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Equilibrium magnetic fluctuations in a Ruderman-Kittel-Kasuya-Yosida spin glass

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Using superconducting quantum interference device magnetometry we have measured the temperature dependence of the equilibrium magnetic fluctuations in the amorphous metallic spin glass $(Fe_{0.15}Ni_{0.85})_{75}P_{16}B_6Al_3$. Above the spin-glass temperature the noise power spectra display pronounced plateaus in the limit of very low frequencies. This new result of spin glasses is in accordance with data obtained from direct measurements of the dynamic susceptibility. Our results give essential implications on the proper dynamic scaling approach in Ruderman-Kittel-Kasuya-Yosida spin glasses.

Experimental observations of spontaneous magnetic fluctuations is the only method that allows a study of low-frequency dynamics in spin glasses at zero magnetic field. This method is particularly important close to the spin-glass freezing temperature T_g, where the equilibrium approach of the spin-glass relaxation exhibits a nonlinear field dependence. The first experimental observations of spontaneous magnetic fluctuations in spin glasses were made by Ocio, Bouchiat, and Monod[1] on some insulating spin-glass systems. They also demonstrated the validity of the fluctuation-dissipation theorem, which relates the magnetic noise power to the dissipative part of the dynamic susceptibility. The electrical Johnson noise can overshadow the magnetic noise in metallic spin glasses. However, measurements on an amorphous metallic spin-glass system[2] have shown the possibility to accurately observe the magnetic noise also in metallic spin glasses. The temperature dependence of the magnetic noise in some insulating spin glasses has recently been reported by Reim, Koch, Malozemoff, and Ketchen[3] and Refregier, Ocio, and Bouchiat.[4] A salient observation in these measurements is a continuing $1/f$ (or $1/\omega$) character of the noise power spectra through the spin-glass temperature.

In this Rapid Communication we report the first experimental measurements of the magnetic noise in a Ruderman-Kittel-Kasuya-Yosida (RKKY) spin glass in the vicinity of the spin-glass temperature. It is found that the noise power spectra show a plateau in the low-frequency limit, which according to the fluctuation-dissipation theorem corresponds to a direct ω dependence of the dissipative part of the dynamic susceptibility. This is in accordance with direct measurements of the dynamic susceptibility and yield fundamental implications on what is a good scaling approach for RKKY spin glasses.

The experiments were performed in a superconducting quantum interference device (SQUID) magnetometer[5] on the amorphous metallic spin glass $(Fe_{0.15}Ni_{0.85})_{75}$-$P_{16}B_6Al_3$. Ribbons of the alloy $(Fe_xNi_{1-x})_{75}$ $P_{16}B_6Al_3$ were prepared by the centrifugal spin quenching technique.[6] The ribbons have a typical cross section of 0.02×1 mm^2. The magnetic phase diagram and general magnetic properties of this alloy system have been reported elsewhere.[7] The high electrical resistivity and the shape of the ribbons limit the electrical Johnson noise and allow an accurate observation of the magnetic noise in a large frequency interval. In the present investigation the sample consists of 90 ribbon pieces 20 mm in length, which are packed together. The sample is attached to a sapphire rod, which constitutes part of the sample holder. The temperature of the sample is measured with a copper resistance thermometer[8] and gives a temperature control of the sample of about 0.1 mK. For the magnetic noise measurements the sample is centered in a third-order gradiometer pick-up coil, and for the measurements of the susceptibility the sample is slightly off centered in the gradiometer coil. The external field is applied by a superconducting magnet operating in the persistent mode. The magnetic flux in the pick-up coil is transferred via a closed superconducting loop to an rf SQUID, which is utilized in the SHE Corporation (San Diego) 330X SQUID system. The transfer ratio of magnetic noise power from the sample to the signal coil of the SQUID sensor is approximately 0.02.

According to the fluctuation-dissipation theorem, the imaginary (dissipative) part of the susceptibility $\chi''(\omega)$ is related to the magnetic noise power through

$$S(f)=4kT\chi''(\omega)/\omega,\ \ \omega=2\pi f. \tag{1}$$

In conventional power-law scaling,[9] it is required that $\chi''(\omega)$ goes to zero as an integral power of ω, and at a specific temperature above T_g the zero-frequency behavior is simply given by

$$\chi''(\omega)\propto\omega. \tag{2}$$

Hence, power-law scaling implies that the magnetic noise power spectra should display a plateau at sufficiently low frequencies. The level of that plateau should gradually increase an approaching T_g, and at temperatures close to and below T_g a $1/f$-like behavior should be found at all experimental frequencies due to a weak frequency dependence of $\chi''(\omega)$. The observations by Reim *et al.*[3] and Refregier *et al.*[4] on insulating spin glasses of a continuing $1/f$ behavior of the noise power through T_g is seemingly at variance with this basic prediction of power-law scaling, but may be explained within the novel "droplet picture" for short-range systems proposed by Fisher and Huse.[10] This picture implies[10] a $1/f$ character of the magnetic noise power only with logarithmic corrections. Recently, within this picture Malozemoff and Pytte[11] have performed an activated (logarithmic) dynamic scaling analysis on $Eu_{0.4}Sr_{0.6}S$ at temperatures above the spin-glass temperature. They found it possible to achieve a good scaling plot, but they could not at the same time rule out the possibility of conventional power-law scaling due to lack of sufficiently accurate experimental data at low frequencies

Experimentally it is very difficult to obtain accurate data in the low-frequency regime using ordinary ac technique. A method which allows a more accurate observation of the equilibrium approach is the zero-field-cooled (ZFC) technique, where the sample is cooled in zero field to the measurement temperature T_m. At T_m the external field is applied and the time dependence of the relaxation is observed. The dynamic susceptibility components may be obtained from such measurements by taking the Fourier transforms of the measured time dependence of the ZFC magnetization. Figure 1 shows the relaxation rate $(1/H)\partial M/\partial \ln t$ of the ZFC magnetization for the $(Fe_{0.15}Ni_{0.85})_{75}P_{16}B_6Al_3$ sample, at various temperatures above T_g, in the observation time window 3×10^{-1}–3×10^3 sec. Figure 2 shows the frequency dependence of $\chi''(\omega)/\omega$ for the same temperatures calculated from the experimental data of Fig. 1. As is shown in Fig. 2, $\chi''(\omega)/\omega$ levels out on a plateau at sufficiently low frequencies. This implies that $\chi''(\omega)\propto\omega$ in the zero-frequency limit in accordance with the basic requirement in power-law scaling. The plateau values of the quantity $[1/\chi(0)]\chi''(\omega)/\omega$ are equal to the "average" correlation times as defined by Ogielski.[13] As is seen in Fig. 2, these correlation times increase rapidly on approaching T_g.

Our measurements indicate deviations from the "$\pi/2$" rule[11,14] at the onset of the plateau. The $\pi/2$ rule relates the imaginary and real parts of the dynamic susceptibility through

$$\chi''(\omega)=(\pi/2)\partial\chi'(\omega)/\partial\ln\omega\,. \quad (3)$$

This relation has been shown to apply in activated scaling,[11] but the present measurements indicate that $\partial\chi'/\partial\ln\omega$ drops faster than $\chi''(\omega)$ at low frequencies. These results imply a low-frequency limit for activated scaling in RKKY spin glasses, and the equilibrium approach needs to be properly addressed within that picture. In general, our susceptibility data favor conventional power-law scaling.

The ZFC measurements were performed at an external field of 40 mG, which was the lowest field for a reasonably accurate observation of the relaxation. In the regime of linear response of the relaxation one expects, according to the fluctuation-dissipation theorem, that the magnetic noise power should exhibit a similar frequency dependence as $\chi''(\omega)/\omega$, obtained from the ZFC measurements. However, due to nonlinear field effects on the relaxation, measurements of the spontaneous magnetic fluctuations is the only method to observe the zero-field low-frequency dynamics in the immediate vicinity of T_g. The present measurements of the magnetic noise were made at a residual external field of less than 1 mG. Figure 3 shows the detected magnetic flux noise as a function of time at some temperatures around T_g. As is seen from Fig. 3 the detected noise increases very rapidly on approaching T_g from above and reaches a maximum amplitude around T_g. The time domain observations in Fig. 3 reflect the fluctuations in the frequency range 10^{-4}–5×10^{-2} Hz. The drastic increase of the fluctuations on approaching T_g is in accordance with the temperature dependence of $\chi''(\omega)$ at various frequencies.[15] Figure 4(a) shows the frequency dependence of the noise power $S(f)$ for $T=1.01T_g$ and $T=1.11T_g$ as obtained from fast Fourier transform (FFT) analyses of the time domain observations. At $T=1.01T_g$, $S(f)$ exhibits a close $1/f$ behavior, which, according to the fluctuation-dissipation theorem, implies a weak frequency dependence of $\chi''(\omega)$. At $T=1.11T_g$ the measured noise power of the sample is indistinguishable from the corresponding spectrum of an "empty coil" experiment. This is an important and funda-

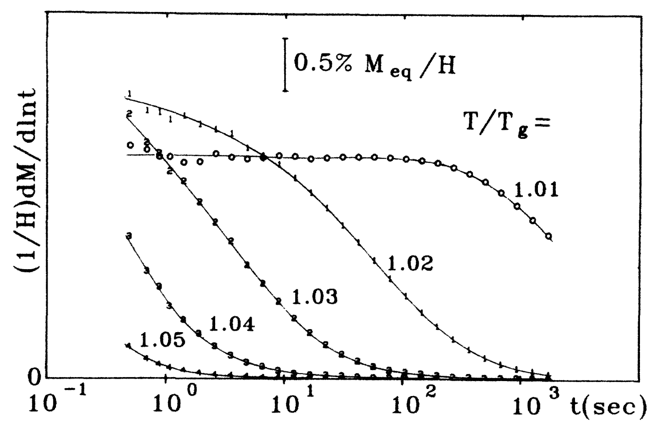

FIG. 1. Relaxation rate of the zero-field-cooled susceptibility, $(1/H)\partial M/\partial \ln t$ vs $\log_{10}t$, for $(Fe_{0.15}Ni_{0.85})_{75}P_{16}B_6Al_3$ at some temperatures above the spin-glass temperature $T_g=22.4$ K. 0.5% of the equilibrium susceptibility M_{eq}/H is indicated. The external field is $H=40$ mG.

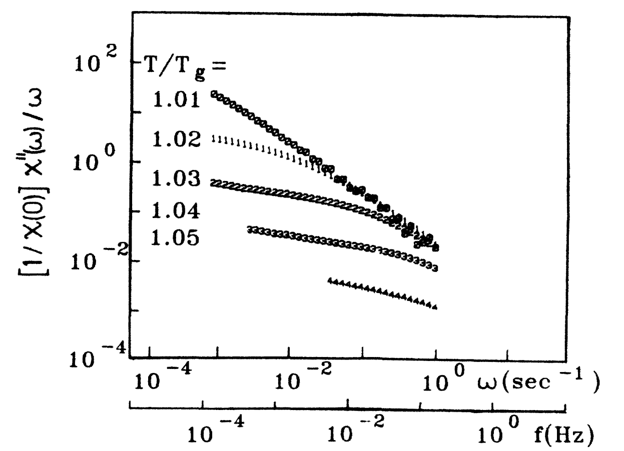

FIG. 2. Frequency dependence of $\chi''(\omega)/\omega$ plotted in a log-log diagram. The data points are obtained from the Fourier sine transforms of the experimental data of Fig. 1. The equilibrium susceptibility $\chi(0)=4.0$ (SI) (Ref. 12).

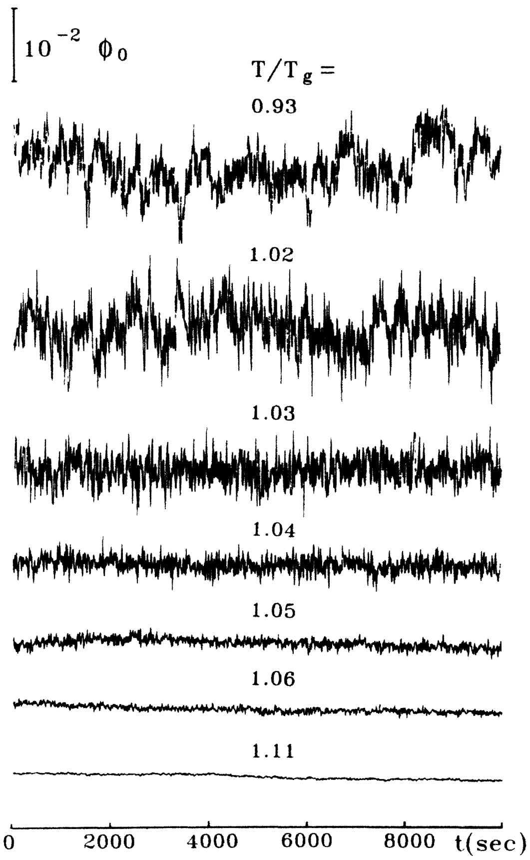

FIG. 3. Magnetic flux noise as a function of time for $(Fe_{0.15}Ni_{0.85})_{75}P_{16}B_6Al_3$ at various temperatures around the spin-glass temperature $T_g=22.4$ K. A relative flux change in the SQUID sensor of $10^{-2}\phi_0$ ($\phi_0=2\times10^{-15}$ Vs) is indicated. The sampling period of the data is 10 sec. An antialiasing filter of 10^{-1} sec (0.1 Hz) is used.

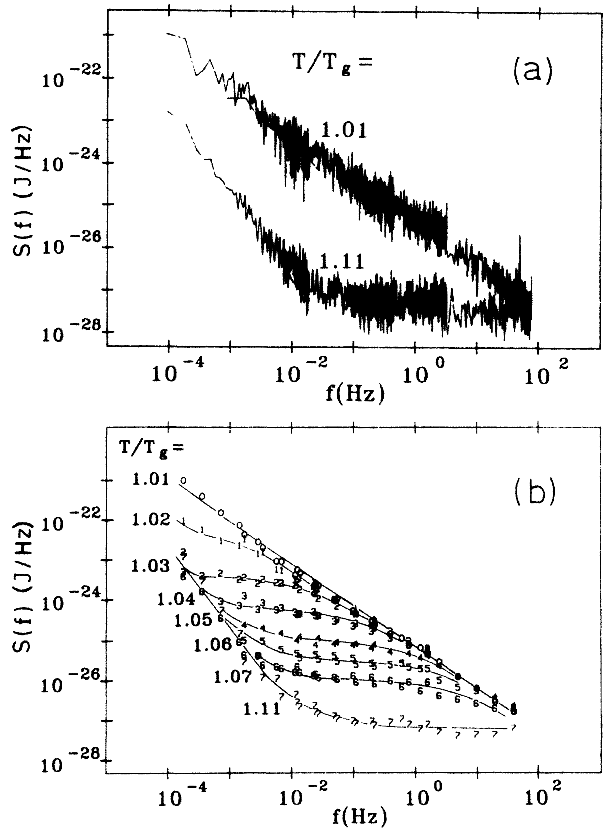

FIG. 4. Magnetic noise power $S(f)$ vs frequency, as detected in the signal coil of the SQUID sensor, for $(Fe_{0.15}Ni_{0.85})_{75}P_{16}B_6Al_3$ at various temperatures above the spin-glass temperature $T_g=22.4$ K. The transfer ratio of noise power from the sample to the signal coil of the SQUID sensor is approximately 0.02. (a) Experimental raw data at two representative temperatures. The spectrum at $T=1.11T_g$ is indistinguishable from that obtained in an "empty coil" experiment. (b) Smoothed spectra, obtained by averaging the experimental raw data over small logarithmic frequency segments.

mental result that should always be obtained in noise measurements on spin glasses. There are no low-frequency magnetic relaxation phenomena in true spin glasses that can give any resolvable contribution to the noise power spectrum at temperatures well above T_g. Figure 4(b) shows noise power spectra at various temperatures above T_g, where the experimental raw data have been averaged over small logarithmic frequency segments. As is seen from Fig. 4(b) the noise power, at a specific temperature, breaks away from a $1/f$ behavior at a certain frequency, and levels out on a plateau at low frequencies. At the four lowest temperatures, the observed frequency dependence of $S(f)$ compares most favorably with the frequency dependence of $\chi''(\omega)/\omega$ (cf. Fig. 2) as obtained from direct susceptibility measurements. At higher temperatures the magnitude of the plateau value of $\chi''(\omega)/\omega$ becomes somewhat smaller than the corresponding value of $S(f)$. This deviation is caused by a too limited time range for the observation of the ZFC relaxation to accurately obtain the proper Fourier sine transforms.

The temperature dependence of the correlation length and average correlation time may be determined from the

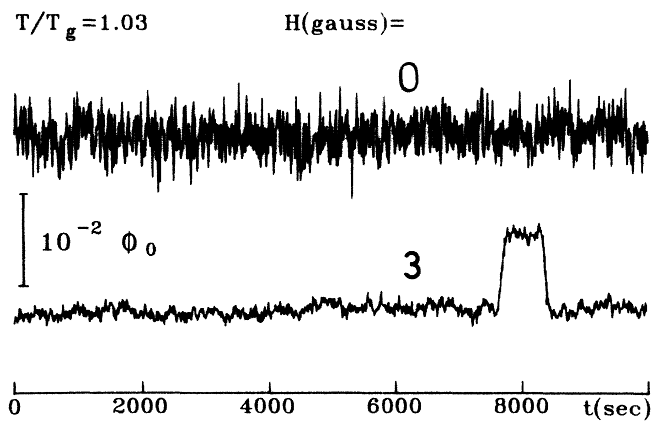

FIG. 5. Magnetic flux noise as a function of time at zero field and at $H=3$ G for $(Fe_{0.15}Ni_{0.85})_{75}P_{16}B_6Al_3$ at $T/T_g=1.03$ ($T_g=22.4$ K). The effect of a temperature cycling of 1 mK is shown for the $H=3$ G curve. A relative flux change in the SQUID sensor of $10^{-2}\phi_0$ is indicated.

plateau values of the noise power spectra. However, we hesitate to squeeze too much out of the present data but also point out that the present measurements have been made with a SQUID sensor of moderate energy resolution and on a sample occupying only 5% of the effective pick-up coil volume. It should be feasible to obtain a substantial improvement of the present data, and thus to extend the present frequency range some orders of magnitude.

At temperatures just above T_g the magnetic noise is gradually quenched with the application of an external field. Figure 5 shows the time domain observation of magnetic noise at zero field and at an external field of $H=3$ G at the same temperature. As is seen from Fig. 5, the noise is reduced by the external field. Also shown is the effect of a temperature change of 1 mK on the $H=3$ G curve, which amply demonstrates the drastic effect of even a minute change of an external parameter on the detected noise. The effect of a temperature change of 1 mK corresponds to a change in bulk magnetization of only 0.5 ppm if the gradiometer pick-up coils should have been wound in a magnetometer configuration. We find it important to study the influence of external fields on the low-frequency dynamics of spin glasses in some more detail by measurements of both the magnetic noise and the dynamic susceptibility.

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