Supercond. Sci. Technol. **21** (2008) 015023 (4pp) doi:10.1088/0953-2048/21/01/015023

N-14 NQR using a high-T_c rf SQUID with a normal metal transformer

D F He[1,3], M Tachiki[1] and H Itozaki[1,2]

[1] SQUID group, Superconducting Materials Center, National Institute for Materials Science, Sengen 1-2-1, Tsukuba, Ibaraki 305-0047, Japan
[2] Graduate School of Science Engineering, Osaka University, 1-3 Machikaneyama, Toyonaka, Osaka 560-8531, Japan

E-mail: he.dongfeng@nims.go.jp

Received 11 September 2007, in final form 2 December 2007
Published 20 December 2007
Online at stacks.iop.org/SUST/21/015023

Abstract
We have improved our high-T_c SQUID-based N-14 nuclear quadrupole resonance (NQR) detection system. By using a reed relay SIL5-1A72-71L (MEDER electronic) in the normal metal transformer, the isolation to the excitation field was much improved. The high-T_c rf SQUID could operate stably when the rf excitation field was over 20 mT. By optimizing the input coil of the transformer, better sensitivity of 0.5 fT Hz$^{-1/2}$ was obtained at the resonant frequency of the tuned normal metal transformer. A spin-locking spin-echo (SLSE) multi-pulse sequence was also used in the system to detect the weak NQR signals produced by samples with longer spin–spin relaxation time T_2, such as trinitrotoluene (TNT).

1. Introduction

Nuclear quadrupole resonance (NQR) of N-14 is a promising method to detect explosives in baggage or landmines. Low-T_c direct current superconducting quantum interference devices (DC SQUIDs) have been used as sensors for low frequency nuclear magnetic resonance (NMR) and NQR [1–4]. We previously used a tuned normal metal transformer to improve the sensitivity of the high-T_c SQUID [5], and successfully detected the N-14 NQR signal of p-nitrotoluene (PNT) [6].

In this paper, we will report the improvements of our SQUID-based NQR system. The transformer was optimized and the spin-locking spin-echo (SLSE) method was used. The signal-to-noise ratio of the NQR detection was much improved, and the weak NQR signal of trinitrotoluene (TNT) was also successfully detected.

2. Improving the switch circuits of the normal metal transformer

Very strong rf fields with the amplitude of several mT are necessary to excite NQR signals, especially for remote sensing. However, the very small leakage of the excitation field will influence the stability of the SQUID. For explosive detection, the N-14 NQR frequencies of explosives are normally high (∼MHz), so it is not easy to isolate the strong excitation field. To reduce the influence of the strong excitation field on the operation of the SQUID, switch circuits were inserted in the normal metal transformer [6]. Figure 1(a) shows the switch circuit we used before [6]. A photo CMOS relay (AQV225N) was used in the switch circuit. For the excitation rf pulse, SW1 was ON and SW2 was OFF, and the strong excitation field was isolated. For the NQR signal, SW1 was OFF and SW2 was ON, and the NQR signal could be coupled to the SQUID through the input coil of the transformer. The ON resistance of the photo CMOS relay was 7 Ω. Even with six CMOS relays in parallel, the 1.2 Ω ON resistance was still too big. The big ON resistance of the switches reduced the Q-value of the transformer, and then reduced the sensitivity of the system. In the meantime, the isolation of the CMOS relay to the rf field was also not so good. When the excitation field was bigger than 0.5 mT, the SQUID became unstable, and the NQR signal could not be detected.

To increase the isolation to the strong excitation field, we improved the switch circuit, as shown in figure 1(b). For the excitation rf pulse, SW1 was ON, the other switches SW2, SW3, SW4 and SW5 were OFF, and the strong excitation field was isolated. For the NQR signal, SW1 was OFF and the other switches were ON, and the NQR signal could be coupled to the SQUID. To get high isolation of the excitation field and high Q-value of the transformer, the choice of switch was also important.

[3] Author to whom any correspondence should be addressed.

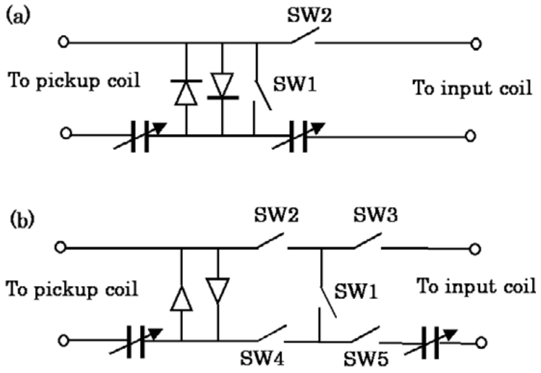

Figure 1. (a) The switch circuit used previously. (b) The improved switch circuit.

Table 1. The comparison of switches: mechanical relay, CMOS relay, and analog switch.

	SIL5-1A72-71L	AQV225N	ADG821
R_{ON} (Ω)	0.4	7	0.8
R_{OFF} (kΩ)	30	0.3	1
T_{ON}	0.3 ms	0.2 ms	50 ns
T_{OFF}	0.1 ms	0.08 ms	20 ns
I_M (A)	1	0.4	0.2
V_B (V)	4200	80	7

Table 1 shows the comparison of several kinds of switches. SIL5-1A72-71L is a reed relay (MEDER electronic). AQV225N is a photoMOS relay. ADG821 is an analog switch. R_{ON} in table 1 is the contact resistance when the switch is ON, R_{OFF} is the isolation resistance of the switch when the switch is OFF. R_{ON} and R_{OFF} are measured at 890 kHz. T_{ON} is the delay time when the switch is turned ON, and T_{OFF} is the delay time when the switch is turned OFF. I_M is the maximum carrying current when the switch is ON. V_B is the breakthrough voltage when the switch is OFF.

From table 1, the reed relay SIL5-1A72-71L has very small ON resistance and much bigger OFF resistance; therefore we used it in the transformer for the NQR detection of PNT.

The isolation to the excitation rf field was much improved. For the previous switch circuit, the isolation was about 60 dB; for the switch circuit with the reed relay, the isolation was better than 90 dB. The SQUID could operate stably when the amplitude of the excitation rf field was over 20 mT, which was 40 times better than before.

3. Optimizing the input coil

The input coil of the transformer has a large influence on the sensitivity of the system. To achieve the best sensitivity, the mutual inductance between the input coil and the SQUID should be large and the loss of the input coil should be small. The loss of the input coil at rf frequency can be equivalent to a resistance, which can be directly measured by an LCR measurement device. Figure 2 shows the experimental equipment to measure the mutual inductance between the SQUID and the input coil. A high-T_c YBCO rf SQUID with a 1 cm $SrTiO_3$ substrate resonator was used [7]. The white flux noise was about 15 $\mu\Phi_0$ $Hz^{-1/2}$ and the magnetic field sensitivity was about 50 fT $Hz^{-1/2}$. The SQUID and the input coil were put in a small cylinder magnetic shielding. A current of 1 MHz was applied to the input coil through a signal generator and a resistance network. From the SQUID response, we could calculate the mutual inductance between the SQUID and the input coil. We tried many input coils with different turns, different diameters and different copper wire diameters, and found that the coil using 0.2 mm copper wire, with 20 turns and 1 cm diameter had the best performance. The mutual inductance between the input coil and the SQUID was about 500 pH. The equivalent resistance was about 1.2 Ω at the frequency of 1 MHz.

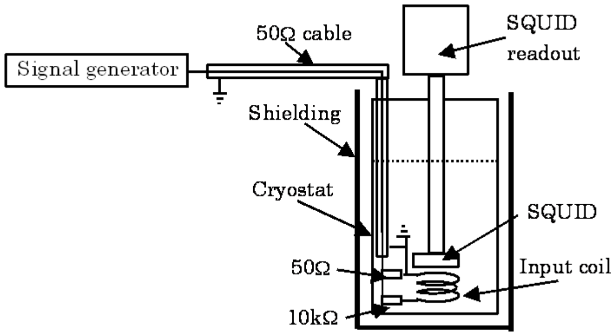

Figure 2. The method to measure the mutual inductance between the SQUID and the input coil.

The input coil L1 was wound using Litz wire with a diameter of 2 mm, which has smaller AC loss than normal copper wire. The coil L1 was 16 turns with a diameter of 10 cm. Using the formula given in [5], we can estimate the magnetic field sensitivity of the high-T_c rf SQUID with the transformer. It was about 0.2 fT $Hz^{-1/2}$ at 1 MHz. Due to the mismatching between the pickup coil and the input coil and AC loss of the transformer, the measured sensitivity was worse than this estimated value. It was about 0.5 fT $Hz^{-1/2}$ around 1 MHz.

We used a 500 g sample of p-nitrotoluene (PNT) powder to check our SQUID-based NQR system. Figure 3(a) shows the free induction decay (FID) of the N-14 NQR signal for PNT after 200 averages. The sample was at room temperature. The pulse applied to the sample is at 890 kHz with the duration of 150 μs, a peak-to-peak amplitude of about 0.3 mT, and a repetition rate of 1 Hz. Figure 3(b) shows the Fourier transform of the FID. The frequency of N-14 NQR in PNT was about 887.1 kHz. For the same measurement time, the signal-to-noise ratio was about four times better than before.

4. Phase cycling SLSE for the SQUID-based NQR system

The multi-pulse technique of SLSE was proposed and used to improve the signal-to-noise ratio of NQR detection by Marino and Klainer [8]. Now it is widely used for N-14 NQR [9–11]. To detect the weak NQR signal of TNT, we used the phase cycling spin-locking spin-echo in our SQUID-based NQR system.

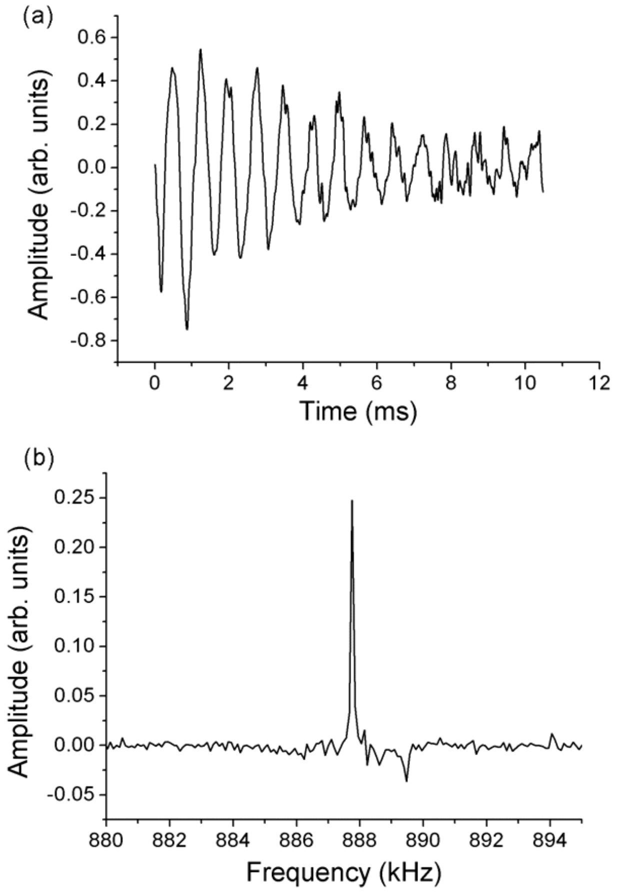

Figure 3. (a) The free induction decay (FID) of the N-14 NQR signal for PNT after 200 averages. (b) The Fourier transform of the FID.

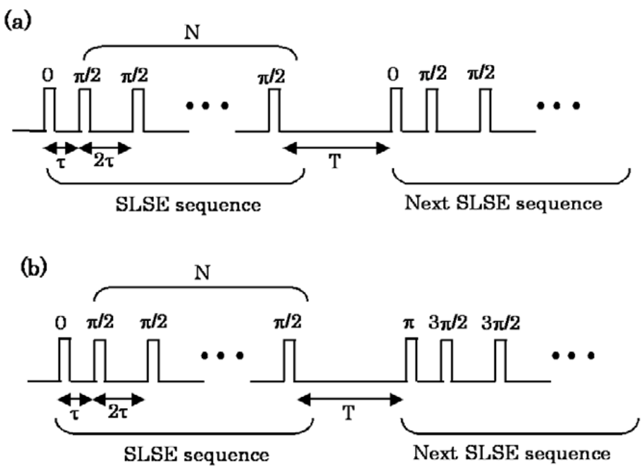

Figure 4. (a) The SLSE pulse sequence. (b) The phase cycling SLSE pulse sequence.

Figure 4(a) shows the SLSE sequence; it can be represented as

$$\theta^0 - (\tau - \theta^1 - \tau)_N. \quad (1)$$

The first pulse is a preparatory pulse with the flipping angle of θ^0. The flipping angle θ^1 of the following pulses is phase shifted by 90° in relation to the first preparatory pulse.

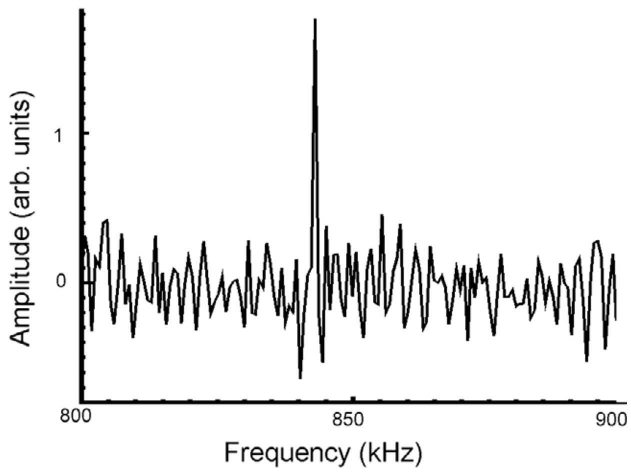

Figure 5. The Fourier transform of the N-14 NQR signal for 300 g TNT detected by the high-T_c rf SQUID.

The time interval between the preparatory pulse and the second pulse is τ, and it is 2τ for the refocusing pulses. After an SLSE sequence, there is a waiting time T, then another SLSE sequence is started. By adding all the NQR signals produced by SLSE sequences, the signal-to-noise ratio of NQR detection can be increased.

To reduce the background signal or the spurious noise, a phase cycling method was used. Figure 4(b) shows the phase cycling pulse sequence in our SQUID-based NQR system. It can be represented as

$$\theta^0 - (\tau - \theta^1 - \tau)_N - T - \theta^0_\pi - (\tau - \theta^1_\pi - \tau)_N. \quad (2)$$

The adjacent SLSE pulse sequence has 180° phase difference and the waiting time between the SLSE sequences is T.

300 g powder TNT was used. The excitation frequency was 0.843 MHz. The rf pulse width was 200 μs. The time interval τ was 0.7 ms. For each SLSE sequence, the echo number was 50. The waiting time between the SLSE sequences was 10 s. The total measurement time was about 20 min. Figure 5 shows the Fourier transform of the N-14 NQR signal for the 300 g TNT detected by the high-T_c rf SQUID.

5. Summary

By improving the switch circuit in the transformer, higher excitation power can be used, and the SQUID could work stably. By optimizing the input coil of the transformer, higher sensitivity of the system was obtained. By using the phase cycling SLSE method in the SQUID-based NQR system, the weak NQR signal of TNT could be detected. We are still improving our system to obtain better results.

Acknowledgments

This research was supported by Japanese Science and Technology (JST) on the project of landmine detection. We also acknowledge help from Mr G Ota.

References

[1] Fan N Q and Clarke J 1991 Low-frequency nuclear magnetic resonance and nuclear quadrupole resonance spectrometer based on a DC superconducting quantum interference device *Rev. Sci. Instrum.* **62** 1453–9

[2] Hürlimann M D, Pennington C H, Fan N Q, Clarke J, Pines A and Haln E L 1992 Pulsed Fourier-transform NQR of ^{14}N with a DC SQUID *Phys. Rev. Lett.* **69** 684–7

[3] Yesinowski J P, Buess M L and Garroway A N 1992 Detection of N-14 and Cl-35 in cocaine base and hydrochloride using NQR, NMR and SQUID techniques *Anal. Chem.* **67** 2256–63

[4] Tonthat D M and Clarke J 1996 Direct current superconducting quantum interference device spectrometer for pulsed nuclear magnetic resonance and nuclear quadrupole resonance at frequencies up to 5 MHz *Rev. Sci. Instrum.* **67** 2890–3

[5] He D F, Itozaki H and Tachiki M 2006 Improving the sensitivity of a high-T_c SQUID at MHz frequency using a normal metal transformer *Supercond. Sci. Technol.* **19** S231–4

[6] He D F, Tachiki M and Itozaki H 2007 ^{14}N Nuclear quadrupole resonance of p-nitrotoluene using a high-T_c rf SQUID *Supercond. Sci. Technol.* **20** 232–4

[7] Zhang Y, Schubert J, Wolters N, Banzet M, Zander W and Krause H J 2002 Substrate resonator for HTS rf SQUID operation *Physica* C **372–376** 282–6

[8] Marino R A and Klainer S M 1977 Multiple spin echoes in pure quadrupole resonance *J. Chem. Phys.* **67** 3388–9

[9] Rudakov T N, Mikhaltsevich V T and Selchikhin O P 1997 The use of multi-pulse nuclear quadrupole resonance techniques for the detection of explosives containing RDX *J. Phys. D: Appl. Phys.* **30** 1377–82

[10] Rudakov T N and Mikhaltsevich V T 2002 Spin locking spin echo in nitrogen-14 quadrupolar spin-system with axially symmetric electric field gradient tensor *Chem. Phys. Lett.* **363** 1–6

[11] Rudakov T N and Mikhaltsevich V T 2003 Multiple NQR spin echoes in phase cycled pulse experiments *Phys. Lett.* A **309** 465–9