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A CONTINUOUS-TIME RANDOM-WALK APPROACH TO THE COLE–DAVIDSON DIELECTRIC RESPONSE OF DIPOLAR LIQUIDS*

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Dedicated to Professor Andrzej Fuliński on the occasion of his 70th birthday

We show how the Cole–Davidson relaxation response, characteristic of alcoholic systems, can be derived within the framework of the continuous-time random walk (CTRW). Using the random-variable formalism, we indicate that the high-frequency power law of dielectric spectra is determined by the heavy-tailed distribution of quantities that provide the spatio-temporal coupling in the random-walk process. As an illustration, we present the dielectric permittivity spectra of several butanediol isomers.

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1. Introduction

Dielectric relaxation spectroscopy is a technique sensitive to the reorientation of dipole moments in materials. The method is widely used to study interactions in dipolar, hydrogen-bonded liquids such as aqueous mixtures or alcoholic systems [1, 2]. Investigations of the dielectric permittivity spectra of water [3, 4], alcohols [3], alcohol/water [5, 6] and alcohol/alcohol mixtures [7–9], electrolyte solutions [10, 11], *etc.* may lead to a better understanding of the unique characteristics of water as a bulk liquid and as

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a solvent embedding solutes. Dipole relaxation experiments yield insight to the wetting properties of water and the physical basis of the hydrophobic effect as a general phenomenon of the interplay among hydrophobic and hydrophilic compounds [12–15]. More specifically, studies of dielectric relaxation in biological systems, especially at interfaces [16], may help elucidate the biological roles of water. Despite vast attention, however, the theoretical understanding of relaxation behaviour in the above mentioned systems is still insufficiently developed.

Experimental data provide clear evidence that the complex permittivity spectra $\varepsilon^*(\omega) = \varepsilon'(\omega) - i\varepsilon''(\omega)$ of aqueous mixtures and alcohols strongly deviate from the classical Debye pattern. The deviations, indicated by broader and asymmetric shapes of the absorption term $\varepsilon''(\omega)$ of the spectra, are often well fitted with the empirical Cole–Davidson function

$$\Phi_{\mathrm{CD}}(\omega) = \frac{\varepsilon^*(\omega) - \varepsilon_\infty}{\varepsilon_{\mathrm{s}} - \varepsilon_\infty} = \frac{1}{\left(1 + \frac{i\omega}{\omega_{\mathrm{p}}}\right)^{\gamma}}\,, \qquad 0 < \gamma < 1\,, \tag{1}$$

where ω_{p} denotes the loss peak frequency, ε_{s} is the static permittivity, and ε_∞ the high-frequency permittivity. The Debye expression corresponds to $\gamma = 1$. Let us note that a fit with the Cole–Davidson function $\Phi_{\mathrm{CD}}(\omega)$ yields the following limiting properties of the complex permittivity [17]:

1. the high-frequency power law

   $$\varepsilon'(\omega) \propto \varepsilon''(\omega) \propto \left(\frac{\omega}{\omega_{\mathrm{p}}}\right)^{-\gamma}, \qquad \text{for} \qquad \omega \gg \omega_{\mathrm{p}}\,, \tag{2}$$

   indicating the deviations from Debye behaviour in the form $(\omega/\omega_{\mathrm{p}})^{-1}$,

2. the low-frequency characteristics

   $$\varepsilon'(0) - \varepsilon'(\omega) \propto \left(\frac{\omega}{\omega_{\mathrm{p}}}\right)^{2} \quad \text{and} \quad \varepsilon''(\omega) \propto \left(\frac{\omega}{\omega_{\mathrm{p}}}\right)^{1}, \quad \text{for} \quad \omega \ll \omega_{\mathrm{p}}\,, \tag{3}$$

   coinciding with the properties of the Debye function.

Such a common property of the dielectric spectra, found in different alcoholic systems (*i.e.*, 1,2 ethanediol, 1,4 butanediol, benzyl, *n*-hexyl, ethanol–water mixture, water–ethanol–glycine mixtures, butanediol–benzyl mixtures, propanediol–ethanediol–hexanol mixtures, *etc.* [8,9]), suggests a kind of general behaviour that is independent of the details of the studied systems. This assumption is justified in the framework of statistical models, where averaging principles like the law of large numbers and limit theorems are in force for the large scale behaviour of complex systems [18–20].

The non-Debye behaviour of complex OH-systems such as alcohols is often interpreted in terms of a "wait-and-switch" relaxation mechanism [21]. In this approach, it is assumed that reorientation follows an activated jump mechanism. The direction of a dipole moment at any given time corresponds to an energy minimum; different dipole orientations are separated by potential energy barriers. Reorientation is initiated by the presence of an additional neighbour in a suitable position. This additional molecule first flattens the potential energy barrier and subsequently offers the possibility of forming a new hydrogen bond. The dielectric relaxation time is hence determined by the period in which an additional neighbour approaches, *i.e.* the local diffusion coefficient. The "wait-and-switch" model resembles the concept of Glarum [22], who assumed that vacancies diffuse within the examined system and that when they meet with an initially prepared excited state the latter is allowed to relax. Following Glarum's idea and the notion of a continuous-time random walk (CTRW), introduced by Montroll and Weiss [23], a target model for describing anomalous (non-Debye) relaxation processes has been proposed by Shlesinger [24] and Blumen *et al.* [25]. An alternative and novel approach to anomalous dynamics of complex systems has been recently proposed in [26, 27].

This paper follows the CTRW approach to relaxation. We demonstrate, however, the power of the mathematical tools underlying the concept of the CTRW by showing how it can be generalised to handle Cole–Davidson relaxation in dipolar-liquid systems. Our attempt is based directly on the definition of a cumulative stochastic process. By using the random-variable formalism [18–20], introduced recently to study random-walk processes, we show that the limit theorems of probability theory lead to rigorous results. This approach avoids the technical difficulties that arise in the classical approach to the CTRW, which is based on Fourier–Laplace transform analysis [28, 29].

As a demonstration of the discussed Cole–Davidson relaxation, we present the dielectric permittivity spectra of several butanediol isomers. These compounds have many applications, *e.g.* as industrial solvents and in cryopreservation. They are often regarded as model systems of biomolecules bearing both polar and non-polar groups. The special interest in the dielectric relaxation of these isomers results from the variable distance of their relaxing groups, giving thus access to the coupling mechanism among these groups. It is hence essential to gain knowledge of their dielectric responses for understanding their behaviour. In this study, the properties of the butanediol isomers were analysed in the frequency range 10 MHz–4 GHz, and the complex permittivity data were fitted with the Cole–Davidson function. Obtained parameters were compared to available literature values [7, 8, 30–32].

2. Experiment and data analysis

2.1. Permittivity measurements

The four butanediol isomers, namely 2,3-, 1,2-, 1,3-, and 1,4-butanediol (structural formulas shown in Fig. 1), were purchased from Sigma–Aldrich Chemie GmbH (Sreinheim, Germany) and used as received. Samples were probed by an alternating electric field, with frequencies in the range 10 MHz $\leq \nu \leq$ 4 GHz, generated by a Vector Network Analyzer (ZVRE, Rhode & Schwarz). The field was introduced into the liquid samples through a small (approximately 2 mm in diameter) cylindrical coaxial electrode; the reflected field was detected by the same electrode and the effective reflection coefficient (ratio of the incoming to outgoing fields) recorded by the Network Analyzer. The amplitude of the generated field was 38 dBmV, and all measurements were performed at approximately $(20 \pm 4)^\circ$ C.

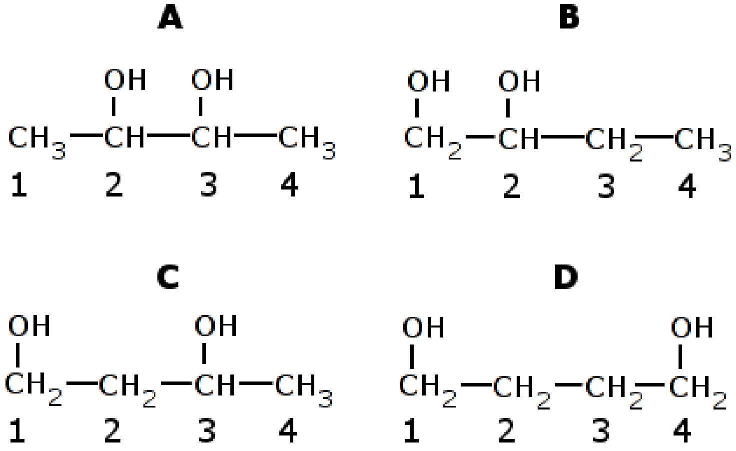

Fig. 1. Structural formulas of the four butanediol isomers: 2,3- (A), 1,2- (B), 1,3- (C), and 1,4- (D) butanediol. The carbon chain positions (1–4) are numbered to show the possible positions of the two hydroxyl groups, which distinguishes the four isomers.

Complex permittivity spectra $\varepsilon^*(\omega)$ were obtained from the measured reflection coefficient $S^*(\omega)$ by using the proportionality:

$$\varepsilon^*(\omega) \propto \frac{1}{i\omega}\, Y^*(\omega) \propto \frac{1}{i\omega}\, \frac{1 - S^*(\omega)}{1 + S^*(\omega)}\,, \tag{4}$$

where $Y^*(\omega)$ is the complex electric admittance of the sample.

In order to compensate for losses in the field and factors of geometrical origin in the proportionality (4), the reflection coefficient was calibrated with respect to air, for which it was assumed to be unity (total reflection, or $S^* = 1.0$), and permittivity was calibrated with respect to water. The final equation used for calculating the permittivity of any sample is then

$$\varepsilon^*(\omega) = \left(\frac{S^*_{\text{air}}(\omega) - S^*(\omega)}{S^*_{\text{air}}(\omega) + S^*(\omega)}\right)\left(\frac{S^*_{\text{air}}(\omega) + S^*_{\text{water}}(\omega)}{S^*_{\text{air}}(\omega) - S^*_{\text{water}}(\omega)}\right)\varepsilon^*_{\text{water}}(\omega)\,, \tag{5}$$

where S^*, S^*_{air}, and S^*_{water} are the non-calibrated reflection coefficients of the sample, air, and water, respectively, as obtained directly from the Network Analyzer output. The permittivity of water in (5), $\varepsilon^*_{\text{water}}(\omega)$, was calculated using a single Debye function with literature parameters interpolated to 20°C [4].

2.2. Data analysis

The obtained complex permittivity spectra were fitted with the Cole–Davidson function in the form:

$$\varepsilon^*(\nu) = \varepsilon_\infty + \Delta\varepsilon\,\Phi_{\text{CD}}(\omega) = \varepsilon_\infty + \frac{\Delta\varepsilon}{(1 + i2\pi\nu\tau)^\gamma}\,, \tag{6}$$

where

$$\Delta\varepsilon = \varepsilon_{\text{s}} - \varepsilon_\infty\,,$$

$$\tau = \frac{1}{\omega_{\text{p}}}\,, \qquad \text{and} \qquad \nu = \frac{\omega}{2\pi}\,.$$

A modified Powell conjugate gradient algorithm was used to minimise the normalised variance χ^2, given by

$$\chi^2 = \frac{1}{2N - m - 1}\left[\frac{1}{\max(|\varepsilon^*(\nu_i)|)}\sum_{i=1}^{N}\left|\delta\varepsilon^*(\nu_i)\right|^2\right], \tag{7}$$

where $\delta\varepsilon^*(\nu_i)$ are the residuals of the complex permittivity, i numbers the frequency data points in logarithmically equidistant steps ($N = 200$), and m is the number of adjustable fit parameters (in this case there are four such parameters, namely ε_∞, $\Delta\varepsilon$, τ, and γ).

2.3. Results

Table I presents the best fit parameters for the measured data of all four butanediol isomers within the Cole–Davidson response (6). The parameter ε_{s} agrees well with literature values (± 1 for all isomers) [30–32], given the inaccuracy of the temperature here. The value of $\gamma\tau$ for 1,4-butanediol has been reported by Hanna *et al.* [8] to be 820 ps at 20°. Our value (785 ps) differs by around 4%, which again is acceptable considering the temperature uncertainty. The parameter χ^2, calculated according to (7), was minimised to obtain the optimised fits.

TABLE I

Dielectric relaxation parameters of the four butanediol isomers for the best fit with the Cole–Davidson function at approximately $20° \pm 4°$C.

	2,3-BD	1,2-BD	1,3-BD	1,4-BD
ε_{s}	23.1	22.1	30.7	32.7
$\Delta\varepsilon$	21.6	20.7	28.6	30.5
γ	0.58	0.65	0.73	0.77
$\gamma\tau$ [ps]	853	482	1043	785
χ^2	$4.4\ 10^{-5}$	$4.9\ 10^{-5}$	$4.8\ 10^{-5}$	$5.4\ 10^{-5}$

Accordingly, Fig. 2 shows the spectra for the most contrasting isomers, namely 2,3-BD and 1,4-BD. The Debye and Cole–Davidson functions that best fit the data have also been plotted.

It is clear that a Debye description is unsatisfactory as compared to the Cole–Davidson one. This can be especially seen for the absorption (imaginary) component of the spectra. The Cole–Davidson function offers a much better description in the studied frequency range, reflecting the slope of the dispersion curve and broadness of the absorption curve. A distinguishable shoulder at high-frequencies, which is more pronounced in the case of 1,4-butanediol than 2,3-butanediol, gives an evidence for another high-frequency process taking over. The frequency range applied in this study does not cover it entirely, therefore, it is not analysed here in more detail.

High-frequency deviations in the absorption spectra of alcoholic systems are often analysed by adding additional terms to the function used to fit the measured data [3,7]. It should be noted that the resulting formal description — a sum of processes $\sum_i f_i(\omega)$ — allows data to be easily compared in literature, but does not provide any distinct physical meaning of each term in itself.

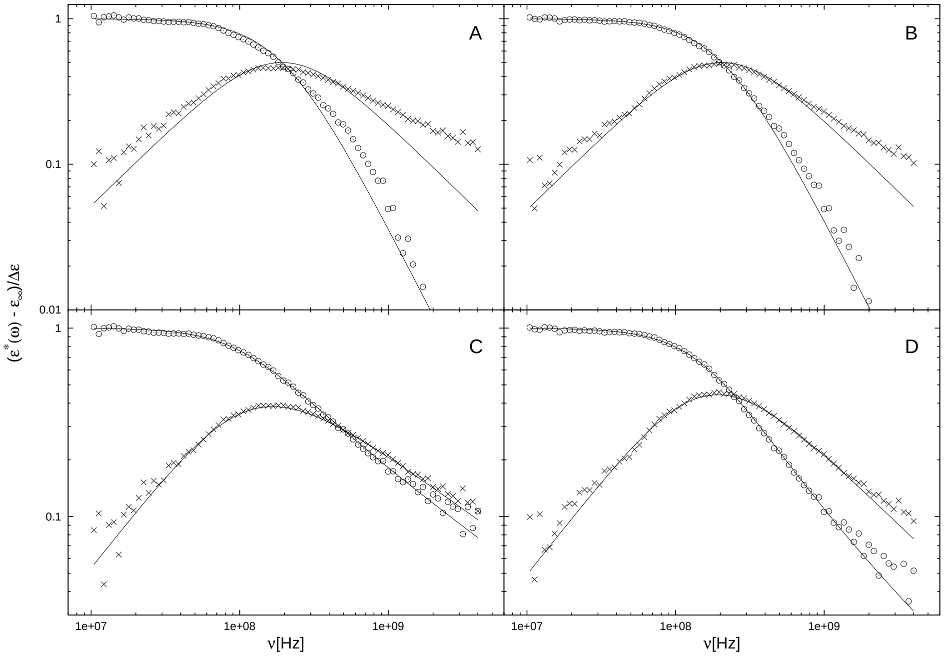

Fig. 2. Debye (A,B) and Cole–Davidson (C,D) fits to the complex permittivity spectra of 2,3- (A,C) and 1,4- (B,D) butanediols at approximately 20° ± 4°C. The spectra are plotted as $(\varepsilon^*(\omega) - \varepsilon_\infty)/\Delta\varepsilon$ to emphasise the power laws (the parameters ε_∞ and $\Delta\varepsilon$ are taken from the corresponding fits). All plots contain the real ($\circ$) and imaginary ($\times$) parts of the spectra, and fits are shown as solid lines.

3. The random-walk mechanism of the Cole–Davidson relaxation

Let us consider a stochastic process generated by a sequence of independent and identically distributed (i.i.d.) random vectors $\{(R_i, T_i), i = 1, 2, \ldots\}$, where the random variable $T_i > 0$ is assigned to the time a particle waits before it jumps. The length and the direction of such a jump is reflected in the random variable R_i. The total distance $R(\tau)$ travelled by the particle, initially at the origin of some coordinate system, during an extended large time τ defines a process known as the continuous-time random walk (CTRW) [23]. In general, this distance may be expressed as a random sum of random jumps

$$R(\tau) = \sum_{i=1}^{\nu(\tau)} R_i \,, \tag{8}$$

where the random number of components $\nu(\tau)$ is determined by the time steps T_i, namely $\nu(\tau)$ is the smallest index k for which the sum of waiting times $T_1 + T_2 + \ldots + T_k$ exceeds the time of observation τ.

It should be noted that there are two types of the CTRWs: *decoupled*, for which the random variables R_i and T_i are independent, and *coupled*, for which the random variables R_i and T_i are dependent.

In this paper, we focus on the special case of coupled processes [20], with the space steps R_i and time steps T_i expressed as random sums:

$$R_i = \sum_{j=1}^{M_i} \Delta R_{ij}\,, \quad T_i = \sum_{j=1}^{M_i} \Delta T_{ij}\,. \tag{9}$$

The sequences of space and time spans, $\{\Delta R_{ij}, i, j = 1, 2, \ldots\}$ and $\{\Delta T_{ij}, i, j = 1, 2, \ldots\}$, consist of i.i.d. positive random variables, and $\{M_i, i = 1, 2, \ldots\}$ consists of i.i.d. positive integer-valued random variables. Additionally, the independence of all sequences is assumed. Although we assume the independence of space and time random spans, a non-degenerate distribution of the number of summands M_i provides a stochastic dependence between them.

In the considered case, the total distance $R(\tau)$ has the same distribution as the following random sum of space spans [33]:

$$R(\tau) = \sum_{k=1}^{L(\tau)} \Delta R_{1k}\,, \tag{10}$$

where the index $L(\tau)$ is given by the following random sum of M_i's:

$$L(\tau) = \sum_{i=1}^{N(K(\tau))} M_i\,, \tag{11}$$

with the number of summands given by the formula

$$\begin{aligned} N(K(\tau)) &= \min\left\{ n : \sum_{i=1}^{n} M_i > K(\tau) \right\}\,, \\ \text{for} \quad K(\tau) &= \min\left\{ l : \sum_{j=1}^{l} \Delta T_{1j} > \tau \right\}\,. \end{aligned} \tag{12}$$

The relaxation function defined within the CTRW framework [34] can be expressed as the Laplace transform of the rescaled total distance (diffusion front) $\tilde{R}(t)$

$$\phi(t) = \left\langle e^{-\mathrm{k}\tilde{R}(t)} \right\rangle\,, \tag{13}$$

where k is a positive constant and the diffusion front is defined as the following limit of the distribution

$$\frac{R(t/\delta\tau)}{f(\delta\tau)} \xrightarrow[\delta\tau\to 0]{\mathrm{d}} \tilde{R}(\tau)\,. \tag{14}$$

Here, $\overset{\mathrm{d}}{=}$ denotes an equality in distributions. In the above formula, $\delta\tau$ is a characteristic time scale and $f(\delta\tau)$ is an appropriately chosen rescaling function. In our calculations, we consider the rescaling function $1/\delta\tau$.

The characteristics of the diffusion front $\tilde{R}(t)$ depend on the assumptions used [20, 33] for the distributions of the variables that describe space spans ΔR_{ij}, time spans ΔT_{ij}, and the counting random variable M_i. Aiming to find the origins of the high-frequency power law (2), we proceed to discuss the assumptions that lead to the classical Debye and its modification expressed in the Cole–Davidson response.

Let us first consider the case when the random variables denoting space and time spans have finite mean values. We assume these values to be determined by the space and time units, *i.e.* $\langle\Delta R_{ij}\rangle = \Delta R$, and $\langle\Delta T_{ij}\rangle = \Delta T$. If we assume that the number M_i also has a finite mean value, then for any non-negative t

$$\frac{R(t/\delta\tau)}{1/\delta\tau} \xrightarrow[\delta\tau\to 0]{\mathrm{a.s.}} \tilde{R}(t) = Ct\,. \tag{15}$$

Here, $\xrightarrow{\mathrm{a.s.}}$ reads "tends with probability 1", and $C = \Delta R/\Delta T$. For a diffusion front of the form (15), the relaxation function (13) is equal to

$$\phi(t) = \left\langle e^{-C\mathrm{k}t}\right\rangle = e^{-\omega_{\mathrm{p}}t}\,, \tag{16}$$

for $\omega_{\mathrm{p}} = C\mathrm{k}$. The exponentially decaying time-domain relaxation function corresponds to the frequency-domain Debye relaxation response given by the Fourier transform

$$\varepsilon^*(\omega) \propto \int\limits_0^\infty e^{-i\omega t}\left(-\frac{d\phi(t)}{dt}\right)\,dt = \frac{1}{1+\frac{i\omega}{\omega_{\mathrm{p}}}}\,. \tag{17}$$

In the next step, while keeping the assumptions put on ΔR_{ij} and ΔT_{ij}, the same as in the Debye case, we additionally assume a heavy-tailed distribution for M_i with a heavy-tail exponent $0 < \gamma < 1$. Let us note that the notion of a heavy tail with a heavy-tail exponent γ means that the inequality $\Pr(M_i \geq m) \propto m^{-\gamma}$ is valid for large values of m. In such a case, the diffusion front, as obtained in (14), is distributed as the inverse of B_γ [20]

$$\tilde{R}(t) \overset{\mathrm{d}}{=} Ct\,\frac{1}{B_\gamma}\,, \tag{18}$$

where B_γ is distributed according to the probability density function

$$f_\gamma(x) = \begin{cases} \dfrac{1}{\mathit{\Gamma}(\gamma)\mathit{\Gamma}(1-\gamma)}\, x^{\gamma-1}(1-x)^{-\gamma} & \text{for } 0 < x < 1\,, \\ 0 & \text{otherwise}\,, \end{cases}$$

which is the beta distribution with parameters $p = \gamma$ and $q = 1 - \gamma$. The corresponding relaxation function has the form

$$\phi(t) = \left\langle e^{-C\mathrm{k}t\frac{1}{B_\gamma}} \right\rangle = \Pr(\mathit{\Gamma}_\gamma \geq C\mathrm{k}t)\,, \tag{19}$$

where $C = \Delta R/\Delta T$ and $\mathit{\Gamma}_\gamma$ is a random variable defined by the gamma density function with the parameter γ:

$$g_\gamma(t) = \begin{cases} \dfrac{C\mathrm{k}}{\mathit{\Gamma}(\gamma)}(C\mathrm{k}t)^{\gamma-1}e^{-\omega_{\mathrm{p}}t} & \text{for } t > 0\,, \\ 0 & \text{otherwise}\,. \end{cases}$$

Inserting $\phi(t)$ given by (19) into formula (17) leads to the following expression for the complex permittivity:

$$\varepsilon^*(\omega) \propto \int\limits_0^\infty e^{-i\frac{\omega}{C\mathrm{k}}t} g_\gamma(t)\, dt \propto \frac{1}{\left(1 + \frac{i\omega}{C\mathrm{k}}\right)^\gamma}\,, \tag{20}$$

which becomes the Cole–Davidson relaxation response (1) if $\omega_{\mathrm{p}} = C\mathrm{k}$.

Let us finally observe that the derivation of the Debye and the Cole–Davidson responses requires detailed information on the probability distribution of neither the space R_{ij} nor time T_{ij} spans. The only important point is to deal in theses cases with finite-expected-value distributions. As a consequence, the space and time spans can be simply considered to be non-random constants to get the same result as for randomly chosen spans. Departure from the Debye to any nonexponential relaxation appears in connection with heavy-tailed distributions of CTRW variates [33]. The high-frequency power law (2), as expressed by the Cole–Davidson response function, is determined by the heavy-tailed exponent γ of the distribution of the random variable M_i counting the number of random space and time spans (see Eq. (9)).

4. Conclusions

In this paper, we have shown how the Cole–Davidson dielectric relaxation response can be rigorously derived using the CTRW concept. Our approach is based on renewal theory and limit theorems for random sums of i.i.d. random jumps, instead of the Fourier–Laplace transform framework commonly used to study CTRWs. The proposed random-variable formalism allows the introduction of a new class of coupled CTRWs, in particular one that leads to the Cole–Davidson function. As a result, the origins of the high- and low-frequency properties of the Cole–Davidson response have been found and the role of the parameter γ has been elucidated. As an illustration to the analysed case, the dielectric permittivity spectra of the four butanediol isomers in the liquid phase have been presented. This empirical evidence essentially concerns a system of OH-dipoles on a molecular backbone that relax in an outer field. We have shown that the Cole–Davidson model provides a good description of the measured data in the frequency range applied, whereas the Debye one does not, as should be expected to some extent due to the interaction of neighbouring relaxing groups.

Comparing the fit parameters for the four butanediol isomers presented in Table I, an evident similarity in the values of the parameters ε_{s} and $\Delta\varepsilon$ for 2,3- and 1,2-butanediol can be seen. These parameters are also similar for 1,3- and 1,4-butanediol. Furthermore, the marked difference in these parameters between the two pairs (2,3-BD and 1,2-BD as opposed to 1,3-BD and 1,4-BD) suggests a connection to the distance between the hydroxyl groups along the chains of these molecules. The values of the parameter γ also seem to reflect this trend.

The assumption of an independent response of each dipole to the external field is, therefore, not valid, and the coupled response is stronger for directly adjacent dipoles. This is also reflected by the random-walk mechanism of the Cole–Davidson response, in which the spatio-temporal coupling (measured by the heavy-tail exponent γ of the coupling variable distribution) is also stronger for the isomers with directly adjacent dipoles.

The trend observed in the fit parameters here can be correlated with other data. For instance, Jesus *et al.*, [35] have calculated the most stable conformations of the butanediol isomers in the gas phase. The weighted averages of the molecular dipole moments over the conformer populations yield a differentiation between isomers with adjacent and separated OH-groups. Moreover, the strengths of both intra- and intermolecular hydrogen bonds in dilute solutions of the four isomers, observed by the same authors, follow the sequence: 2,3-BD $\approx$ 1,2-BD $<$ 1,3-BD $<$ 1,4-BD.

Let us finally emphasise that the high-frequency process is more pronounced in the case of 1,4-butanediol than 2,3-butanediol, *i.e.* in the system where the spatio-temporal coupling is weaker as measured by the heavy-tail exponent γ of the coupling variable distribution (the tails become weaker as $\gamma \to 1$).

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