers: 05.40.Fb, 77.22.Gm, 02.50.Ey

I. INTRODUCTION

No doubt, the linear response theory \cite{1} plays a great role in physics, being a theoretical background and fundamental accomplishments of statistical physics. Its conventional formulation is based on two assumptions \cite{2}: 1) The time evolution of the system variables is governed by Hamiltonian operators; 2) The external perturbation arising from an initial disturbance makes the system depart from canonical equilibrium weakly, and the linear response function is expressed in terms of the derivative of a stationary correlation function (of dipole orientation in the theory of relaxation, for example). This gives a rigorous and efficient approach to the temporal description of some Hamiltonian systems \cite{3}. However, the foundations are crashed on many-body kernel in dynamics of complex systems. The main cause is that the macroscopic evolution of such systems cannot be attributed to any particular object chosen from those forming the systems. Any form of many-body interactions in the complex systems should be introduced through cross-correlations between their different objects, yielding hence insurmountable mathematical problems. Therefore, it is very difficult, if possible generally, to describe the time evolution of many-body systems by Hamiltonian operators, and any stationary correlation function is not available at all. Since, in general, the dynamical processes in complex systems are strictly stochastic in nature, they should be analyzed in a corresponding manner.

For description of relaxation and transport properties in such systems as glasses, liquid crystals, polymers, etc., the continuous-time random walk (CTRW) processes are one of the most useful mathematical tools. Despite their long history, started with the brilliant Montroll-Weiss idea \cite{4}, the CTRWs are still far from their full exploration. However, their connection with anomalous diffusion has been already recognized (see e.g., \cite{5}). Recently, a progress in understanding of this mathematical tool \cite{6–10} stimulated new developments in diffusive scenarios of the non-exponential relaxation phenomena \cite{11–13}.

In this paper we present experimental data which confirm that the non-exponential relaxation behavior, in fact, is governed by compound counting processes strictly connected with clustered CTRWs \cite{9}. In Sec. II we study dielectric spectroscopy data measured for gallium-doped Cd\textsubscript{0.99}Mn\textsubscript{0.01}Te:Ga semiconducting mixed crystals possessing deep, metastable defects. We observe that this material exhibits such a relaxation pattern which cannot be fitted with any of the well-known empirical relaxation functions. Hence, we apply a novel relaxation law being a modification of the result derived recently in \cite{13}. In Sec. III we propose an anomalous diffusion scenario, based on the notion of a compound counting process, by means of which the observed relaxation behavior may be explained. We end with conclusions in Sec. IV.
II. EXPERIMENT

Dielectric spectroscopy studies carried out on various physical systems revealed that a wide class of materials follows the anomalous relaxation mechanism [14, 15] represented by low- and high-frequency fractional power-law dependences of the imaginary part $\varepsilon''(\omega)$ of the complex dielectric permittivity $\varepsilon^*(\omega) = \varepsilon'(\omega) - i\varepsilon''(\omega)$:

$$
\varepsilon''(\omega) \sim (\omega/\omega_p)^m, \quad \omega \ll \omega_p, \\
\varepsilon''(\omega) \sim (\omega/\omega_p)^{m-1}, \quad \omega \gg \omega_p, \tag{1}
$$

where $\omega_p$ denotes the loss peak frequency and $0 < m, n < 1$. Depending on a mutual relation between the power-law exponents two different types of relaxation responses can be distinguished. The relaxation response is called typical when the power-law exponents satisfy relation $m \geq 1 - n$. In order to interpret this type of experimental data the well-know Havriliak-Negami (HN) function [14, 15]

$$
\varphi_HN(\omega) = \frac{1}{[1 + (i\omega/\omega_p)^\alpha]^\gamma}, \quad 0 < \alpha, \gamma < 1 \tag{2}
$$

is used: $\varepsilon^*(\omega) = (\varepsilon_0 - \varepsilon_\infty) \varphi_HN^*(\omega) + \varepsilon_\infty$, where $\varepsilon_0$ is the static permittivity and $\varepsilon_\infty$ represents the asymptotic value of the dielectric permittivity at high frequencies. For the HN function the two-power-law property (1) is fulfilled with the power-law exponents $m = \alpha$ and $1 - n = \gamma\alpha$. For a long time period the HN function with extended parameters’ range $0 < \alpha, \gamma < 1$ was also used to fit the less typical relaxation data for which the power-law exponents yield the opposite inequality $m < 1 - n$. Unfortunately, none of the known relaxation models [12, 16] can justify the values $\gamma > 1$ appearing in the extended range of the power-law exponents. Recent progress in stochastic modeling of relaxation processes has resulted in derivation of a new relaxation pattern [12, 13] underlying the less typical responses:

$$
\varphi^*(\omega) = 1 - \frac{1}{[1 + (i\omega/\omega_p)^\alpha]^\gamma}, \quad 0 < \alpha, \gamma < 1. \tag{3}
$$

This function exhibits the two-power-law property (1) with the power-law exponents $m = \alpha\gamma$ and $1 - n = \gamma\alpha$. It not only properly describes the less typical class of relaxation responses but also relates the experimentally observed power-law properties with random characteristics of the investigated system. Let us notice that both formulas (2) and (3) can be used as fitting functions also with parameters $\alpha = 1$ and/or $\gamma = 1$. In such cases they result from slightly different diffusion scenarios and may exhibit one fractional power-law only or even none.

The less typical relaxation pattern is observed in semiconducting mixed crystals of Cd$_{0.99}$Mn$_{0.01}$Te:Ga possessing deep metastable defects – the so called DX centers. It is widely accepted that depending on the position of gallium (Ga) dopants in the CdMnTe lattice shallow donor states or deep metastable traps may be formed [17]. As shown in Ref. [18] the relaxation response of Cd$_{0.99}$Mn$_{0.01}$Te:Ga is mainly influenced by presence of deep, metastable traps within the band gap of the investigated Au-Cd$_{0.99}$Mn$_{0.01}$Te Schottky junction. For purpose of the present study we analyzed the frequency-domain response of two samples of the same x = 0.01 manganese (Mn) content labeled as sample 1 and sample 2, respectively. Both the samples posses the same net donor concentration of approximately $10^{15}$ cm$^{-3}$ estimated from the capacitance-voltage measurements. Description of the samples preparation can be found in details elsewhere [19]. Gold Schottky contacts were thermally evaporated on the front side of the samples. Measurements were performed at zero bias using Novocontrol impedance analyzer. Applied ac probe signal amplitude was equal to 10 mV.

In Figure 1 the normalized imaginary part of the dielectric permittivity for two samples, investigated in a broad temperature range, is presented. It is clear from the plot that both the samples exhibit the less typical, two-power-law relaxation pattern $m < 1 - n$, however, with different values of the low-frequency power-law exponent $m$. The change in values of this exponent, in the samples of the same manganese content and gallium concentration, may be associated with locally different surroundings of DX centers contributing to the effective relaxation response of the investigated sample.

![FIG. 1: Normalized imaginary part of dielectric permittivity obtained for two samples of Cd$_{0.99}$Mn$_{0.01}$Te:Ga. Both the samples exhibit two-power-law relaxation behavior with power-law exponents satisfying relaxation $m < 1 - n$.](image)
and formula (3) are presented. It can be observed that this function $C_d$ is significantly different depending on the sample considered. Since $\alpha$ is approximately equal to 1, we observe lack of the high-frequency fractional power-law behavior. Such a relaxation pattern, being a very special case of (1), suggests a non-standard diffusion scenario underlying the experimental result.

In Figure 2 the experimental data fitted by means of formula (3) are presented. It can be observed that this function perfectly covers the experimental data points. Values of the fitting parameters are collected in Figure 3. Both $\alpha$ and $\gamma$ values are temperature independent. It should be pointed out that in case of both analyzed samples $\alpha$ parameter remains the same, whereas value of $\gamma$ is significantly different depending on the sample considered. Since $\alpha$ is approximately equal to 1, we observe lack of the high-frequency fractional power-law behavior. Such a relaxation pattern, being a very special case of (1), suggests a non-standard diffusion scenario underlying the experimental result.

### III. DIFFUSION SCENARIO

The CTRW process $R(t)$ determines the total distance reached by a random walker until time $t$. It is characterized by a sequence of independent and identically distributed (i.i.d.) spatio-temporal random steps $(R_i, T_i), i \geq 1$. If we assume stochastic independence between jumps $R_i$ and waiting times $T_i$, we get a decoupled random walk; otherwise, we deal with a coupled CTRW. The distance reached by the walker at time $t$ is given by the following sum

$$R(t) = \sum_{i=1}^{\nu(t)} R_i,$$

where $\nu(t) = \max\{n : \sum_{i=1}^{n} T_i \leq t\}$, counts the performed steps.

Theoretical studies of the relaxation phenomenon in the above framework are based on the idea of an excitation undergoing (anomalous, in general) diffusion in the system under consideration [5]. The relaxation function $\phi(t)$ is then defined by the inverse Fourier or the Laplace transform of the diffusion front $\tilde{R}(t) \approx R(t)/\tau_0 / f(\tau_0)$, where the dimensionless rescaling parameter $\tau_0 \approx 0$ and $f(\tau_0)$ is appropriately chosen renormalization function. The diffusion front $\tilde{R}(t)$ approximates a position at time $t$ of the walker performing rescaled spatio-temporal steps $(R_i/f(\tau_0), \tau_0 T_i)$. The characteristics of the relaxation process are related to the properties of the diffusion front resulting from assumptions imposed on the spatio-temporal steps of the random walk. For example, the decoupled CTRW process with power-law waiting-time distributions (i.e., with the random variables $T_i$ satisfying $\text{Prob}(T_i \geq t) \sim (t/t_0)^{-a}$ as $t \to \infty$ with some $0 < a < 1$ and $t_0 > 0$) leads to the Cole-Cole relaxation [20].

However, the frequency-domain Cole-Cole relaxation with the corresponding time-domain Mittag-Leffler pattern is only one of the cases measured in various experiments with complex media, and derivation of those more general patterns requires considering diffusion scenarios based on a compound coupled CTRW representation. It should be pointed out that the simple coupling of type $R_i \sim T_i^p$ (with positive power exponent $p$) does not lead behind the Cole-Cole relaxation [21]. In contrast, introducing a dependence between the jumps and waiting times by a random clustering procedure, we can obtain another empirical relaxation laws like the Cole-Davidson or Havriliak-Negami patterns [12,13,22]. Below, we present the diffusion scenario which leads directly to the results discussed in the preceding section.

Let $M_i$ be a sequence of i.i.d. positive integer-valued random variables independent of the pairs $(R_i, T_i)$. Next, assume that the jumps and waiting times are assembled into clusters of random sizes $M_1, M_2, \ldots$. This assumption allows one to transform the sequence of spatio-temporal steps $(R_i, T_i)$ into a new sequence $(\tilde{R}_j, \tilde{T}_j)$ of random sums

$$(\tilde{R}_1, \tilde{T}_1) = \sum_{i=1}^{M_1} (R_i, T_i),$$
Then the position \( R^M(t) \) of the walker is determined by \((\tilde{R}_j, \tilde{T}_j)\) and, in accordance with the general formula \([4]\), it is given by

\[
R^M(t) = \sum_{j=1}^{n} \tilde{R}_j,
\]

(6)

where \( \tilde{\nu}(t) = \max\{n : \sum_{j=1}^{n} \tilde{T}_j \leq t\} \). The dependence between the jumps \( \tilde{R}_j \) and the waiting times \( \tilde{T}_j \) of the coupled CTRW process \( R^M(t) \) is determined by the distribution of the cluster sizes \( M_j \).

In the simple case when the waiting times are represented by equal intervals in time, i.e., \( T_i = \Delta t \), we have

\[
R(t) = \sum_{i=1}^{\lfloor t/\Delta t \rfloor} R_i,
\]

(7)

where \( \lfloor \cdot \rfloor \) denotes the integer part, while the clustering procedure \([5]\) yields \( \tilde{T}_j = M_j \Delta t \) for \( j \geq 1 \), and the coupled process \( R^M(t) \) in \([6]\) takes an equivalent form

\[
R^M(t) = \sum_{i=1}^{\tilde{\nu}(t/\Delta t)} R_i .
\]

(8)

Here \( U^M(\tilde{\nu}(t)) \) is a compound counting process obtained from \( U^M(n) = \sum_{j=1}^{n} \tilde{T}_j/\Delta t = \sum_{i=1}^{n} M_j \), and \( \tilde{\nu}(t) = \max\{n : \sum_{j=1}^{n} \tilde{T}_j \leq t\} = \max\{n : \tilde{U}^M(n) \leq t/\Delta t\} \). Observe that formula \([8]\) is an analog of \([7]\) with the compound counting process \( U^M(\tilde{\nu}(t)) \) substituting the deterministic number \( \lfloor t/\Delta t \rfloor \) of performed jumps \( R_i \). The counting process \( U^M(\tilde{\nu}(t)) \) is always less than \( \lfloor t/\Delta t \rfloor \), and it is hence a special case of the undershooting compound counting process \([12]\). It is also a clear signature of the spatio-temporal coupling provided by the clustering procedure \([5]\).

The idea of compound counting processes in CTRW approach is not new in physics. The resulting CTRW processes were examined in the context of the rareness hypothesis in the fractal-time random walk models (see, e.g., \([23, 24]\)). In general, the compound counting process cumulates random number of random events. Physical situations where the relevance of this scheme holds are numerous. For instance, take into account the energy release of individual earthquakes in geophysics, the random magnitude of claims’ sequence in insurance risk theory or random water inputs flowing into a dam in hydrology where summing the individual contributions yields the total amount of the studied physical magnitude over certain time intervals.

The diffusion front \( \tilde{R}^M(t) \) related to \([8]\) takes the subordinated form

\[
\tilde{R}^M(t) \overset{d}{=} X(Z(t)) ,
\]

(9)

where the parent process \( X(\tau) \) is just the diffusion front corresponding to the simple random walk \( R(t) \) given by \([4]\), while the undershooting subordinator \( Z(\tau) \) corresponds to the limit of the rescaled counting process \( U^M(\tilde{\nu}(t/\tau_0))/g(\tau_0) \) as \( \tau_0 \to 0 \) (with renormalizing function \( g(\tau_0) \) chosen appropriately). Both the parent process and the undershooting subordinator are well defined if the distributions of the spatial steps \( R_i \) and cluster sizes \( M_j \) satisfy some conditions, referring to their asymptotic behaviors. In particular, if \( \langle M_j \rangle < \infty \) (or equivalently \( \langle \tilde{T}_j \rangle < \infty \) we have \( Z(\tau) = t/\Delta t \). On the other hand, taking into account clustering with a heavy-tailed cluster-size distribution

\[
\text{Prob}(M_j \geq m) \sim m^{-\gamma}/m^{-\gamma}
\]

with the tail exponent \( 0 < \gamma < 1 \) and some scaling constant \( c > 0 \), we obtain the compound form \( Z(t) = U^-_{\gamma}[S_{\gamma}(t/\Delta t)] \).
where $U_{\gamma}^{-}(\tau) = \lim_{x \to -\tau} U_{\gamma}(x)$ is the left limit of the $\gamma$-stable subordinator $U_{\gamma}(\tau)$, and $S_{\gamma}(t) = \inf\{t \geq 0 : U_{\gamma} > t\}$ is its inverse process. For a fixed $t > 0$, the compound undershooting subordinator $U_{\gamma}^{-}[S_{\gamma}(t/\Delta t)]$ has the general-
ized arcsine distribution rescaled by $t/\Delta t$. The corresponding probability density function reads

$$p_{\gamma}(t, \tau) = \frac{\sin \pi \gamma}{\pi} \tau^{\gamma - 1} (t/\Delta t - \tau)^{-\gamma}, \quad 0 < \tau < t/\Delta t. \quad (10)$$

It is easy to check that all moments of the random variable $U_{\gamma}^{-}[S_{\gamma}(t/\Delta t)]$ are finite.

The numerical approximation of the process $U_{\gamma}^{-}[S_{\gamma}(t/\Delta t)]$ is shown in Fig. 4. For simplicity, we take $\Delta t = 1$. To simulate the process $U_{\gamma}^{-}[S_{\gamma}(t)]$, one only needs to generate the values $U_{\gamma}(n \Delta t)$ (see Fig. 4(a)), where $n = 1, 2, \ldots$, and $\Delta t$ is the step length. This can be carried out by the standard method of summing up the independent and stationary increments of the Lévy process $[25]$. Then the approximation of the process $U_{\gamma}^{-}[S_{\gamma}(n \Delta t)]$ is a simple continuous-time random walk in which each waiting time is exactly equal to the jump. In the result, Fig. 4(b) demonstrates the process $U_{\gamma}^{-}[S_{\gamma}(t)]$ non-decreasing in time.

As far as the parent process is concerned, for simplicity, but without loss of generality, we can consider the standard Brownian motion $B(t)$ that can easily be obtained if we take into account the spatial jumps satisfying conditions $R_{\gamma} = 0$ and $R_{\gamma}^{2} < \infty$ (see Fig. 4(c)). Then the subordinated process $B(U_{\gamma}^{-}[S_{\gamma}(t)])$ represents the corresponding diffusion front. Its sample paths are shown in Fig. 4(d). The main feature of the subordinated process consists in random jumping along random trajectories of the parent process. For better visualization of this feature we can consider a walk on a plane with 2D vector jumps satisfying analogous conditions that leads to 2D Brownian motion as a parent process, see Fig. 5. The difference between Figs. 5(a) and (b) is that the walker, moving along a Brownian trajectory in presence of the compound sub-
ordination, stops from time-to-time and overjumps through intermediate positions in the Brownian trajectory. In other words, the trajectory of Fig. 5(b) is something like a partition of the trajectory in Fig. 5(a) on random intervals. The appearance of the overjumps, as applied to the relaxation of semiconductors with metastable defects described in Sec. 11, is connected with long-range interactions among DX centers.

The probability density function of $B(U_{\gamma}^{-}[S_{\gamma}(t/\Delta t)])$ reads

$$p(x, t) = \int_{0}^{\infty} p^{B}(x, \tau) p_{\gamma}(t, \tau) \, d\tau, \quad (11)$$

where $p^{B}(x, \tau) = \frac{1}{\sqrt{2\pi \tau}} e^{-x^{2}/2\tau}$. Taking the inverse Fourier transform with respect to $x$ for $p(x, t)$ in Eq. (11), we derive the relaxation function $\phi(t) = \langle e^{-ikB(t)} \rangle$ that characterizes the temporal decay of a given macroscopic mode $k$. We get

$$\phi(t) = \frac{\sin \pi \gamma}{\pi} \int_{0}^{t/\Delta t} e^{-(k^{2}/2\tau)} \tau^{\gamma - 1} (t/\Delta t - \tau)^{-\gamma} \, d\tau, \quad (12)$$

where $k^{2}/2$ may be denoted as a characteristic frequency $\omega_{p}$ of the relaxation system. The integral representation of the relaxation function may be substituted by a series expansion observing that the relaxation function $\phi(t)$ can be expressed as

$$\phi(t) = E_{\alpha, \beta}^{\gamma}(\omega_{p}, t),$$

is the generalized Mittag-Leffler function $[26]$. Here $(\gamma, k) = \gamma(n + 1)(\gamma + 2) \ldots (\gamma + k - 1)$ is the Appell’s symbol with $(\gamma, 0) = 1, \gamma \neq 0$.

Based on the Laplace image of $\phi(t)$ with respect to $t$ we find the kinetic equation of relaxation in the pseudodifferential form

$$\left(\frac{d}{dt} + \omega_{p}^{\gamma}\right) \phi(t) = \frac{t^{-\gamma}}{\Gamma(1 - \gamma)},$$

with the initial condition $\phi(0) = 1$. For the experimental studies of the dielectric spectroscopy data (dielectric permittivity or the corresponding susceptibility) the frequency-domain representation of the latter function

$$\varphi^{\ast}(\omega) = \int_{0}^{\infty} e^{-i\omega \tau} \left(-\frac{\partial \phi(t)}{\partial t}\right) dt \quad (13)$$

is of interest. Taking the Fourier transform, we get

$$\varphi^{\ast}(\omega) = 1 - \left(\frac{i\omega/\omega_{p}}{1 + i\omega/\omega_{p}}\right)^{\gamma}, \quad 0 < \gamma < 1. \quad (14)$$

Let us note that formula (14) coincides with (3) for $\alpha = 1$. Moreover, for $\gamma = 1$ it takes the form of the Debye re-
sponse, corresponding to the spatio-temporal clustering of $(M_{x}) < \infty$, where the undershooting subordinator becomes deterministic: $Z(t) = t/\Delta t$.

Considering the dielectric susceptibility $\chi^{\ast}(\omega) = \chi^{\prime}(\omega) - i\chi^{\prime\prime}(\omega) = \chi^{0}(0) \varphi^{\ast}(\omega)$ with $\varphi^{\ast}(\omega)$ as in (13) one can show that the real and imaginary parts of the susceptibility fulfill the following frequency-independent relations:

$$\lim_{\omega \to 0} \frac{\chi^{\prime\prime}(\omega)}{\chi^{\prime}(\omega)} = \tan \left(\frac{\pi \gamma}{2}\right),$$

$$\lim_{\omega \to \infty} \frac{\chi^{\prime\prime}(\omega)}{\chi^{\prime}(\omega)} = \infty. \quad (15)$$

This means that for small $\omega$ the gallium-doped Cd$_{0.99}$Mn$_{0.01}$Te mixed crystals obey the energy crite-
ron $[13]$, while for large $\omega$ the high-frequency energy lost per cycle does not have a constant relationship to the extra energy that can be stored by a static field. The similar feature takes place in the Cole-Davidson law where, however, the energy criterion is obeyed for large $\omega$ only. As it is well known $[15]$, the presence of the universal law in the relaxation in gallium-
doped Cd$_{0.99}$Mn$_{0.01}$Te mixed crystals is caused strictly by many-component interactions. This effect is just accounted for using the compound counting random processes.
IV. CONCLUSIONS

In the diffusive framework we have brought to light the fractional dynamics of gallium-doped Cd$_{0.99}$Mn$_{0.01}$Te:Ga mixed crystals, for which the particular case of less typical relaxation behavior with the high-frequency power-law exponent $1 - n = 1$ has been detected. We have proposed the sub-ordination scenario of anomalous diffusion underlying the observed pattern, based on random, heavy-tailed spatio-temporal clustering procedure applied to the random walk hidden behind the standard Brownian motion. Such a procedure has led to a substitution of the renewal counting process, used in the classical CTRW model, by the undershooting compound counting one. As a consequence, the explicit stochastic structure of the diffusion front as a subordinated Brownian motion has been obtained. The resulting frequency-domain relaxation function has been shown to fit the studied dielectric spectroscopy data.

V. ACKNOWLEDGEMENTS

Work of J.T. and A.J. was partially supported by project PB NN 507503539.

A.S. is much obliged to the Institute of Physics and the Hugo Steinhaus Center for pleasant hospitality during his visit in Wroclaw University of Technology.

J.T. is grateful to dr hab. Ewa Popko for making the samples of Cd$_{0.99}$Mn$_{0.01}$Te:Ga accessible for measurements.

A.S. thanks dr Marcin Magdziarz for his useful remarks.

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