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Nuclear magnetic resonance magnet actively cooled by pulse tube refrigerator

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High field NMR spectrometers have been an essential tool for biomolecular scientists for many years. They have been instrumental in the pursuit of understanding of the structure, function and dynamics of proteins and other biological molecules. In addition, NMR is increasingly used for small molecule applications such as metabonomics, providing capabilities that aid drug discovery, as well as general organic and inorganic chemistry [M. Pellecchia *et al.*, Nature Reviews Drug Discovery **1**, 211 (2002)]. However, access to these systems is restricted due to the requirement to periodically refill them with liquid cryogens. This is both logistically demanding and expensive. A new system combining NMR spectrometry and Pulse Tube Refrigeration (PTR) has been developed and successfully tested. This approach eliminates the dependence on liquid cryogens, reduces spectrometer downtime, and also significantly reduces the size of the system. In the near future this new type of analytical tool may become ubiquitous in biomedical and chemical laboratories. [DOI: 10.1063/1.1896949]

I. INTRODUCTION

While NMR magnets are an essential tool for biomolecular scientists, access to them is restricted due to the requirement to have them periodically refilled with liquid cryogens by trained personnel. In addition, systems cannot be operated properly during cryogen transfer due to instrument instability. The cost of the helium itself is also an increasingly important issue. The helium gas is produced mainly by separation from natural gas. Deposits are limited while demand from both industry and scientific research has increased significantly in the past few years. As a result, there is a shortage of supply and significant price rises by helium gas producers.[1] If the trend continues the helium price could increase about seven times during the next decade.

II. SYSTEM DESIGN

The newly developed system presented in this paper makes possible the "active cooling" of a 400 MHz NMR superconducting magnet by a Pulse Tube Refrigerator (PTR).[2] This approach eliminates the dependence on liquid cryogens, reduces spectrometer downtime, and significantly reduces the size of the system. The design that has been employed avoids distortion of the NMR spectrum by the vibration[3,4] and magnetic effects of the PTR. In addition, the operation of the system is in many respects the same as with conventional systems, including the presence of a reservoir of liquid helium that provides temperature uniformity and stability, together with the ability to tolerate short duration power failures. A photograph of the system is presented in Fig. 1.

The NMR system cryostat utilizes two-stage 4 K pulse tube technology. The cooling effect is produced through the periodic pressure variation and displacement of helium gas in the pulse tube. Conventional coolers based on the Gifford–McMahon (GM) cycle, generate high levels of mechanical vibration and distort magnetic fields due to the displacement of magnetic materials contained in a moving regenerator-displacer. A unique feature of the PTR technology is the absence of cold moving parts, which considerably reduces the vibration of the cold head and increases reliability. However vibrations are still generated by the helium pressure oscillation in the PTR. As it was shown,[5] the vibration of the room temperature part of the PTR cold head is two orders of magnitude smaller than that of the GM. At the same time the vibrations of the cold stages for both types were of the same order of magnitude. A major challenge was therefore the delivery of cooling power from the cold stages of the PTR to the magnet with a level of vibration tolerable in NMR.

An unconventional and innovative solution for the control of PTR vibration was found. A schematic of the cryostat is presented in Fig. 2. The two-stage PTR (SRP-052A Sumitomo Heavy Industries Ltd.) marked on the schematic by (1) was suspended in helium gas without any direct mechanical contact with the magnet (2). The cooling power of the PTR's second stage (3) was transported to the magnet by a "rain" of condensed helium. Gaseous helium passed from the main section of the helium vessel to the heat exchanger (5) attached to the second stage of the PTR where it was condensed and returned as a liquid to the main section. The cooling power transport was found to be so effective, that the maximum temperature gradient between magnet and cold head was less than 100 mK. At the same time the effective transport of cooling power from the first stage (4) of the PTR to a nitrogen vessel (6) through copper braids (7) enabled the use of the empty nitrogen vessel as a 50 K radiation shield. Future systems will need only a radiation shield, eliminating the nitrogen vessel, thus reducing the diameter of the cryostat. The diameter of a prototype 600 MHz magnet cryostat was reduced by 20%.

FIG. 1. (Color online) 400 MHz NMR cryostat actively cooled by a Pulse Tube Refrigerator.

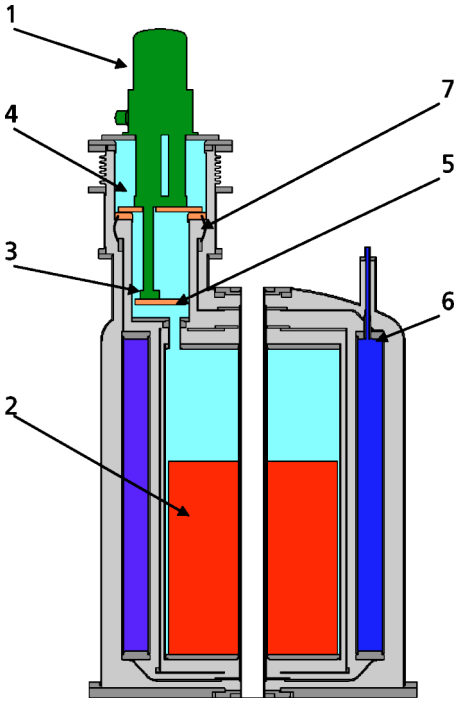

FIG. 2. (Color online) System schematic: (1) PTR, (2) magnet, (3) second stage of PTR, (4) first stage of PTR, (5) heat exchanger (condenser), (6) 50 K-radiation shield, (7) copper braids.

The solution that has been described exposed both stages of the PTR to helium gas at atmospheric pressure. In this situation, there was the risk of a significant reduction in cooling power, due to heat-exchange caused by helium gas convection between the pulse and regenerator tubes of the PTR, and also between the cold parts of the PTR and its enclosure. In order to minimize this convection, the PTR was surrounded by Styrofoam baffles. The design and efficiency of these baffles will be the subject of a future paper.

III. MEASUREMENTS AND TEST DATA ANALYSIS

Figure 3 shows the variation of PTR first and second stage temperatures over time. A simple system comprising a temperature sensor and heater was used to control the rate of condensation at the heat exchanger. The level of liquid helium remained constant during a seven-day test. It should be noted that the second stage temperature was controlled at a constant 4.1 K in order to keep the pressure of the helium gas in the vessel close to atmospheric. The estimated heat-load to the first stage of PTR was about 20–25 W and to the second stage was about 0.3 W.

An additional benefit of this design is that it allows the magnet to stay at field for a long period even after power or PTR failure. It was shown in a test that only 11% of the liquid helium was lost during the first 12 h after the PTR was switched off. There was no liquid nitrogen in the nitrogen vessel during the test.

Another potential risk that was considered was the distortion of the magnetic field homogeneity by the second stage regenerator material of the PTR. The perturbation of the magnetic field by the PTR in the NMR sample space consists of permanent and time-dependent components. The permanent component was taken into account by magnet shimming, however, the time-dependant component remained an important issue and was the subject of further investigation. The temperature of the PTR regenerator material oscillates and since the magnetic susceptibility of the material is strongly temperature dependent[6] it induces an oscillating magnetic field component. In order to minimize the influence of this component on the homogeneity of the magnetic field, the position of the PTR was optimized for minimum stray field.

To investigate the possibility of magnetic coupling between the NMR sample and magnetic material contained within the second stage regenerator of the PTR, two NMR spectrum measurements were performed. The first measurement was obtained with a second stage temperature of 4.1 K. A second measurement held the temperature of the second stage at 20 K. At 20 K the magnetic susceptibility of the regenerator material is just 10% of the susceptibility value at 4.2 K and therefore if magnetic coupling were present, then a difference in the spectra at the two temperatures would be expected. In the experiment no difference between the NMR spectra was detected. It can therefore be concluded that in this case the magnetic coupling was negligibly small.

An array of NMR experiments were acquired to evaluate the effect of the PTR on routine spectra, such as those typically required by chemists performing structure identification or verification. The results obtained are shown in Figs. 4 and 5. As can be seen from the figures, the NMR spectra generated while the PTR was running are comparable in resolution to those generated on conventional systems, indicating effective vibration isolation.

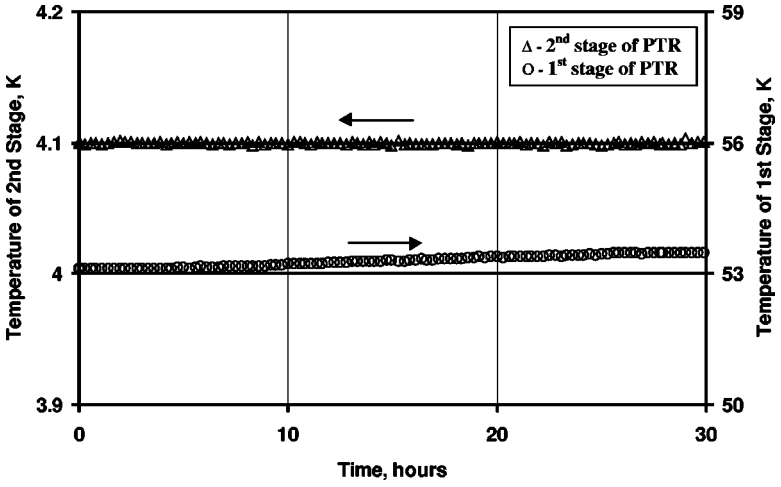

FIG. 3. Results of the system test in zero helium boil-off regime.

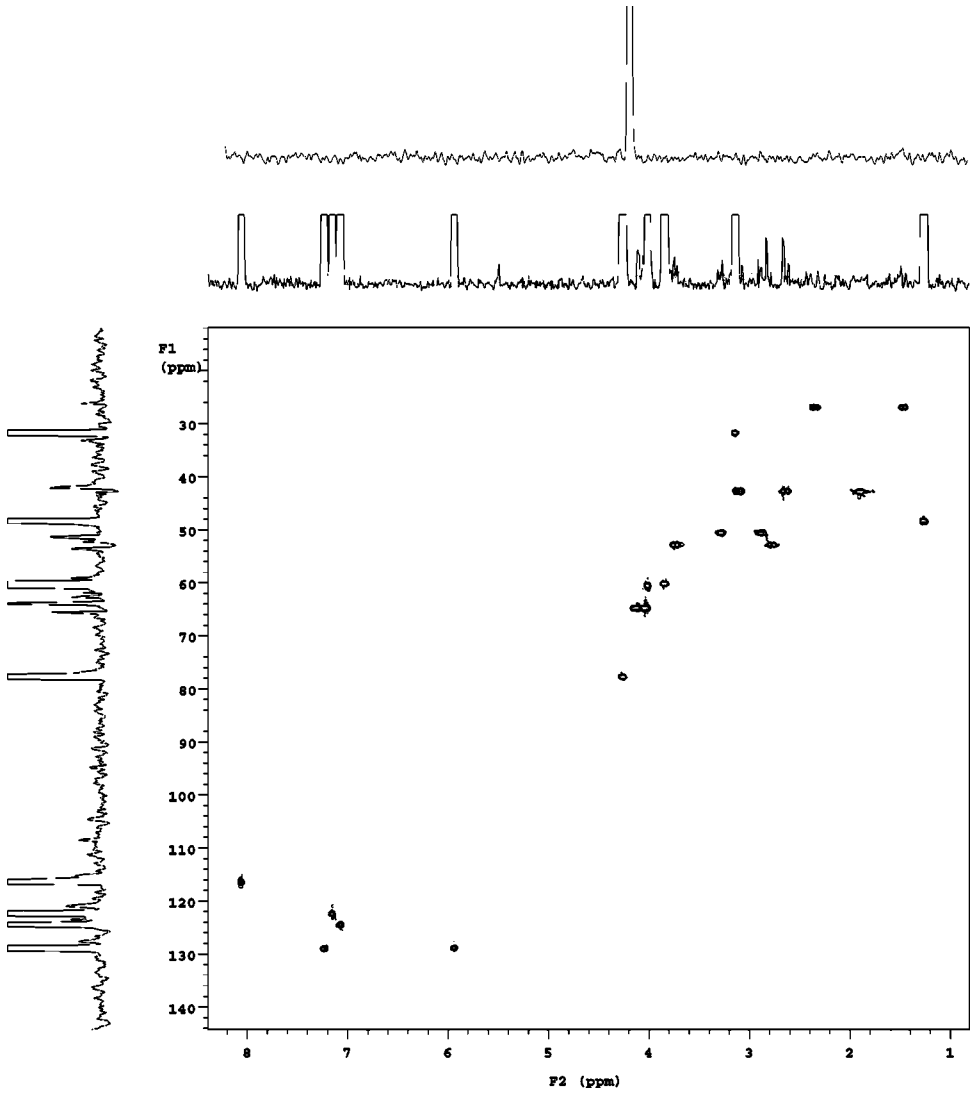

FIG. 4. PTR on gHSQC of 10 mg strychnine in $CDCl_3$ ≈10 min. Varian UnityINOVA 400 MHz, Oxford Instruments ActivelyCooled™, Actively Shielded Magnet, triple resonance PFG probe.

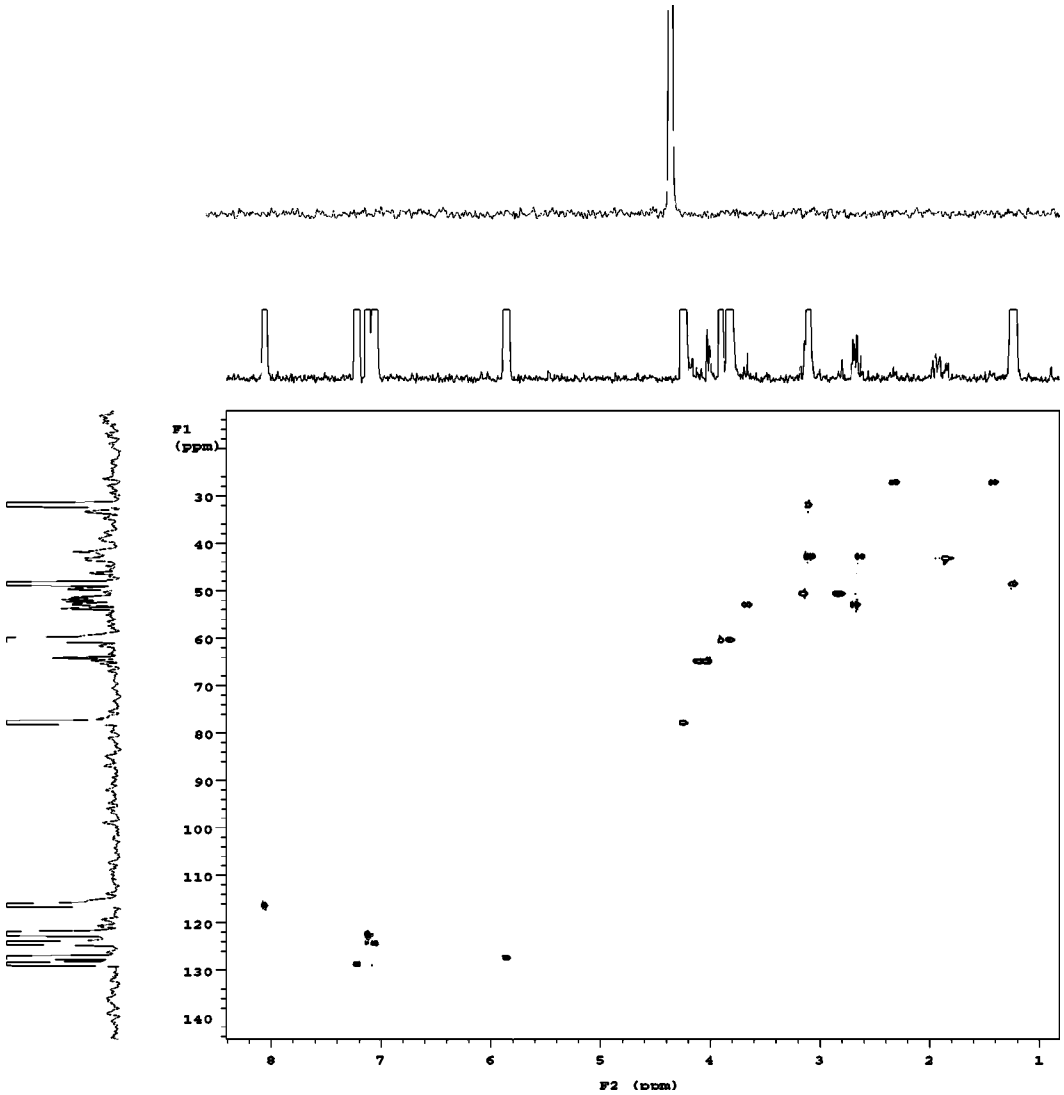

FIG. 5. As Fig. 4 but with PTR off.

ACKNOWLEDGMENTS

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[1] J. Lesser, B. Ziegler, and S. Ebner, Economical aspects on helium recovery due to increased demand of helium gas and shortage of supply *IIR International Conference CRYOGENICS 2002*, Prague, Czech Republic, Biii. Rare Gases, 2002.

[2] Preliminary test results of first in the world NMR spectrometer actively cooled by PTR were presented at Varian User Meeting, Stanford University, California, USA, April 2004.

[3] A. F. Mehlkopf *et al.*, J. Magn. Reson. (1969-1992) **58**, 315 (1984).

[4] P. J. Bowyer *et al.*, J. Magn. Reson. **152**, 234 (2001).

[5] T. Tomaru *et al.*, Cryogenics **44**, 309 (2004).

[6] C. Wang and P. E. Gifford, "Performance characteristics of a 4 K pulse tube in current applications," ICC13 Proceedings (to be published).