Journal of Low Temperature Physics, Vol. 110, Nos. 1/2, 1998

Nonlinear Response of a Continuous Wave NMR Spectrometer for Samples with a Large Magnetization

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We present a new insight in the response of a CW NMR spectrometer for highly magnetized samples above 100 MHz. The spectrometer is a bridge made of a magic "T". The output of the bridge is proportional to the reflection coefficient, Γ, of a resonant circuit, which is built with a coil containing the sample. The sensitivity of the reflection coefficient to the complex susceptibility of the sample, $\chi(\omega) = \chi'(\omega) - j\chi''(\omega)$, depends on the quality factor, Q, of the circuit and filling factor, η. When the condition $Q\eta\chi'' \ll 1$ is not fulfilled, we show indeed that the use of a simple crystal detector, which is only sensitive to $|\Gamma|$ gives rise to a strong nonlinear response of the spectrometer. Measurements of the complex value of Γ by means of phase sensitive detection allow to recover a linear behavior. We discuss and illustrate those issues with a few circuits we designed for our measurements on liquid ^3He with spin polarizations up to 15 %. A method is described to build in a reproducible and predictable way resonant circuits matched to 50 Ω in the frequency range 100 - 400 MHz with a quality factor as high as 1000 at 4 K.

PACS numbers: 07.58.+g

1. INTRODUCTION

A continuous wave NMR spectrometer is used to perform experiments on liquid ^{3}He, polarized to 15 % in a 7 T magnetic field by out of equilibrium methods [1]. The experimental data at the higher polarizations lead to the conclusion that the output of the spectrometer does not increase linearly with the polarization: the polarization should be higher than measured by the spectrometer. In this paper, we investigate the origins of this nonlinear

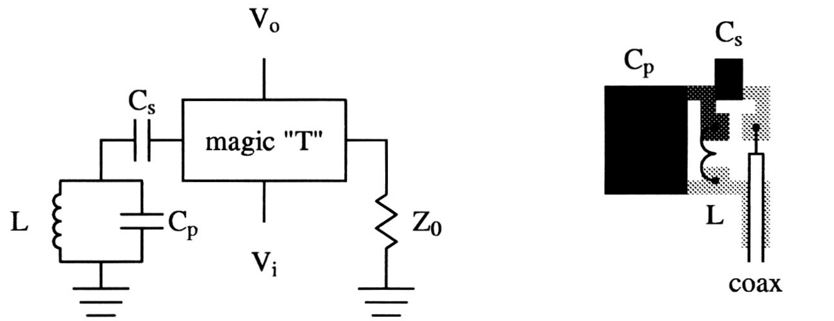

Fig. 1. Left: simplified circuit diagram of the spectrometer; $V_o \propto (\Gamma_{LCR} - \Gamma_{50\Omega})V_i$. Right: C_p and C_s made out of a copper-Kapton-copper laminate; the copper on one side is outlined by the light-gray and black regions, the copper on the other side is outlined by the dark-grey and black regions, C_p and C_s are the overlapping black regions.

behavior.

The detection method used by the spectrometer boils down to a measurement of the impedance of a tuned circuit with a coil containing the sample with a complex r.f. susceptibility $\chi = \chi' - j\chi''$. The magnitude of the impedance of such a circuit, $|Z_{LCR}|$, is in first order proportional to χ'' if

$$Q\eta\chi'' \ll 1 \tag{1}$$

where η is the filling factor of the coil and Q the quality factor of the resonant circuit [2]. In this case, a measurement of $|Z_{LCR}|$ is sufficient to obtain χ''. In practice, Eq. (1) may be very hard to satisfy for any experiments on samples with a large magnetization but we will show that even in this case it should be possible to obtain a linear response of the spectrometer if phase sensitive detection is used.

2. THE SPECTROMETER

Fig. 1 shows the spectrometer, which is a bridge built from a magic "T" to compare the reflection coefficient of the resonant circuit, Γ_{LCR}, with the reflection coefficient of a 50 Ω termination, $\Gamma_{50\Omega}$ [3]. The reflection coefficient of a circuit terminating a coaxial line is given by $\Gamma = (Z - Z_0)/(Z + Z_0)$, where Z is the impedance of the circuit and Z_0 the characteristic impedance of the coaxial line. The output voltage, V_o is related to the input voltage, V_i by the relation $V_o \propto (\Gamma_{LCR} - \Gamma_{50\Omega})V_i$ with $\Gamma_{50\Omega} = 0$ [4].

For optimal NMR sensitivity, the resonant circuit should be matched to 50 Ω at the resonance frequency of the nuclear spins. The circuit consists of an inductance, L, and a capacitance, C_p in parallel, both in series with a capacitance C_s. L and C_p form a circuit with an impedance, $Z_p(f)$, which is high at its resonance frequency $f_0 \approx 1/2\pi(LC_p)^{1/2}$. C_s is chosen so that at a frequency, f_r, where the real part of $Z_p(f_r) = 50\ \Omega$, the imaginary part of $Z_p(f_r)$ is canceled: the circuit is resonating at the frequency f_r ($f_r < f_0$) with an impedance of 50 Ω.

In the experiments mentioned above, variations of $|\Gamma|$ have been measured using a HP 8471D crystal detector while sweeping the frequency through the NMR line [5]. The polarization has been assumed to be proportional to $\int |\Gamma_{sample}(f)| - |\Gamma_{empty}(f)| df$. Although, the circuit was not very well matched to 50 Ω, the nonlinear effects at the resonance frequency f_r were so large that the signal coming from the spectrometer initially increased with increasing polarization, went through a maximum and finally decreased at the highest polarizations. Therefore, the data have been taken at a frequency well below f_r, where $|\Gamma|$ is less sensitive to the r.f. susceptibility, but the nonlinear response did still exist.

3. SIMULATIONS

To understand and to be able to eliminate the nonlinear response of the spectrometer, we have compared three plausible methods to extract the polarization, P, from measurements of the reflection coefficient. The three methods are characterized by the following assumptions:

1. $P \propto S_1 = \int |\Gamma_{sample}(f)| - |\Gamma_{empty}(f)| df$. This is the simplest method, limited in usefulness but requiring only a crystal detector.

2. $P \propto S_2 = \int |\Gamma_{sample}(f) - \Gamma_{empty}(f)| df$. This method requires phase sensitive detection.

3. $P \propto S_3 = \int |Z_{sample}(f) - Z_{empty}(f)| df$. This method requires phase sensitive detection and a calibration relating V_o/V_i to Γ necessary for the calculation of Z_{sample} and Z_{empty}.

To simplify the calculations we assumed a Lorentzian NMR line shape with a width, $\Delta f/f = 10^{-4}$, equal to the field inhomogeneity of the magnet over the sample volume. The real line shape is determined by the field gradient. The filling factor, $\eta = 0.025$, is estimated being the volume of the sample divided by twice the volume bounded by the coil. The other parameters, corresponding to a real circuit, are: $L = 64$ nH, $r = 0.4\ \Omega$ (the

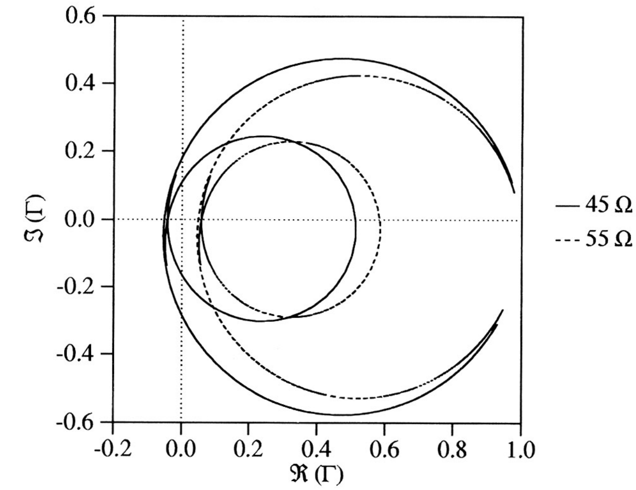

Fig. 2. The reflection coefficient for resonant circuits matched at 45, and 55 Ω with a fully polarized sample ($\eta = 0.025$, $P = 100$ %). The large circles represent Γ_{empty} and the smaller circles Γ_{sample}.

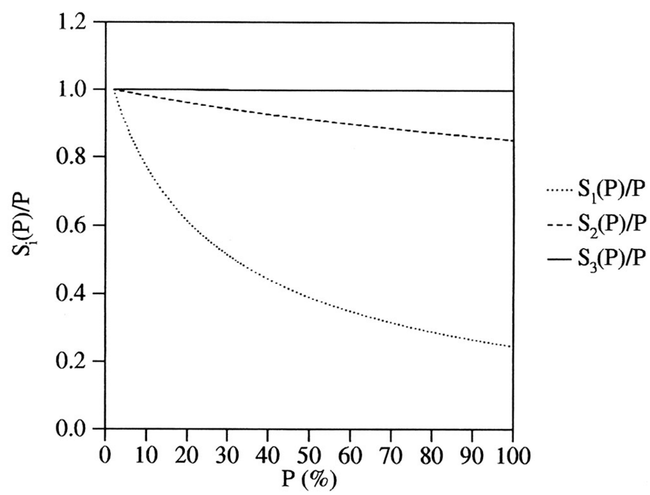

Fig. 3. $S_i(P)/P$ versus P for the resonant circuit adapted to 55 Ω. $S_3(P)/P$ at $P = 100$ % is 2×10^{-3} smaller than at $P = 2$ %.

dissipation is modeled by adding a series resistance to the coil) and C_p = 9.22 pf. For $Z(f_r)$ = 45, 50 and 55 Ω, C_s = 0.961, 0.907 and 0.861 pF at f_r = 197.1914, 197.7181 and 198.1701 MHz.

Fig. 2 shows $\Re(\Gamma(f))$ versus $\Im(\Gamma(f))$ in the presence and absence of a fully polarized sample for resonant circuits adapted to 45 and 55 Ω. For the circuit adapted to 45 Ω and at small polarization, $\Re(\Gamma(f)) < 0$ when sweeping through the resonance line. At higher polarization, this is not true. In this case, the first method is clearly bad, because $|\Gamma_{sample}(f)|-|\Gamma_{empty}(f)|$ does not increase continuously with increasing polarization: it is preferable to match $Z_{LCR}(f_r)$ to a value slightly higher than 50 Ω.

Fig. 3 shows $S_i(P)/P$ versus the polarization P for the circuit matched to 55 Ω. $S_i(P)$ is normalized such that $S_i(P_{eq})/P_{eq} = 1$ at the equilibrium polarization, P_{eq}. The first method is not suitable to polarization measurements of highly magnetized samples. The second method shows an improvement but leads also to a nonlinear response, which arises from the transformation $\Gamma = (Z - Z_0)/(Z + Z_0)$, as is clear from the nearly ideal response of the third method (the nonlinearity $\approx 10^{-3}$ for an increase of $|Z|$ with a factor 3).

4. CONSTRUCTION OF RESONANT CIRCUITS

Before assembling a resonant circuit, we measure the impedance of the NMR coil (a saddle coil with the parasitic capacitance between the wires minimized by a Teflon tube at the wire crossing) at 4.2 K in a ^{4}He storage vessel with a HP 8720B network analyzer (0.13 - 20 GHz). We guess values for C_p and C_s necessary to obtain a reasonable match at the NMR frequency. The capacitors are etched out of a copper-Kapton-copper laminate (35 - 127 - 35 μm thickness and a capacitance per surface of 16 pF/cm^2) according to the layout shown in Fig. 1b. The complete circuit is measured to improve the value of the series resistance, r, of the coil. In general, one iteration is needed to obtain a circuit with $|\Gamma| \leq 0.1$ at the NMR frequency, either by etching a new set of capacitors or by scratching the capacitors with a scalpel knife. We have found that this way to make the capacitances is essential for the construction of a resonant circuit which is well described by the model consisting of L, r, C_p and C_s.

The use of 0.4 mm Ø copper wire leads to circuits with $Q \approx 1000$ at 4.2 K. Such a quality factor, which is compatible with the dielectric losses of Kapton at 77 K, is too high for our experiments. Brass wire (0.4 mm Ø) leads to $150 < Q < 250$ with the additional advantage that the tuning does not change very much in the temperature range 4.2 - 300 K. Because of the good control over and the reproducibility of the manufacturing of these resonating circuits, they are a viable alternative with respect to split-ring resonators [6] at these frequencies.

The 50 Ω coaxial lines running down from T = 300 K to T ≈ 10 mK at a field of 7 or 12 T are well characterized. We mounted 2.2 mm Ø coaxial cable (stainless steel or BeCu) with one end terminating in the experimental room and the other side in the vacuum can at 4.2 K without any precaution against leaks. From 4.2 K to lower temperatures we use home made coaxial cables with a characteristic impedance of 55 Ω, a wave velocity of 2.355×10^8 m/s and an attenuation coefficient of $0.0054 f^{0.5}$ m^{-1} with the frequency f in MHz. The cable is made out of a 0.22 mm Ø superconducting inner conductor, a 0.8 × 1.0 mm Ø CuNi outer conductor and a 0.4 × 0.7 Teflon tube as insulation. In the high field region of the 12 T magnet we use commercial 1.7 mm Ø copper coaxial cable.

5. CONCLUSION

The reflection coefficient in the ^{4}He storage vessel and in the experimental setups agree very well. Although experiments using the techniques described above are still in a preliminary stage, the NMR signal at small polarization and the calculations agree; we are confident that the nonlinear response is under control.

ACKNOWLEDGMENTS

We thank Olivier Buisson for the time he spent to instruct us in the use of the network analyzer and to discuss the measurements.

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