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14. ABSTRACT

This report results from a contract tasking The Provost, Fellows and Scholars of the College of the Holy and Undivided Trinity of Queen Elizabeth as follows: The contractor will investigate approaches to solve the underlying stochastic Langevin equations developed in previous works.

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U.S. Air Force; contract **EOARD** FA8655-03-01-3027

Dear Dr. Milligan,

I am writing for the purpose of presenting to you a final report of our EOARD project FA8655-03-01-3027 (*ANOMALOUS DIFFUSION AND RELAXATION IN COMPLEX DISORDERED SYSTEMS*) for 2003. The results of this period are contained in 8 papers [1-8] which are available as an attachment to this note.

To be specific, in Refs. [1,2,4], the fractional Klein-Kramers (Fokker-Planck) equation describing the fractal time dynamics of an assembly of fixed axis dipoles rotating in an N -fold cosine potential representing the internal field due to neighboring molecules is solved using matrix continued fractions. The result can be considered as a generalization of the solution for the normal Brownian motion in a cosine periodic potential to fractional dynamics (giving rise to anomalous diffusion) and also represents a generalization of Fröhlich's model of relaxation over a potential barrier. The solution includes both inertial and strong internal field effects, which in combination produce a strong resonance peak (Poley absorption) at high frequencies due to librations of the dipoles in the potential, an anomalous relaxation band at low frequencies mainly arising from overbarrier relaxation and a weaker relaxation band at mid frequencies due to the fast intrawell modes. The high frequency behavior is controlled by the inertia of the dipole, so that the Gordon sum rule for dipolar absorption is satisfied, ensuring a return to optical transparency at very high frequencies. Application of the model to the broadband (0-THz) dielectric loss spectrum of a dilute solution of the probe dipolar molecule CH_2Cl_2 in glassy decalin is demonstrated.

In ref. 3, we show how Fröhlich's model [H. Fröhlich, *Theory of Dielectrics*, 2nd ed. (Oxford University Press, London, 1958)] of relaxation over a potential barrier based on the concept of *normal* diffusion of a dipolar particles in a potential may be generalized to *anomalous* diffusion in disordered energyscapes giving rise to temporally non local behavior. We present exact and approximate solutions of the fractional diffusion equation for an assembly of fixed-axis dipoles in a double-well periodic potential. It is shown that a knowledge of 3 time constants characterizing the *normal* rotational diffusion, viz., the integral relaxation time, the effective relaxation time, and the inverse of the smallest eigenvalue of the Fokker-Planck operator, is sufficient to accurately predict the *anomalous* relaxation behavior for all time scales of interest. Simple equations for the characteristic frequencies of the two bands, which appear in the dielectric loss spectrum, are obtained in terms of the physical model parameters (barrier height and fractional exponent). The model can explain the anomalous

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relaxation of complex dipolar systems where the anomalous exponent σ differs from unity (corresponding to the classical Debye theory of dielectric relaxation), i.e., the relaxation process is characterized by a broad distribution of relaxation times.

In Refs. 5 and 6, the linear dielectric response of an assembly of symmetric top dipole molecules (each of which is free to rotate in space) is evaluated in the context of fractional dynamics. The infinite hierarchy of differential-recurrence relations for the relaxation functions appropriate to the dielectric response is derived by using the underlying inertial fractional Fokker-Planck (fractional Klein-Kramers) equation. On solving this hierarchy in terms of continued fractions (as in normal rotational diffusion), the complex dynamic susceptibility is calculated for typical values of the model parameters for the particular case of linear and symmetric top molecules. It is shown that the model can reproduce nonexponential anomalous dielectric relaxation behaviour at low frequencies ($\omega\tau \leq 1$, where τ is the Debye relaxation time) and the inclusion of inertial effects ensures that optical transparency is regained at very high frequencies (in the far infrared region) so that Gordon's sum rule for integral dipolar absorption is satisfied.

In Ref. 7 and 8, the Cole-Cole behaviour of the complex susceptibility is derived from the diffusion limit of a random walk of permanent dipoles that is the discrete orientation model with a Lévy like distribution of waiting times at the sites. The method yields a generalisation of the Smoluchowski equation of the classical theory of the Brownian motion in a potential which describes the behaviour of the distribution function in configuration space with a fixed waiting time to a system which exhibits chaotic behaviour of the waiting times. It is indicated how the fundamental solution of the Smoluchowski equation in the absence of a potential may be obtained as a Lévy distribution by simply using the properties of the characteristic function which is the Mittag-Leffler function. Such a representation also yields in a simple manner both the mean square displacement of a dipole and the after effect function of the Cole-Cole process.

The results will be presented at the 2004 March Meeting of the American Physical Society in Montreal (three papers have been accepted for oral presentation).

Profs. W. T. Coffey and Y. P. Kalmykov attended the March Meeting of the American Physical Society, which was held in the Austin Convention Center from Monday 2 March to Friday 7 March 2003. During the course of the Conference, Profs. W. T. Coffey and Y. P. Kalmykov held extensive discussions with Dr. T. Bunning (Air Force Research Laboratory/MLP) with a view towards development of scientific collaboration. Secondly, extensive discussions were held with John F. Maguire (Air Force Research Laboratory, Material & Manufacturing Directorate/MLBR, WPAFB, OH) with a view towards possible

applications of our theory in polymer dynamics. Profs. Kalmykov and Coffey also presented material concerning the project at the NATO-ARD conference on non linear dielectric phenomena held in Ustron, Katowice Poland in May 2003. Their contributions are to be published in the Kluwer NATO conference series of volumes. The proceedings of the Royal Irish Academy workshop on diffusion and relaxation in complex systems in September 2002 sponsored by EOARD and dealing with some of the topics covered by this contract are also being published as a special volume of the *Journal of Molecular Liquids* published by Elsevier Amsterdam. EOARD will receive a copy in due course.

Prof. Coffey (Principal Investigator) is devoting all his research time to the project at TCD. Dr. S.V. Titov (IREE RAS) is in Ireland for two 3 month visits per year working full time on the project. All Prof. Kalmykov's research time in Perpignan and about 50% of Prof. D. S. F. Crother's time is being spent on the project. Mr. D. Holland is also preparing his PhD in Belfast on the topic of the project under the joint supervision of Prof. Crothers and Prof. Coffey who as joint supervisor is a Distinguished Visitor Fellow at Queens University Belfast 2002-2005.

List of publications on the project

1. W. T. Coffey, Yu. P. Kalmykov and S. V. Titov, *Anomalous dielectric relaxation in an N-fold cosine potential*, Phys. Rev. E, **67**, 061115 (2003).
2. W. T. Coffey, Yu. P. Kalmykov and S. V. Titov, *Anomalous diffusion and dielectric relaxation in periodic potential*, March Meeting of the APS, Austin 2003, Bull. Am. Phys. Soc., Series II, 2003, V.48, No 1, Part I, p. 442.
3. W. T. Coffey, Yu. P. Kalmykov and S. V. Titov, *Bimodal approximation for anomalous diffusion in a potential*, Phys. Rev. E, **69**, No. 1 (2004).
4. W. T. Coffey, Yu. P. Kalmykov and S. V. Titov, *Anomalous dielectric relaxation in a double-well potential*, J. Mol. Liquids, in press (2003).
5. W. T. Coffey, Yu. P. Kalmykov and S. V. Titov, *Inertial effects in anomalous dielectric relaxation*, J. Mol. Liquids, in press (2004).
6. W. T. Coffey, Yu. P. Kalmykov and S. V. Titov, *Inertial effects in anomalous dielectric relaxation of symmetric top molecules*, Phys. Rev. E, **69**, No. 3 (2004).
7. W. T. Coffey, *Theory of anomalous dielectric relaxation*, J. Mol. Liquids, in press (2004).
8. W. T. Coffey, D.S.F. Crothers, D. Holland, and S. V. Titov, *Anomalous dielectric relaxation in complex systems*, J. Mol. Liquids, in press (2004).

Prof. W. T. Coffey
FAPS, MRIA, Docteur honoris causa (Perpignan)
Department of Electronic & Electrical Engineering
School of Engineering
Trinity College
Dublin 2
Ireland

Tel. +(353)-1-608-1433

FAX +(353)-1-677-2442

e-mail: seccr@ee.tcd.ie or wcoffey@mee.tcd.ie