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Observation of $1/f$ magnetic fluctuations in a spin glass

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Résumé. — Nous observons les fluctuations spontanées de l'aimantation à 4,2 K en champ nul dans le verre de spin amorphe $(\text{Al}_2\text{O}_3)_{0,1}(\text{MnO})_{0,5}(\text{SiO}_2)_{0,4}$ et dans le verre de spin cristallin CsNiFeF_6 . Dans les deux cas la densité spectrale du bruit montre une dépendance en $1/f$ dans la gamme de fréquences 10^{-2} Hz- 10^2 Hz. L'amplitude du bruit en $1/f$ est en bon accord avec la valeur mesurée de la susceptibilité hors phase. Ces observations vont à l'appui de l'applicabilité du théorème de fluctuation-dissipation aux verres de spins et révèlent le bruit magnétique comme un nouveau domaine d'investigation.

Abstract. — We observe spontaneous magnetic fluctuations at 4.2 K in zero field for the amorphous spin glass $(\text{Al}_2\text{O}_3)_{0,1}(\text{MnO})_{0,5}(\text{SiO}_2)_{0,4}$ and for the crystalline spin glass compound CsNiFeF_6 . In both cases the noise power spectrum exhibits an $1/f$ dependence in the range 10^{-2} Hz to 10^2 Hz. The $1/f$ noise amplitude is in good agreement with the measured value of the out of phase susceptibility. This observation supports the applicability of the fluctuation dissipation theorem to spin glasses and suggests the magnetic noise as a new area of investigation.

Although the observation of electrical noise due to conduction electrons (by the voltage fluctuations across a resistor) is a very old one [1] the corresponding phenomenon of spontaneous fluctuations of the magnetization in a magnetic material at equilibrium has scarcely been studied directly [2]. The difficulty of detecting thermally excited magnetic fluctuations at low frequencies can be appreciated from this simple example : if a coil of inductance L is completely filled with a material of susceptibility $\chi = \chi' - i\chi''$ at temperature T , its impedance at a frequency $\omega/2\pi$ becomes $Z = i\omega L(1 + 4\pi\chi)$. The out of phase susceptibility $\chi''(\omega)$ gives rise to a resistive term $r = 4\pi\chi'' L\omega$ which, by the Nyquist theorem, produces the mean fluctuation voltage $\overline{V}^2 = 4kTr\Delta F$ in a frequency range $\Delta\omega/2\pi$. In order to evaluate r we take a simple model to relate $\chi''(\omega)$ to the static susceptibility $\chi(0)$ by $\chi''(\omega) = \chi(0)\omega\tau/1 + \omega^2\tau^2$ where τ is a relaxation time. Taking $\tau = 10^{-12}$ s, $\chi(0) = 0.1$, $L = 1\mu\text{H}$, and $\omega/2\pi = 1$ Hz, we find $r = 10^{-18}\Omega$, which readily explains why the magnetic noise is so much more difficult to detect at low frequencies than the corresponding electrical one. Our comparison is, however, not strictly honest : since the magnetic fluctuations originate microscopically in the random motion of

magnetic dipole moments, comparison should be made with the dielectric fluctuations of an assembly of electrical dipoles rather than with those of free electrical charges. A detailed analysis [3] shows that the ratio of the amplitudes of the magnetic energy fluctuations to the dielectric ones is equal to the square of the ratio of the Bohr magneton to the Debye dipole unit, which, in turn, is equal to the square of the fine structure constant $\alpha = (137)^{-1}$. Furthermore it is obvious in our example that another unfavourable factor is that the noise power available in the frequency spectrum of $\chi''(\omega)$ extends up to τ^{-1} and is proportional to τ at low frequency. These elementary remarks show that direct observation of magnetic fluctuations at low frequencies in concentrated paramagnetic systems or ordered magnetic materials, for which the relaxation times are always very short, is quite hopeless [4]. However a special case arises with spin glass-like phases which are well known to display a wide spectrum of relaxation times, extending to very low frequencies below the spin glass temperature.

In this letter we report the direct observation at 4.2 K of magnetic fluctuations at equilibrium for the amorphous system $(\text{Al}_2\text{O}_3)_{0.1}(\text{MnO})_{0.5}(\text{SiO}_2)_{0.4}$, which is a spin glass below 5.3 K [5], and for the compound CsNiFeF_6 . This compound is a crystalline insulator belonging to the modified pyrochlore family. The crystal structure is fcc but the magnetic ions Ni^{2+} and Fe^{3+} are distributed at random on a network of corner-sharing tetrahedra with near neighbour anti-ferromagnetic interactions : this structure provides both disorder and frustration among the spins [6]. dc susceptibility (field cooled and zero field cooled) [7], specific heat [8] and ac susceptibility measurements from 20 Hz to 50 kHz lead to a consistent description of the low temperature magnetic state (below 5.4 K) as a spin glass phase close to the ferromagnetic limit [9].

The technique used is essentially that of a high sensitivity susceptibility measurement using a pickup coil coupled by a superconducting flux transformer to an r.f. SQUID detector [10]. In order to minimize the noise from external magnetic field sources, a geometry of third order cylindrical gradiometer was adopted for the pickup coil. This non-conventional geometry raises the question of the relation between the fluctuations of the sample magnetization M and the detected variation of the magnetic flux ϕ in the gradiometer coil. A general analysis of this relation has been done [11], based on the reciprocity theorem. Two simple limits exist depending on whether the length scale over which the fluctuations occur is large or small compared to the characteristic size of the gradiometer coils : in the case of long range spatial correlation (i.e. a single domain) the gradiometer geometry effectively cancels most of the magnetic flux ; on the contrary, local magnetic fluctuations (as in a powder) give rise to spatially uncorrelated magnetic fluxes whose spectral density at ω is detected according to :

$$(\phi^2)_\omega = (M^2)_\omega V \int_V h^2(\mathbf{r}) d\mathbf{r}^3 \quad (1)$$

where V stands for the volume occupied by the sample and $h(\mathbf{r})$ is the field induced by the coil when carrying a unit of current at frequency $\omega/2\pi$. This relation shows the essential difference between a magnetic noise measurement and a magnetization measurement for which case the same quantities are related by :

$$\phi = M \int_V h(\mathbf{r}) d\mathbf{r}^3. \quad (2)$$

The measurements are performed at 4.2 K in a He⁴ dewar. Two mumetal shields and a superconducting lead shield screen the experimental space against stray magnetic field [12]. The third order gradiometer coil is wound on a 7.2 mm diameter with basic spacing 5 mm. The measured self inductance is $1.3 \pm 0.2 \mu\text{H}$. The coupling coil of the r.f. SQUID is connected to the gradiometer coil by a pair of twisted Nb wires : this ensemble forms a superconducting flux transformer of transfer ratio $(200 \pm 40)^{-1}$. The output voltage of the SQUID electronics is transferred to

a 400 channel digital spectrum analyser operating in the ranges 0-1 Hz to 0-100 kHz. The position of the sample can be manually shifted away from the pickup coil in order to test the « empty coil » situation. The intrinsic noise of a SQUID system being strongly sensitive to the experimental conditions, a record of the noise spectrum with the coil empty is first performed, then the sample is inserted into the coil and the noise spectrum is again recorded.

The difference between the spectra of $(\phi^2)_\omega/L_0$, where L_0 is the pick up self inductance, with the sample in and out of the coil, is taken as the intrinsic noise spectrum of the sample. As can be seen in figure 1a the noise of the empty system is flat above 1 Hz at a level of 3×10^{-20} erg/Hz and has an approximate $1/f^2$ contribution below 1 Hz [10]. Due to the subtraction procedure, the limiting sensitivity is obtained when the intrinsic noise of the sample is equivalent to that of the empty system.

Contrary to the case of the electrical noise, no standard « Johnson » [1] magnetic noise source exists. In order to test the ability of the system to detect magnetic fluctuations of known amplitude we have chosen to measure the random flux associated with the current fluctuations in a pure copper cylinder having the same geometry as the sample holder. The observed noise spectrum at 4.2 K is flat below 1 Hz at 5.9×10^{-16} erg/Hz (dashed line in Fig. 1a) and decreases

Fig. 1a. — Noise power spectrum of the empty system (lower curve) at 4.2 K. The plateau above 1 Hz corresponds to the electronic noise of the SQUID system ($1.5 \times 10^{-4} \phi_0$ rms/ $\sqrt{\text{Hz}}$). The upper set of points is the intrinsic noise power spectrum of CsNiFeF₆ at 4.2 K (i.e. after subtracting out the empty system noise). The points (■ and □) at frequencies 1 Hz correspond to waiting times of 1 hour and 28 hours after initial cooling, respectively. Below 1 Hz, the points (●) correspond to waiting times $54 \text{ hours} \leq t \leq 145 \text{ hours}$, after initial cooling. The dashed line is a smoothed average of the eddy currents noise from a pure copper cylinder sample at 4.2 K, for calibration purposes.

Fig. 1b. — Calculated values of $\chi''(\omega)$ from figure 1a for CsNiFeF₆, using the fluctuation dissipation theorem (Eq. (3)). For comparison, measured values of $\chi''(\omega)$ at higher frequencies (□) [14] are reported. The dashed parallel lines show our estimate of the absolute calibration error.

like $1/f^{3/2}$ above 1 Hz. This is well accounted for by a change of regime of the eddy currents out of phase susceptibility, depending on whether the skin depth is larger or smaller than the cylinder radius [13]. The calculated value of the noise level at low frequencies agrees within 30 % with our measured value. This ascertains the validity of quantitative noise determination with our set up.

The CsNiFeF_6 powder is packed into a cylindrical sample holder (31.5 mm $\times$ 6.5 mm diam). The noise spectrum of this sample at 4.2 K is plotted in erg/Hz *versus* frequency in figure 1a in the range 10^{-2} Hz- 10^2 Hz for different times after the initial cooldown of the system (the minimal time is about 1 hour). One sees that above 1 Hz an $1/f^n$ dependence is obtained with $n = 1 \pm 0.04$ for the noise power, independent of the time of measurement after cooling. A slight dependence of the spectrum appears as a function of the waiting time t_w below 0.5 Hz : $n \sim 1.5$ for $t_w \leq 27$ hours after cooldown. For $t_w \geq 54$ hours the $1/f$ dependence is recovered in the whole range of frequencies investigated. Apart from this effect (which needs further investigation) the general $1/f$ character of the noise spectrum is well established, since the CsNiFeF_6 magnetic noise level is well above that of the empty system, except above 200 Hz where both contributions are equal. The measured $1/f$ noise amplitude is $8 \pm 1 \times 10^{-18}$ erg/Hz at 1 Hz.

Application of the fluctuation dissipation theorem to the magnetization yields for the flux the relation :

$$(\phi^2)_\omega = 16 \pi q_0 L_0 kT \chi''(\omega)/\omega . \quad (3)$$

We have introduced here a filling factor q_0 . Taking into account that $L_0 = (4 \pi)^{-1} \int_\infty h^2(\mathbf{r}) d\mathbf{r}^3$, q_0 is given by

$$q_0 = \frac{\int_V h^2(\mathbf{r}) d\mathbf{r}^3}{\int_\infty h^2(\mathbf{r}) d\mathbf{r}^3} .$$

The magnetic noise is estimated as follows : the susceptibility χ'' at 19 Hz is measured [14] to be 4×10^{-3} emu (very weakly dependent on frequency). The quantity q_0 is calculated [11] to be 0.163 ± 0.045 . By equation (3) one deduces that, at 1 Hz frequency, $(\phi^2)_{1\text{Hz}}/L_0 = 1.8 \pm 1 \times 10^{-17}$ erg/Hz where the uncertainty is mainly due to the evaluation of q_0 and of the transformer ratio. The agreement within a factor 2 between the calculated and measured values of the magnetic noise power of CsNiFeF_6 means that we are indeed observing the fluctuations associated with the imaginary component of the spin susceptibility. In figure 1b we have used equation (3) in order to calculate $\chi''(\omega)$ from the measured value for $t_w \geq 54$ hours : this allows a direct comparison with the $\chi''(\omega)$ measurements made at 19 Hz, 112 Hz and 400 Hz [14]. The noise determination of $\chi''(\omega)$ does extrapolate these measurements to much lower frequencies within a factor 2 due to our limited absolute accuracy.

Exactly similar measurements performed with a bulk cylinder of $(\text{Al}_2\text{O}_3)_{0.1}(\text{MnO})_{0.5}(\text{SiO}_2)_{0.4}$ (23 mm $\times$ 6 mm diam) give essentially the same inverse frequency dependence of the noise as is visible in figure 2, but the amplitude of the $1/f$ noise is now $\sim 5 \times 10^{-20}$ erg/Hz, that is two orders of magnitude below that of CsNiFeF_6 . The total noise power level now exceeds only by a factor two, at most, the empty system noise so that subtracting one from the other is safe only in a restricted range of frequencies (10^{-1} Hz to 2 Hz). This reduction factor in amplitude is expected since it is close to the ratio of the out of phase susceptibility of both systems, which is measured to be of the order of, or less than, 10^{-2} at 4.2 K [15].

What is the value of this overall agreement ?

Statistical mechanics assumes [16] for the fluctuation dissipation theorem that the system is ergodic, i.e. that time averages are identical to thermodynamic averages. This is not true in

Fig. 2. — Noise power spectrum of $(\text{Al}_2\text{O}_3)_{0.1}(\text{MnO})_{0.5}(\text{SiO}_2)_{0.4}$ after subtraction of the empty system noise (●) at 4.2 K. The lower level of the $1/f$ noise compared with figure 1a for CsNiFeF_6 is consistent with the reduction in $\chi''(\omega)$ between these two systems. Dashed lines : pure copper (see Fig. 1a).

spin glasses. What we measure is rather

$$(M^2)_{\omega,t} = \text{Re} \int_0^t \langle m(0) \cdot m(t') \rangle_t e^{i\omega t'} dt'$$

where $\langle \rangle_t$ means a thermodynamic average over the phase space that the system can visit during time t , starting from given initial conditions. Inasmuch as the same restrictive conditions apply to a measurement of χ'' , one may hope that a fluctuation dissipation relation holds between $(M^2)_{\omega,t}$ and $\chi''_t(\omega)$. The present work suggests that it is the case above 1 Hz. On the other hand the non-stationary noise amplitude observed at low frequency could be manifestation of the non-ergodicity of the system [17].

A more ambitious goal is to relate our noise measurements to what is known from the time decay of the remanent magnetization. The time decay of the remanent magnetization of CsNiFeF_6 at 4.2 K, measured between 3 s and 8×10^3 s, is best fitted by a $t^{-0.04 \pm 0.002}$ time power law on 4 decades, implying an $1/f^{0.96}$ noise spectrum : this is not incompatible with our direct noise measurements. Studies of AgMn suggest that a stretched exponential would be a good description for the time dependence of the remanent magnetization [18]. The corresponding power spectrum is expected to be $\propto \omega^{n-2}/\omega_0^{n-1}$ for $\omega > \omega_0$ and constant for $\omega < \omega_0$, where n and ω_0 are the characteristic parameters of the stretched exponential. The values $n = 0.8$ and $\omega_0 = 10^{-6}$ Hz found for AgMn are consistent with a $1/f^{1.2}$ noise power spectrum.

Opposite to our macroscopic scale of measurement, NMR spin longitudinal relaxation $T_1^{-1}(\omega)$ experiments [19] give access to a local determination of the electron correlation function. This quantity is found to vary like $\omega^{-0.7 \pm 0.3}$ in CuMn for $10^7 \text{ Hz} < f < 10^9 \text{ Hz}$, suggesting as well as muon spin resonance results [20] a high frequency extension of the range of the $1/f$ law.

Finally our results may serve to test theoretical predictions for the exponent of the frequency dependence of the correlation function [21]. In the mean-field dynamical picture a $\omega^{\nu-1}$ power spectrum is expected with $0.25 < \nu < 0.5$. We note that numerical simulations seem to agree better with our experiments since a $1/f$ dependence is found for the noise in 2-dimensional [22] and 3-dimensional [23] Ising systems.

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