Nuclear-Spin Noise

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The spectral density of Nyquist noise current in a tuned circuit coupled to a sample of nuclear spins has been measured at ^{4}He temperatures with a dc SQUID used as a rf amplifier. When the sample is in thermal equilibrium, a dip is observed in the spectral density at the Larmor frequency. For zero spin polarization, on the other hand, a bump in the spectral density is observed. This bump is due to temperature-independent fluctuations in the transverse component of magnetization, and represents spontaneous emission from the spins into the circuit.

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In his pioneering paper[1] on nuclear induction, Bloch noted that in the absence of any external radiofrequency (rf) driving field a sample of N spins of magnetic moment μ contained in a pickup coil would induce very small voltage fluctuations proportional to $N^{1/2}\mu$. In this Letter, we report the observation of these temperature-independent fluctuations at liquid-^{4}He temperatures arising from the ^{35}Cl nuclei in $NaClO_3$ at the nuclear quadrupole resonance (NQR) frequency of about 30 MHz.

In the experiment, a sample of nuclear spins is placed in the inductor L_p of a tuned LCR circuit and the spectral density of the current fluctuations is measured over the bandwidth of the circuit. The circuit resistance R_i produces a Nyquist voltage noise and therefore a current noise that, in the absence of a sample, has a Lorentzian spectral density. The presence of the sample is found to modify the shape of this noise-power spectrum in the region of the NQF frequency. The influence of the sample is determined from its complex spin susceptibility[2] $\chi(\omega)=\chi'(\omega)-j\chi''(\omega)$, where χ' and χ'' are the dispersion and absorption. The complex impedance of the coil in the presence of the sample is written as

$$Z_p'=j\omega L_p'=j\omega L_p[1+4\pi\xi\chi(\omega)]$$
$$=j\omega[L_p+L_s(\omega)]+R_s(\omega)\ ,$$

where $\xi=\mathscr{V}_s/\mathscr{V}_c$ is the sample filling factor; $\mathscr{V}_s$ and $\mathscr{V}_c$ are the volumes of the sample and the pickup coil. The added spin inductance $L_s=4\pi\xi L_p\chi'$ shifts the circuit resonant frequency, while the added spin resistance $R_s=4\pi\xi\omega L_p\chi''$ modifies the damping of the circuit and acts as a source of Nyquist noise. This noise is due to spin fluctuations in the transverse direction. To observe these fluctuations in a reasonable averaging time one requires, first, that R_s/R_i be not too small, and second, that the noise current be measured by an amplifier with a noise temperature comparable with or smaller than the bath temperature T.

We can compute the Nyquist noise generated by the spins in terms of the microscopic parameters of the sample. Since the NQR sample is equivalent to a two-level system,[3] we take as a model an ensemble of spins in an external magnetic field $H_z\hat{\mathbf{z}}$ with spin $I=\frac{1}{2}$, spin density $n=N/\mathscr{V}_s$, and Larmor frequency $\omega_s/2\pi=\gamma H_z/2$, where γ is the gyromagnetic ratio. The axis of the pickup coil is along the x direction. We ascribe a spin temperature T_s to the magnetization

$$M_z=(n\gamma\hbar/2)\tanh(\hbar\omega_s/2k_BT_s)\ . \tag{1}$$

We assume that Bloch's equations apply, so that $\chi''=\chi'/\Delta\omega T_2$ is given by

$$\chi''(\omega)=M_z\gamma T_2/2[1+(\Delta\omega)^2T_2^2]\ , \tag{2}$$

where $\Delta\omega=\omega_s-\omega$, and the linewidth is given by $\Delta f_s=1/\pi T_2$. The spectral density of the Nyquist noise voltage produced by R_s is[4]

$$S_v^s(\omega)=\frac{2}{\pi}R_s\frac{\hbar\omega}{2}\coth\left[\frac{\hbar\omega}{2k_BT}\right]\ . \tag{3}$$

Combining Eqs. (1)–(3) with $R_s=4\pi\xi\omega L_p\chi''$, and neglecting terms of order $\Delta\omega/\omega_s \ll 1$, we obtain

$$S_v^s(\omega)=\frac{\xi L_p\omega_s^2 n\gamma^2\hbar^2T_2}{1+(\Delta\omega)^2T_2^2}\ . \tag{4}$$

Remarkably, because of the cancellation of the hyperbolic terms in Eqs. (1) and (3), $S_v^s(\omega)$ is independent of temperature throughout both the quantum ($\hbar\omega \gg k_BT_s$) and classical ($\hbar\omega \ll k_BT_s$) regimes. This temperature independence which holds only for a two-level system, can be understood in a different way by applying Faraday's law of induction to obtain the total mean square voltage $\langle V_s^2\rangle=4\pi\xi L_p\omega_s^2\mathscr{V}_s\langle M_x^2\rangle$ across the coil in terms of the mean square magnetization $\langle M_x^2\rangle$. For a sample with N spins, $\langle M_x^2\rangle=N\langle\mu_x^2\rangle/\mathscr{V}_s^2=n\gamma^2\hbar^2\langle I_x^2\rangle/\mathscr{V}_s$, where the magnetic moment of a single spin is $\mu_x=\gamma\hbar I_x$ and

$\langle I_x^2 \rangle = \frac{1}{4}$ for spin $I = \frac{1}{2}$. Making these substitutions, we find $\langle V_s^2 \rangle = \pi \xi L_p \omega_s^2 n \gamma^2 \hbar^2$, which is just $\int_{-\infty}^{\infty} S_v^s(\omega)\, d\omega$.

The configuration of the experiment is shown in Fig. 1. The sample is contained in a superconducting coil L_p which is in series with a capacitor C_i that can be adjusted from outside the cryostat. A superconducting inductance L_i ($\ll L_p$), also in series, forms the input coil of a dc superconducting quantum interference device (SQUID).[5] The resistance R_i represents contact resistance and losses in the capacitor. Current fluctuations in the input circuit generate a magnetic flux in the loop of the SQUID, which in turn produces an output voltage. In the configuration shown, the SQUID is a linear rf amplifier which operates at a signal frequency of $\omega/2\pi = 30$ MHz with a gain of about 20 dB and a noise temperature of about 0.2 K for a bath temperature of 1.5 K. The output from the SQUID is recorded by a spectrum analyzer interfaced to a computer.

Noise measurements were made on 0.63 cm^3 of powdered $NaClO_3$, with a filling factor $\xi = 0.35$. The ^{35}Cl nucleus with $I = \frac{3}{2}$ has two doubly degenerate energy levels with a transition frequency at 1.5 K of $\omega_s/2\pi = 30.6857$ MHz and a transverse relaxation time $T_2 = 240$ μs corresponding to a linewidth $\Delta f_s = 1.3$ kHz. We neglect departures from ideal behavior that occur in a solid—for example, inhomogeneous broadening—and assume that χ has the form given by Eq. (2) with the value of T_2 given above.

We carried out measurements for two cases. In the first, the spin-lattice relaxation time T_1 of the sample was reduced from the order of days to 20 min by γ irradiation of the sample, and the spins were allowed to reach equilibrium[6] ($T_s = T$) with the helium bath at 1.5 K. In the high-temperature limit $\hbar\omega \ll k_B T$, the spectral density of the current noise in the input circuit is given by

$$S_I(\omega)\,|_{T_s = T} = \frac{(2/\pi) k_B T [R_i + R_s(\omega)]}{[R_i + R_s(\omega)]^2 + |X|^2}, \qquad (5)$$

where $X = j\omega[L_p + L_i + L_s(\omega)] - j/\omega C_i$. When the circuit is tuned exactly to the Lamor frequency $\{\omega = [(L_p + L_i)C_i]^{-1/2}\}$, Eq. (5) reduces approximately to $2k_B T/\pi[R_i + R_s(\omega)]$ near resonance if we assume that Δf_s is much less than the circuit bandwidth Δf_c. Under these conditions the effect of the spins is to produce a "dip" in the spectral density as ω is scanned through the NQR frequency and $R_s(\omega)$ passes through a maximum. An example of such data, averaged over 3 h, is shown in Fig. 2; each of the 1001 points was averaged over a bandwidth of about 300 Hz. The minimum of the dip occurs at the ^{35}Cl NQR frequency (indicated by an arrow), which was measured in a separate, pulsed NQR experiment at the same temperature. Taking into account impedance reflected from the SQUID,[5] we fitted the data with Eq. (5), and find good agreement with the parameters ω_s and T_2 measured separately. The data are consistent with the values $Q = 7320$ and $R_s(\omega_s)/R_i = 0.12$, where Q is the quality factor of the circuit in the absence of the sample. These values lead to $R_s(\omega_s)/R_i Q = 4\pi\xi\chi''(\omega_s) = 1.6 \times 10^{-5}$. The dotted line in Fig. 2 indicates the response we would expect from this fit in the absence of the sample.

In the second case, the spins of a sample of $NaClO_3$ with an extremely long T_1 (days) were saturated by application of continuous rf excitation at resonance. After the excitation was turned off, the spectral density was measured over a time much less than T_1. A saturated sample has zero spin-population difference, so that $m_z = \chi = R_s = L_s = 0$ and $T_s = \infty$. However, according to Eq. (4), the product $R_s(\omega) T_s$ (for $\hbar\omega \ll k_B T$) is independent of T_s for a given frequency. The spectral density of the current becomes

$$S_I(\omega)\,|_{T_s = \infty} = \frac{(2/\pi) k_B [R_i T + R_s(\omega) T_s]}{R_i^2 + [\omega(L_p + L_i) - 1/\omega C_i]^2}. \qquad (6)$$

Thus, one would expect to observe a "bump" in the spectral response arising from the term $R_s T_s$ in the numerator of Eq. (6). An example of our data, averaged over 7 h, is shown in Fig. 3(a) with the NQR frequency indicated by an arrow. Fitting the data with Eq. (6) we find $Q = 3.430$, $R_s(\omega_s) T_s / R_i T = 0.06$, and $4\pi\xi\chi''(\omega_s) = 1.7 \times 10^{-5}$, a value in good agreement with that obtained in the previous (equilibrium) case. The dotted line indicates

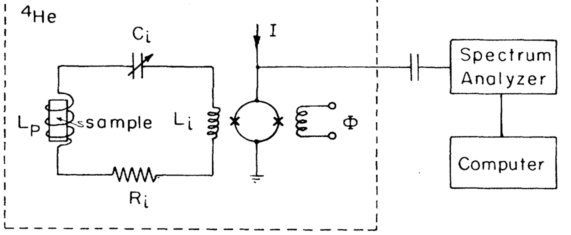

FIG. 1. Experimental configuration. Components in dashed box are immersed in liquid ^4He.

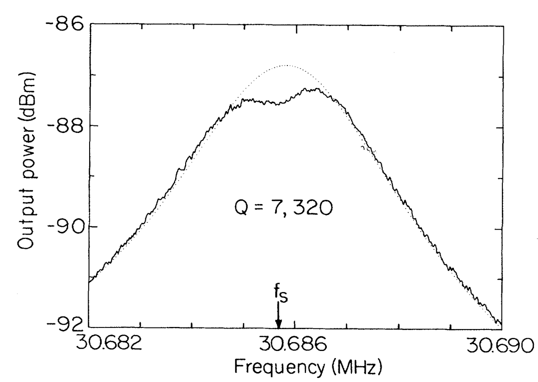

FIG. 2. Spectral density of noise current for a $NaClO_3$ sample in thermal equilibrium at 1.5 K.

the expected power spectrum in the absence of the sample. Figure 3(b) shows the excess noise in Fig. 3(a) due to the spins, and represents the first observation of nuclear-spin noise.

To discuss our results, we note that the spin system at temperature T_s is coupled to the resistance R_i at bath temperature T by absorbing noise power P_R generated by R_i, and simultaneously emitting power P_s into R_i. The net power flow $\Delta P = P_s - P_R$ into R_i (if we assume $\Delta f_s \ll \Delta f_c$ and $\hbar\omega \ll k_B T$) is

$$\Delta P = \int_{-\infty}^{\infty} \frac{2k_B R_i R_s(\omega)}{\pi[R_i + R_s(\omega)]^2}(T_s - T)\,d\omega \ . \qquad (7)$$

Alternatively, we can derive this expression in terms of the dynamics of the spin system if we express the power ΔP in terms of Bloch's equations, modified to include the effect of the circuit (radiation damping).[7] For $R_s \ll R_i$ we find

$$\Delta P = \mathscr{V}_s H_z\, dM_z/dt$$
$$= 4\pi\xi \mathscr{V}_s Q\omega_s \langle M_x^2 \rangle - \mathscr{V}_s \omega_s S_H(\omega_s) \int_{-\infty}^{\infty} \chi'' d\omega \ , \quad (8)$$

where $\langle M_x^2 \rangle = n\gamma^2\hbar^2/4\mathscr{V}_s$. The first term on the right-hand side of Eq. (8) is the power P_s emitted by the spins into R_i and arises from the radiation damping of fluctuations in M_x. This power can also be expressed as $P_s = 2k_B T_s/\tau_R$ (for $k_B T_s \gg \hbar\omega$) where $1/\tau_R = 2\pi\xi\gamma M_z Q$ is the radiation damping rate.[7,8] The second term on the right-hand side of Eq. (8) represents the absorption by the spins of the magnetic field energy produced by noise in R_i at temperature T. The spectral density $S_H(\omega_s)$ of the magnetic field H_x is determined from

$$\frac{S_H(\omega_s)}{8\pi} = \mathscr{D}_c(\omega_s)\frac{\hbar\omega_s}{4}\coth\left[\frac{\hbar\omega_s}{2k_B T}\right],$$

where $\mathscr{D}_c(\omega_s) = 2Q/\pi\omega_s\mathscr{V}_c$ is the density of states for the single-cavity mode. With some manipulation, Eq. (8) results in the familiar form of Einstein's detailed balance equation,

$$\Delta P = (\hbar\omega_s/2)d\Delta N(T_s)/dt$$
$$= \frac{\hbar\omega_s}{2}A\left[N - \Delta N(T_s)\coth\left[\frac{\hbar\omega_s}{2k_B T}\right]\right], \qquad (9)$$

where $\Delta N(T_s)$ is the total spin-population difference and $A = 2\pi Q\hbar\gamma^2/\mathscr{V}_c$ is the spontaneous emission rate into a resonant cavity.[9,10] The equivalence of Eqs. (8) and (9) demonstrates the close connection between spin fluctuations and spontaneous emission and shows that the bump observed in Fig. 3 represents spontaneous emission from the spins into the circuit.

Although for our experiment the spontaneous emission rate for one spin is extremely low, $A = 2\times 10^{-16}$ sec^{-1} (about one spin flip in 10^8 years), since $N = 2\times 10^{21}$ the total emission rate ($NA/2$) is $\approx 2\times 10^5$ sec^{-1}, corresponding to a total emissive power $P_s \approx 5\times 10^{-21}$ W. This power is about 5% of the Nyquist noise power $4k_B T/\pi T_2 = 10^{-19}$ W generated in the bandwidth of the spin noise $1/\pi T_2$.

In conclusion, we note that the value of T_1 does not affect the noise measurements except to determine the rate at which the sample reaches thermal equilibrium. This longitudinal relaxation process would induce very small voltage fluctuations about zero frequency across a pickup coil with its axis parallel to the direction of the spin polarization. Finally, this method of observing fluctuations from saturated spins provides a means for determining the resonant frequency and linewidth in systems where T_1 is impractically long for conventional techniques.

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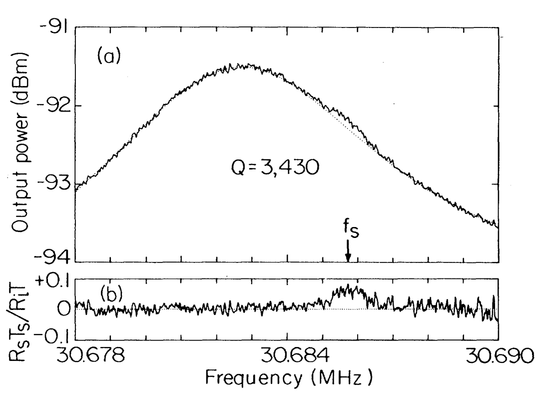

FIG. 3. Spectral density of (a) noise current for a $NaClO_3$ sample with saturated spins ($T_s = \infty$), and (b) nuclear-spin noise of $NaClO_3$ sample obtained from (a).

[1]F. Bloch, Phys. Rev. **70**, 460 (1946).

[2]C. P. Slichter, *Principles of Magnetic Resonance* (Springer-Verlag, Berlin, 1978), 2nd ed., p. 37.

[3]M. Bloom, E. L. Hahn, and B. Herzog, Phys. Rev. **97**, 1699 (1955).

[4]H. B. Callen and T. A. Walton, Phys. Rev. **83**, 34 (1951).

[5]C. Hilbert and J. Clarke, Appl. Phys. Lett. **43**, 694 (1983), and IEEE Trans. Magn. **21**, 1029 (1985), and J. Low Temp. Phys. **61**, (1985); C. Hilbert, J. Clarke, T. Sleator, and E. L. Hahn, Appl. Phys. Lett. **47**, 637 (1985), and Bull. Am. Phys. Soc. **30**, 478 (1985).

[6]Related noise measurements on other systems in equilibrium are reported by J. J. Brophy, in *Fluctuation Phenomena in Solids,* edited by R. E. Burgess (Academic, New York, 1965); G.

Alzetta, E. Arimondo, and C. Ascoli, Nuovo Cimento **54B**, 163 (1968).

[7]S. Bloom, J. Appl. Phys. **28**, 800 (1957).

[8]N. Bloembergen and R. V. Pound, Phys. Rev. **95**, 9 (1954); C. R. Bruce, R. E. Norberg, and G. E. Pake, Phys. Rev. **104**, 419 (1956).

[9]E. M. Purcell, Phys. Rev. **69**, 681 (1946). For recent work concerning effects of cavity modes on spontaneous emission, see D. Kleppner, Phys. Rev. Lett. **47**, 233 (1981); G. Gabrielse and H. Dehmelt, Phys. Rev. Lett. **55**, 67 (1985).

[10]Equations (8) and (9) are identical with Eq. (7) to first order in $R_s/R_i(\ll 1)$. To obtain agreement to higher order, we take into account the effect of R_s on the density of states by writing $Q=\omega(L_p+L_i)R_i/(R_i+R_s)^2$ in the steps leading to Eq. (8). This yields a result in agreement with Eq. (7) to second order in R_s/R_i.