Fluctuation-Dissipation relation in a spin glass in the non-stationary regime

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Abstract

We present the first experimental determination of the time autocorrelation $C(t',t)$ of magnetization in the non-stationary regime of a spin glass. Quantitative comparison with the corresponding response, the magnetic susceptibility $\chi(t',t)$, is made possible by the use of a new experimental setup allowing both measurements in the same conditions. Clearly, we observe a non-linear fluctuation-dissipation relation between C and χ, depending weakly on the waiting time t'. Following theoretical developments on mean-field models, and lately on short range ones, it is predicted that in the limit of long times, the $\chi(C)$ relationship should become independent on t'. A scaling procedure allows us to extrapolate to the limit of long waiting times.

Key words: spin glass; non-stationarity; fluctuation-dissipation relation;

1. Introduction

The fluctuation dissipation theorem (FDT) [1,2] which links the response function of a system to its time autocorrelation function, is from more than fifty years ago one of the masterpieces of statistical physics. Nevertheless, FDT applies only to ergodic systems at equilibrium. Yet, such systems represent a very limited part of natural objects, and there is now a growing interest on non-ergodic systems and on the related challenging problem of the existence of fluctuation dissipation (FD) relations valid in off-equilibrium situations.

In non-stationary systems, FDT is not expected to hold. A quite general FD relation can be written as [3,4] $R(t',t) = \beta X(t',t)\partial C(t',t)/\partial t'$, where $R(t',t)$ is the impulse response of an observable to its conjugate field, $C(t',t)$ the autocorrelation function of the observable and $\beta = 1/k_BT$. FDT corresponds to $X = 1$. Determination of X, the fluctuation-dissipation ratio (FDR), or of an "effective temperature", $T_{eff} = T/X$, is the aim of many recent theoretical studies which predicted a generalization of FDT [5,3,4] in "weak ergodicity breaking" systems [6].

2. Experimental

We have investigated the FD relation in the insulating spin glass $CdCr_{1.7}In_{0.3}S_4$ [7], an already very well known compound, with $T_g = 16.7K$. Above T_g, the susceptibility follows a Curie-Weiss law $\chi = \mathcal{C}/(T-\Theta)$ where $\mathcal{C}$ corresponds to ferromagnetic clusters of about 50 spins, and $\Theta \approx -9K$ [8]. The sample is a powder with grain sizes around $10\,\mu m$, embedded in silicon grease to insure good thermal contact between grains, and compacted into a coil foil cylindrical sample holder $5\,mm$ wide and $40\,mm$ long. The two times dependence of the magnetic relaxation (TRM) of this compound was extensively studied [9].

In principle, SQUID measurement of magnetic fluctuations is very simple [10,11]. The difficulty lies in the extreme weakness of the thermodynamic fluctuations (of the order of the response to a field about

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$10^{-7}G$ in our case). Our system, where the pick-up (PU) coil is a third order gradiometer, allows time analysis of the magnetic fluctuations signal over up to 2000 s of sample fluctuations with more than 20 dB of signal/noise ratio. Moreover, in the non-stationary regime, the time autocorrelation of magnetic fluctuations $C(t',t) = \frac{1}{N}\sum_i \langle \mathbf{m}_i(t')\mathbf{m}_i(t)\rangle$, where $\mathbf{m}_i$ is the elementary moment at site i, must be determined as an ensemble average over a large number of records of the fluctuation signal, each one initiated by a quench from above T_g ("birth" of the system). And finally, we want to compare *quantitatively* correlation and relaxation data. If the relaxation function $\sigma(t',t) = \frac{1}{N}\sum_i \langle \mathbf{m}_i(t)\rangle / \mathbf{H}_i$ is measured using a classical magnetometer with homogeneous field, quantitative comparison between C and σ is almost impossible due to the strong discrepancy between the coupling factors in both experiments. Therefore, we have developed a new bridge setup depicted in Fig.1a, allowing measurements of both fluctuations and response. The PU coil of self inductance L_0 is connected to the input coil of a SQUID, of self inductance L_S. The whole circuit is superconducting. Relaxation measurements use a small coil l inserted in the pick-up circuit, and coupled inductively with mutual inductance M to an excitation winding. A current I_0 injected in the excitation results in a field induced by the PU coil itself ($\leq 1mG$ here, clearly in the linear regime though inhomogeneous), and the sample response is measured by the SQUID. To get rid of the term L_0, the sample branch is balanced by a similar one without sample, excited oppositely (see Fig.1a). Detailed analysis of the system will be published elsewere. The main features are as follows.

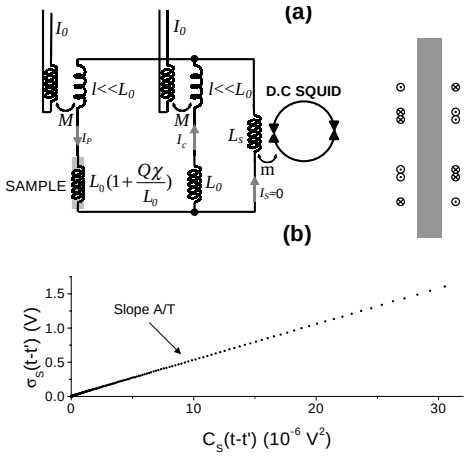

Fig. 1. a) Schematic of the detection circuit. The pick-up coil (right side), containing the cylindrical sample, is a third order gradiometer made of +3 -6 +6 -3 turns. b) Calibration is obtained by measuring relaxation versus correlation in a high conductivity copper sample at equilibrium at $4.2K$.

As the fluctuations of elementary moments in the sample are homogeneous and spatially uncorrelated at the scale of the PU, the SQUID output voltage autocorrelation is given by:

$$C_S(t',t) = \langle V_S(t')V_S(t)\rangle = C(t',t)Q\frac{G^2}{(L_0+2L_S)^2} \quad (1)$$

where G is the gain of the SQUID ($V_S = GI_S$). $Q = \sum_i \mathbf{h}^2(\mathbf{r}_i)$ where the index i refers to a moment site, is the coupling factor to the PU, including demagnetizing field effects since $\mathbf{h}$ is the internal field.

The elementary moment response at site i is $R_i(t',t) = \partial\langle \mathbf{m}_i(t)\rangle/\partial \mathbf{h}(\mathbf{r}_i,t')$. Taking into account that the medium is homogeneous, the relaxation function of the SQUID output voltage is given by

$$\sigma_S(t',t) = \frac{V_S(t',t)}{I_0} = \sigma(t',t)Q\frac{MG}{L_0(L_0+2L_S)}. \quad (2)$$

Thus, the coupling factor Q disappears in the relation between C_S and σ_S, independently on the nature and shape of the sample. There remains only the inductance terms M, L_0 and L_S. These being difficult to determine with enough accuracy, absolute calibration was performed using a copper sample of high conductivity, by measuring $\sigma_S(t',t)$ and $C_S(t',t)$ — computed by standard FFT algorithm — at $4.2K$ (^{4}He boiling temperature at normal pressure): with this ergodic material, the relation between both measured quantities is linear with slope A/T, where A is the ***sample independent*** calibration factor (see Fig.1b). From the knowledge of A, determined at $4.2K$, the system is equivalent to a thermometer, i.e. the FDT slope is known ***exactly*** at any temperature.

In the spin glass sample, $C_S(t',t)$ and $\sigma_S(t',t)$ were measured at $T = 0.6T_g$, $0.8T_g$ and $0.9T_g$ after quench from a temperature $T \approx 1.2T_g$. To get a precise definition of the "birth" time, a minimum value of 100 s was chosen for t'. The autocorrelation was determined from an ensemble of 320 records of up to 12000 s of the fluctuation signal. The ensemble averages were computed in each record from the signal at t', averaged over $\delta t' \leq t'/20$, and the one at t, averaged over $\delta t \leq (t-t')/10$ — the best compromise allowing a good average convergence still being compatible with the non-stationarity — , and averaging over all records. As there is an arbitrary offset in the SQUID signal, the connected correlation was computed. Nevertheless, this was not enough to suppress the effect of spurious fluctuation modes of period much longer than 2000 s, giving a non-zero average offset on the correlation results. Thus, as a first step, we have plotted all correlation data, taking as ***the*** origin the value of $\langle V_S^2(t')\rangle$. Due to the elementary measurement time constant this last term corresponds to an average over $t-t'$ about 10^{-2}s, i.e. a range of $(t-t')/t'$ corresponding to stationary regime. Thus,

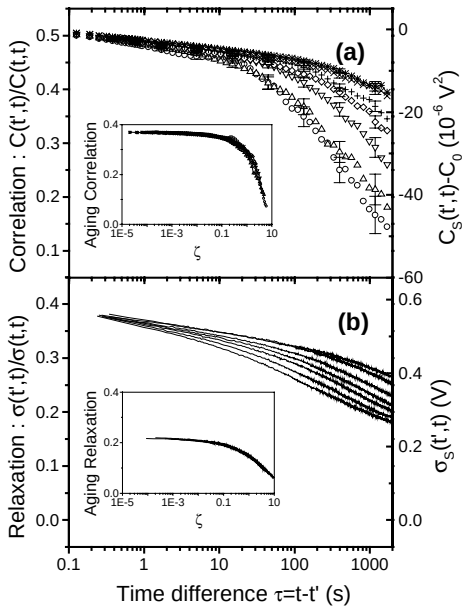

Fig. 2. Aging and scaling of (a) correlation (b) relaxation at $T = 0.8T_g$. Both are measured for waiting times $t' = 100$, 200, 500, 1000, 2000, 5000 and 10000 seconds from bottom to top. Reported error-bars on correlation have a length of two standard-deviation, corresponding to averages over records. In insets, scaling of the aging parts versus $\zeta = (t^{1-\mu} - t'^{1-\mu})/(1-\mu)$, using $\mu = 0.87$. The stationary parts are found to obey a power-law decrease with an exponent $\alpha = 0.05$.

all C_S data are shifted by a common offset C_0. The result at $0.8T_g$ is shown in Fig.2a (right sided scale), as a function of $t - t'$ for values of t' from 100 s to 10000 s. Residual oscillations —and large error bars— for $t' = 100s$ reveal the limit of efficiency of our averaging procedure. Corresponding relaxation data are plotted on Fig.2b. In both results, one can see that the curves merge at low $t - t'$, meaning that they do not depend on t' (stationary regime). At $t - t' \geq t'$, they strongly depend on t', the slower decay corresponding to the longer t'.

Since zero of correlation is unreachable in experimental time, correction of the offset could be obtained from the knowledge of $C(t,t)$. Nevertheless, due to clustering, $C(t,t)$ depends on temperature and cannot be determined from the high temperature susceptibility. In canonical compounds like 1% Cu:Mn [12], with negligible clustering, the field-cooled susceptibility is temperature independent in agreement with the PaT hypothesis [14,15], yielding $C(t,t) = T_g \chi_{FC}(T)$. We used a generalization of this relation with the condition that a smooth dependence of $C(t,t;T)/T$ must result [16]. This was obtained by using for T_g a slightly different value, $T_g^* = 17.2K$. Then, from the value of the calibration factor A, and writing $C(t,t;T) = 17.2\chi_{FC}(T)$, $C_S(t,t;T)$ can be determined, and suppression of the offset can be performed by using the $\chi(C)$ plot, first introduced by Cugliandolo and Kurchan [3]. We plot the normalized susceptibility function $\widetilde{\chi}(t',t) = 1 - \widetilde{\sigma}(t',t)$ where $\widetilde{\sigma}(t',t) = \sigma_S(t',t)/\sigma_S(t,t)$ (note that $\sigma(t,t) = \chi_{FC}$) versus normalized autocorrelation $\widetilde{C}(t',t) - \widetilde{C}_0 = (C_S(t',t) - C_0)/C_S(t,t;T)$ for all experimental values of t'. In this graph, the FDT line has slope $-T_g^*/T$ and crosses the $\widetilde{C}$ axis at $\widetilde{C} = 1$. On the data, a clear linear range appears at large $\widetilde{C}$ (small $t - t'$), with slope corresponding to the calculated FDT slope with less than 3% error. Suppression of the correlation offset is then obtained by horizontal shift of the data. This adjustment is of course based on a rough ansatz on $C(t',t;T)$ which needs further justifications, but we stress that the induced uncertainty concerns only the position of the zero on the $\widetilde{C}$ axis, and not the shape and slope of the curves. The result at the three investigated temperatures is shown in Fig.3. With decreasing $\widetilde{C}$ (increasing $t - t' \geq t'$), the data points depart from the FDT lines. Despite the scatter of the results, a tendency for the data at small t' to depart the FDT lines at larger values of $\widetilde{C}$ is clear.

3. Discussion

In the asymptotic limit of large times, in the SK model, the FDR should depend on time only through the correlation function: $X(t',t) = X(C(t',t))$ for t' (and $t > t'$) $\to \infty$. The dependence of X on C would reflect the level of thermalization of different degrees of freedom within different timescales [4]. Thus, the integrated forms of the FD relation would become $\chi(t',t) = \beta \int_{C(t',t)}^{C(t,t)} X(C)dC$ (susceptibility function) and $\sigma(t',t) = \beta \int_0^{C(t',t)} X(C)dC$ (relaxation function). They would depend on t and t' only through the value of C. Analytical [13] and numerical [14] investigations confirm the above properties in short range models.

Experimentally, our results show that it is impossible to fulfill the condition of timescales separation underlying the theory. The left sided scales in Fig.2a and b correspond to $\widetilde{C}(t',t)$ and $\widetilde{\sigma}(t',t)$ respectively, at $0.8T_g$. In former works, it was shown that the whole relaxation curves could be scaled as the sum of two contributions, one stationary and one non-stationary [9]

$$\widetilde{\sigma}(t',t) = (1-\Delta)(1 + (t-t')/t_0)^{-\alpha} + \Delta\varphi(\zeta), \qquad (3)$$

where t_0 is an elementary time of order 10^{-11} s, φ is a scaling function of an effective time parameter $\zeta \propto t^{1-\mu} - t'^{1-\mu}$ depending on the sub-aging coefficient $\mu < 1$ [9], and α can be determined with good precision from the stationary power spectrum of fluctuations $S(\omega) \propto \omega^{\alpha-1}$. The inset in Fig.2b displays the result of the scaling on the relaxation curves with $\alpha = 0.05$, $\Delta =$

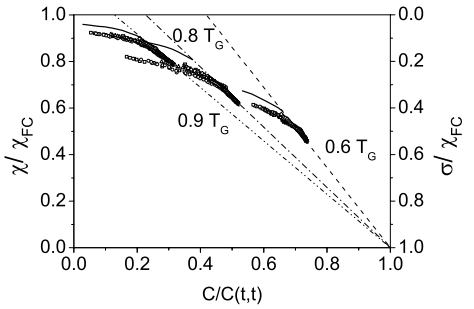

Fig. 3. FD-plot. Relaxation functions are plotted versus correlation functions at each temperature for each t'. The lines (FDT lines) are calculated from the calibration obtained with the copper sample. The full lines represent the scaling extrapolation for $t' \to \infty$. The branching points with the FDT lines, correspond to $\widetilde{C} = q_{EA}$.

0.21 and $\mu = 0.87$. As shown in the inset of Fig.2a, the scaling works rather well on the autocorrelation curves with the same exponents, but now, q_{EA}, the Edwards Anderson order parameter, replaces Δ. We get $q_{EA} = 0.37$. These results show clearly that the stationary part of the dynamics is still important yet in the aging regime, i.e. that the limit of long t' is not reached within the timescale of our experiments (in fact, timescale separation is realized if $t' \geq \tau$ where τ is the observation time such that $C_{stat}(\tau) \ll q_{EA}$). Even if the long t' limit for $\widetilde{\chi}(\widetilde{C})$ does exist, it is not reached in the plot of data in Fig.3 and a t' dependence of the $\widetilde{\chi}(\widetilde{C})$ curves is expected.

Nevertheless, if granted, the scaling gives the long time limit of the non-stationary part of the dynamics, allowing a plot of the long times asymptotic non-stationary part of the $\widetilde{\chi}(\widetilde{C})$ curve. Of course, here we verify it only over 2 decades of time, up to $t' = 10000\,s$, but it was proven to be relevant on TRM up to $t' = 100000\,s$ [7]. The full lines in Fig.3 are obtained by plotting the smoothed curves of aging parts of $\widetilde{\chi}(\zeta)$ versus $\widetilde{C}(\zeta)$ at the three temperatures investigated. According to theoretical conjectures, $d\widetilde{\chi}(\widetilde{C})/d\widetilde{C}$ would represent the static quantity $x(q)$ [13]. One can see that the curves does not align perfectly, though they follow roughly a law like $\widetilde{\chi} = (1 - \widetilde{C})^B$ with $B \simeq 0.5$ corresponding astonishingly to the mean field prediction. Nevertheless, this result in based on the validity of our time scaling at all timescales, an hypothesis whose theoretical justifications are still lacking.

4. conclusions

In conclusion, we have presented the first experimental determination of the non stationary time autocorrelation of magnetization in a spin glass, an archetype of a complex system. With the help of the time scaling properties of both the relaxation and the autocorrelation, we were able to propose a first experimental approach of a possible generalization of FDT to non-stationary systems.

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