Generalized Spin Glass Relaxation

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Spin relaxation close to the glass temperature of CuMn and AuFe spin glasses is shown, by neutron spin echo, to follow a generalized exponential function which explicitly introduces hierarchically constrained dynamics and macroscopic interactions. The interaction parameter is directly related to the normalized Tsallis non-extensive entropy parameter, \( q \), and exhibits universal scaling with reduced temperature. At the glass temperature \( q = 5/3 \) corresponding, within Tsallis' \( q \)-statistics, to a mathematically defined critical value for the onset of strong disorder and non-linear dynamics.

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The dynamical correlations associated with the onset of a glassy state have been the focus of considerable interest and controversy over the last four decades, yet despite this attention such dynamics are still poorly understood. Above the glass temperature, \( T_g \), the dynamics are generally characterized by a stretched exponential (Kohlrausch or KWW) relaxation, \( \sim \exp[−(t/τ)^β] \). This form appears to be almost ubiquitous in nature, describing phenomena as diverse as human dynamics \([3]\), the conductivity close to the metal-insulator transition \([4]\), the jamming transition \([5]\), frustrated magnets \([6]\), experimental \([7]\) and theoretical \([8]\) spin glasses, as well as conventional glass forming liquids above the glass temperature \([9, 10]\).

However the stretched exponential form does not adequately describe the relaxation of either structural or magnetic glasses close to \( T_g \) and below, where self-similarity in time (i.e. fractal behaviour in time) occurs. This is demonstrated in Monte Carlo calculations by Ogieski \([8]\), based upon a 3d ±1 Ising spin glass model, which show that close to the spin glass temperature the time dependent spin autocorrelation function should take a phenomenological modified Kohlrausch form which incorporates a power law dependence:

\[
q(t) = \langle S_i(0) . S_i(t) \rangle \propto t^{-x} \exp(-(t/\tau)^β)
\]

where \( β \) is associated with the fractal geometry of the system and \( k(>0) \) is an effective interaction parameter related to the waiting time and providing a measure of the relative contribution of hierarchical relaxation processes. Conveniently, the Werón power law reduces to the Kohlrausch form in the limit \( k \rightarrow 0 \), in which case \( 0 < β \leq 1 \) has precisely the same meaning as before, with the limit \( β \rightarrow 1 \) implying simple Debye (exponential) relaxation.

Phenomenologically, the Werón model can be readily extended to the spin glass transition problem yet, despite its rigor and elegance, this approach has not previously been used in the analysis of spin relaxation. We have therefore investigated the applicability of this generalized relaxation function to spin glasses by analysing neutron
spin echo spectra collected from the archetypal metallic systems Cu$_{1-x}$Mn$_x$ (x=0.1, 0.16, 0.35) and by revisiting our previously measured NSE spectra from Au$_{1-x}$Fe$_x$ (x=0.14)\cite{11}. We have also explored the interconnections between the probabilistic Weron model and a more general approach based on non-extensive thermodynamics introduced by Tsallis \cite{15} to describe highly disordered systems governed by Lévy-stable distributions.

Cu$_{1-x}$Mn$_x$ samples with x=0.1, 0.16 and 0.35 were prepared by argon arc-melting and subsequent cold-rolling. The resulting disks were homogenised at 900°C before being quenched directly into water. The samples were found to have glass temperatures, T$_g$, of 45K, 74K and 153K respectively. The Cu$_{1-x}$Mn$_x$ NSE spectra were collected using the IN11C spectrometer \cite{16} at the Institut Laue Langevin (Grenoble) at $Q = 4$$\text{nm}^{-1}$ with an incoming wavelength of 0.55nm. Our previously reported spectra from Au$_{0.86}$Fe$_{0.14}$ ($T_g$=41K) were obtained on IN15 at $Q = 0.4$$\text{nm}^{-1}$ and a wavelength of 0.8nm \cite{11}.

All NSE spectra were independent of scattering vector, Q. Characteristic results are shown in figures 1 and 2. In figure 3 we show a direct comparison of least squares fits of the Kohlrausch stretched exponential, Ogielski (eq 1) and Weron (eq 2) functions to the relaxation of Au$_{0.86}$Fe$_{0.14}$ at 45.7 K. Whilst both of the latter functions afford equally acceptable descriptions over the Fourier time range covered by the data, the Ogielski function must be modified to avoid an unphysical increase to values greater than unity at short times \cite{17}. In comparison, the Weron function is able to describe all spectra, from the high temperature simple exponential limit to the complex power law decay found below T$_g$, with consistent and physically meaningful parameters.

The spectra of Au$_{0.86}$Fe$_{0.14}$ give a temperature dependent $\beta$, decreasing from 1 at high temperatures to approximately 0.3 at the glass temperature, which implies a continuing evolution of the geometric and dynamical fractal character of the spin clusters. The CuMn spectra cover a smaller dynamic range, and equally valid fits can be obtained with a temperature independent $\beta \sim 0.5$ or with $\beta$ varying with temperature as found for AuFe. However, the most informative parameter is the interaction parameter, $k$, which diverges as the spin glass temperature is approached. We suggest that $k$ provides an interesting and novel insight into the underlying thermodynamics which drive the non-exponential spin glass relaxation, and closely links such relaxation to that observed in other disordered systems.

A physical interpretation of the Weron function is provided by Tsallis’ generalisation of Boltzman-Gibbs thermostatistics for complex and multifractal systems \cite{13}. In this context Tsallis introduces the concept of non-extensive entropy, proposing that the total entropy of any self-organising, strongly interacting systems may be either greater or less than the sum of the entropies of the individual components of the system.

\[
\frac{S_q(A + B)}{k_B} = \frac{S_q(A)}{k_B} + \frac{S_q(B)}{k_B} + (1 - q) \frac{S_q(A)}{k_B} \frac{S_q(B)}{k_B} \quad (3)
\]

where $q$ is the so-called non-extensivity parameter. For

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{Temperature dependence of the NSE spectra of (a) Cu$_{0.9}$Mn$_{0.10}$ (b) Cu$_{0.84}$Mn$_{0.16}$ and (c) Cu$_{0.65}$Mn$_{0.35}$ at Q = 4nm$^{-1}$. In each case the solid lines represent fits of the Weron function, eq(2), to the data.}
\end{figure}
Here the relaxation time, $\tau$, scales with the characteristic volume $v$ of the relaxing clusters according to $\tau = v^{1/\beta}$ and $0 < \beta \leq 1$ is related to the fractal geometry of the system, as in the Weron analysis. By maximising the Tsallis non-extensive entropy they obtain a (normalised) cluster size distribution function

$$f(v) = \left[1 - v^{1 - q}/(2 - q)\right]^{-\frac{1}{\beta}}$$

where $1 \leq q < 2$. This distribution function belongs to the family of Lévy-stable distributions characterized by asymptotic power law tails $^{[13, 20]}$ and the non-extensivity parameter $q$ quantifies the statistics of the system. An analysis of eq. (6) shows that the moments are defined only for $1 < q < 3/2$, whereas for $q \geq 3/2$ all moments, including the mean cluster size, diverge. However, within the modified $q$-statistics introduced by Tsallis finite moments are obtained only for $q < 5/3$ $^{[21]}$. The critical value $q=5/3$ marks the transition to the limit of strong disorder, where most statistical weight is in the wings of the distribution and the macroscopic behavior is governed by a high number of highly improbable collective events.

The relaxation function deduced from the cluster size distribution function takes a form identical to that proposed by Weron, subject to the simple substitutions $k = (q - 1)/(2 - q)$

$$\varphi(t) = \left[1 + \left(q - 1 \over 2 - q\right) \left(\frac{t}{\tau}\right)^{\beta}\right]^{-\frac{1}{\beta}}$$

The values of the interaction parameter, $k$, obtained from fits of the Weron function of eq. 2 to the AuFe and CuMn NSE spectra can thus be related directly to the sub-extensivity parameter, $q$.

As shown in figure 3, the resulting $q$-values for the all the spin glass alloys discussed here, together with those obtained from a re-analysis of the earlier published NSE measurements $^{[22]}$ on the more dilute Cu$_{90.95}$Mn$_{90.05}$, exhibit the same scaling with reduced temperature, $(T/T_g)$. At first sight this is surprising as the fractal character of dilute and concentrated CuMn spin glasses and of AuFe is expected to be quite different: Although an oscillatory RKKY magnetic exchange dominates the interactions in both systems, in AuFe the Fe atoms are known to cluster, whilst in CuMn the Mn atoms anticluster. Correspondingly ferromagnetic percolation is found at 15at%Fe in AuFe, but incipient antiferromagnetism is not evident in CuMn until 70at%Mn $^{[23]}$. It is a remarkable property of phase transitions that critical phenomena do not depend on microscopic details but on the topology (i.e. interactions and dimensionality) of the systems.

At high temperatures $q$ is close to 1, indicating an essentially independent, parallel, relaxation of cluster moments. $q$ increases continuously with decreasing temperature and the first limit at $q = 3/2$ is reached at $\sim 1.2$
From the fits of eq(2) to NSE spectra of the Au and CuMn samples, as function of reduced temperature. The values of $q$ for the CuMn sample have been obtained by fitting the previously published data of ref [22].

Significantly, this evolution appears to be universal, at least for the fundamentally different AuFe and CuMn spin glasses discussed here suggesting that spin glasses are very similar to many other complex disordered systems such as financial markets, earthquakes, turbulence or jamming which are governed by self-similarity and the underlining Lévy stable distributions.

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1. R. Kohlrausch, Ann. Phys. Lpz. 12, 393 (1847).
2. J. C. Phillips, Rep. Prog. Phys. 59, 1133 (1996).
3. T. Nakamura, K. Kiyono, K. Yoshiuchi, R. Nakahara, Z. R. Struzik, and Y. Yamamoto, Phys. Rev. Lett. 99, 138103 (2007).
4. J. Jaroszyski and D. Popović, Phys. Rev. Lett. 96, 037403 (2006).
5. P. Chaudhuri, L. Berthier, and W. Kob, Phys. Rev. Lett. 99, 060604 (2007).
6. H. Mútka, G. Ehlers, C. Payen, D. Bono, J. R. Stewart, P. Fouquet, P. Mendels, J. Y. Mevellec, N. Blanchard, and G. Collin, Phys. Rev. Lett. 97, 047203 (2006).
7. I. A. Campbell, A. Schenck, D. Herlach, F. N. Gygax, A. Amato, R. Cywinski, and S. H. Kilcoyne, Phys. Rev. Lett. 72, 1291 (1994).
8. A. Ogilvieski, Phys. Rev. B. 32, 7384 (1985).
9. F. Mezei, W. Knaak, and B. Farago, Phys. Rev. Lett. 58, 571 (1987).
10. D. Richter, R. Zorn, B. Farago, B. Frick, and L. J. Betters, Phys. Rev. Lett. 68, 71 (1992).
11. C. Pappas, F. Mezei, G. Ehlers, P. Manuel, and I. A. Campbell, Phys. Rev. B. 68, 054431 (2003).
12. R. G. Palmer, D. L. Stein, E. Abrahams, and P. W. Anderson, Phys. Rev. Lett. 53, 958 (1984).
13. K. Weron, J. Phys. C: Solid State Phys. 3, 9151 (1991).
14. L. A. Dissado and R. M. Hill, J. Appl. Phys. 66, 2511 (1989).
15. C. Tsallis, S. V. F. Levy, A. M. C. Souza, and R. Maynard, Phys. Rev. Lett. 75, 3589 (1995).
16. B. Farago, Physica B 241, 113 (1997).
17. A. Keren, J. S. Gardner, G. Ehlers, A. Fukaya, E. Segal, and Y. J. Uemura, Phys. Rev. Lett. 92, 107204 (2004).
18. F. Bouzou and O. Sotolongo-Costa, Europhysics Letters 62, 808 (2003).
19. J. P. Nolan, Stable Distributions - Models for Heavy Tailed Data (Birkhäuser, 2009), in progress, Chapter 1 online at academic2.american.edu/~jpnolan.
20. O. E. Barndorff-Nielsen, T. M. Sidney, and I. Resnick, Lévy Processes - Theory and applications (Birkhäuser, 2001).
21. D. Prato and C. Tsallis, Phys. Rev. E. 60, 2398 (1999).
22. F. Mezei and A. P. Murani, J. Magn. Magn. Mater. 14, 211 (1979).
23. J. A. Mydosh, Spin Glasses: An Experimental Introduction (Taylor & Francis, London, 1993).