Design Evaluation of Serial and Parallel sub-mK Continuous Nuclear Demagnetization Refrigerators

David Schmoranzer · James Butterworth · Sébastien Triqueneaux · Eddy Collin · Andrew Fefferman

Received: date / Accepted: date

Abstract We present the evaluation of two different design configurations of a two-stage PrNi$_5$ continuous nuclear demagnetization refrigerator. Serial and parallel configurations of the two stages are considered, with emphasis on the attainable cooling power at sub-mK temperatures and the impact of the design choices on the operation of the refrigerator. Numerical simulations of heat transfer in the setup are used to evaluate the performance of the refrigerator as well as the technological requirements for the essential thermal links. In accord with similar findings for adiabatic demagnetization refrigerators [Shirron, Cryogenics 62, 2014], our simulations show that the performance of both configurations improves as the thermal links improve, and that the parallel configuration yields a higher cooling power than the series design for a given thermal link resistance and sample temperature.

Keywords ultra low temperatures · adiabatic nuclear demagnetization · continuous refrigeration techniques · PrNi$_5$

1 Introduction

Recently, significant effort was devoted to the development of a novel ultra-low temperature refrigerator [1][2] which would be able to overcome the limitations of traditional nuclear demagnetization setups [3][4] and continually maintain sub-mK temperatures. Such a continuous nuclear demagnetization refrigerator (CNDR) would find use in ultra-low temperature research of numerous physical phenomena such as superfluidity of $^3$He [5][6][7], quantum states of...
macroscopic objects \[8, 9\], or dissipation in amorphous matter \[10, 11, 12, 13\] or modern materials such as graphene \[14\]. The main factor limiting the use of traditional nuclear demagnetization setups is the fact that at sub-mK temperatures, the duration of the experimental window is often less than the thermal relaxation times of the materials of interest \[4\] (spin-lattice relaxation in non-conducting materials, relaxation processes in amorphous materials, heat exchange between metal walls and liquid helium), which precludes proper sample thermalization. This issue becomes crucial if the demagnetization setup is mounted on a dry dilution refrigerator, as these typically have higher intrinsic heat leaks due to vibrations from the pulse tube assembly \[15\], but even on “wet” systems, the thermal cycling might be disruptive for precise measurements. Additionally, the ability to maintain sub-mK temperature indefinitely may provide an important stepping stone for the development of further cooling techniques and eventually lead to the experimental realization of novel states of matter, such as the dual Bose-/Fermi-type superfluidity in \(^3\)He-\(^4\)He mixtures \[16\].

Previously, similar designs of a continuous (electronic) adiabatic demagnetization refrigerator (CADR) were discussed in detail in the literature \[17, 18\], and a review may be found in Ref. \[19\]. The design presented below builds on the above-mentioned work and extends the temperature range of the refrigerator to sub-mK temperatures by replacing paramagnetic salt pills with a suitable nuclear magnetic refrigerant, in our case PrNi\(_5\). The fundamental principle of operation of the CNDR is the same as for CADR, except that it is the entropy of nuclear spins rather than of outer-shell electronic spins that is manipulated using the external magnetic field.

The construction of the CNDR requires the use of at least two nuclear demagnetization stages with separate superconducting solenoids providing the magnetic field. The stages have to be connected to each other and/or to the sample space via thermal links consisting of metal wires/blocks and superconducting heat switches. Gas-gap heat switches are unfeasible at temperatures below 10 mK. In previous work on CNDR \[1, 2\] it was shown that thermal links are indeed the crucial elements of the entire setup, requiring the use of highest purity metals prepared using specialized treatments and contacting techniques. Specifically, it was demonstrated by means of numerical simulations \[1, 2\] that the equivalent electrical resistance (from the well-known Wiedemann-Franz law) of the thermal link between the two demagnetization stages in a series configuration should be comparable to 150 nΩ or less to attain a cooling power of 20 nW at 1 mK, see also Fig. 6 further below.

Practically, to obtain such a low resistance, one would need to use 5N or 6N purity aluminium blocks (RRR over 5000 was obtained in CNRS Grenoble on 6N aluminium), 5N or better copper wires annealed under oxygen (RRR over 10000 was obtained) or silver wires of equal purity as well as specialized contacting techniques (stable Al to Cu contacts were made with contact resistances of 30 nΩ or less on the area of several cm\(^2\)). Another crucial issue is the contact to PrNi\(_5\) and the purity of the nuclear refrigerant (RRR values near 30 have been reported \[20\]). The technical developments and the details
of processes used in the construction of the CNDR setup represent a separate topic and will be published elsewhere. The necessary material properties are mentioned solely to justify the values of the model parameters and link them to the ongoing development of the refrigerator.

In this manuscript, we use numerical simulations to compare two fundamentally different design approaches to the construction of the CNDR. Namely, the already discussed series configuration [1, 2] is contrasted to the parallel configuration of the two demagnetization stages. Design considerations and the thermal models for both configurations are presented and, subsequently, numerical simulations similar to the calculations in Ref. 2 are used to provide quantitative comparison of the performance of the CNDR in these two configurations, with special attention devoted to the quality of the thermal links used.

2 CNDR Design and Operation

Both principal design configurations have already been discussed for CADR in Ref. 19 and are illustrated for CNDR in Fig. 1. In the series configuration (S-CNDR), the first of the nuclear demagnetization stages (NDS-1) is connected to the mixing chamber plate (MXC) of a dilution refrigerator via a superconducting heat switch (HS-1). The second stage (NDS-2) is directly linked to the sample space (load) and is separated from the first NDS via another heat switch unit (HS-2). The stages may be designed asymmetrically, with NDS-2 containing a lower quantity of the nuclear refrigerant than NDS-1, or with a different nuclear refrigerant altogether.

The operation of the S-CNDR consists of three main steps and can be described as follows (see Fig. 2). Assuming steady-state operation, we start with NDS-2 at the sample temperature and at a moderate magnetic field (of order 60 mT), and thermally decoupled from NDS-1, which is at low temperature and minimum magnetic field (≈10 mT).

Step 1: a slow demagnetization is started on NDS-2 (Fig. 2a) to keep the sample temperature stable and offset any external heat leaks, while NDS-1 is magnetized to full magnetic field (of order 1 T) and warms up (Fig. 2A1). Eventually, it is thermally coupled to the MXC and let to exchange heat (Fig. 2A2), transferring its excess entropy to the MXC.

Step 2: once the final magnetic field and a temperature sufficiently close to the MXC are reached, NDS-1 is decoupled from the MXC and its demagnetization begins (Fig. 2B), aiming for a temperature close below that of the sample space. Step 3: once the temperature of NDS-1 drops below that of NDS-2 (this should coincide with the end of the slow demagnetization of NDS-2), the two stages are thermally linked via the HS and heat exchange between them takes place (Fig. 2C), leading to transfer of entropy from NDS-2 to NDS-1. During this time, the demagnetization of NDS-1 may continue for some time until the minimum field (≈10 mT) is reached, while NDS-2 undergoes a magnetization to its starting value of the magnetic field (Fig. 2b) accompanied with a thermal relaxation to NDS-1 temperature.
As the performance-limiting factor is the heat transfer rate between the two stages, it is advantageous to perform the magnetization of NDS-2 in the initial phase of this step, and thus increase the total heat transferred by inducing a higher temperature difference between NDS-2 and NDS-1.

On the other hand, the parallel configuration (P-CNDR) requires each of the two NDS to be linked to both the MXC and the sample space via separate HS units. In this case, both NDS should be designed symmetrically, with balanced heat capacities. The extra heat switches necessary for the P-CNDR configuration represent some design complications as each HS unit requires its own small superconducting solenoid which must be mounted on the dilution refrigerator and operated independently. When choosing between S-CNDR and P-CNDR, one must therefore weigh any benefits found in the performance of the P-CNDR against the increased complexity of the setup and its larger footprint on the dilution refrigerator.

The operation of the P-CNDR has two main steps (see Fig. 3) during which either of the NDS is in thermal contact with the sample space and providing the cooling via a slow demagnetization (Fig. 3A), while the other NDS is being “regenerated” at the MXC temperature. The regeneration procedure involves decoupling from the sample and magnetization to the maximum field (Fig. 3B1), heat exchange with the MXC (Fig. 3B2), followed by thermal decoupling and demagnetization to or below the sample temperature (Fig. 3B3), where the newly regenerated NDS will be switched for its counterpart. Based on the comparison with CADR setups [19], one would expect a higher cooling power for the P-CNDR setup than for S-CNDR, as with the two stages in parallel, heat rejection at MXC occurs during a longer part of the operating cycle. Nevertheless, it remains to be seen if other complications, specific to ultra-low temperatures arise due to the increased number of heat switches.
The steady-state operation cycle of the series CNDR. In this design, NDS-1 provides precooling for NDS-2 during its up-magnetization, and rejects entropy at the MXC temperature. Ideally, the loop of NDS-2 should be as narrow as possible to minimize losses and provide a stable temperature, but with realistic durations of each step, a finite span of temperatures is necessary. The span of entropies covered by NDS-2 is determined by the choice of its maximum and minimum magnetic field values.

and thermal links, potentially influencing the overall heat leak in the sample space or introducing additional dissipative processes into the operation of the refrigerator, such as heat switch manipulation.

3 Thermal Models

The thermal models corresponding to both configurations are shown in Fig. 4. In the S-CNDR setup, the thermal link between NDS-1 and NDS-2 (including the heat switch HS-2) is the crucial component of the entire setup [1, 2], as it determines the heat exchange rate at which NDS-2 can be precooled from NDS-1 during its magnetization. The duration of this step, together with the heat leaks involved determine the efficiency of the S-CNDR setup and hence the final temperature it can attain.

The necessary heat exchange between NDS-1 and NDS-2 can be, in principle, shortened if a lower capacity stage is used for NDS-2, with the drawback of having to cycle the entire refrigerator faster in order compensate for the external heat leak. This would in turn lead to faster magnetization and demagnetization rates, increasing eddy current heating. As a consequence, the heat capacity of the second stage (unless taken to extremes) has a relatively weak effect on the overall performance of the S-CNDR, if the durations of each cycle step (and values of magnetic field) are tuned to near-optimum values.
The steady-state operation cycle of the parallel CNDR. While the initial cooldown and the first few cycles would differ slightly for both demagnetization stages, once a steady-state is reached, NDS-1 and NDS-2 follow the same operation cycle (out of phase with each other) and both reject entropy at the MXC temperature. As the rate of entropy rejection per cycle is effectively doubled compared to S-CNDR, a higher cooling power is expected for the P-CNDR configuration.

For the purposes of a direct comparison with P-CNDR, we therefore assume a symmetrically designed S-CNDR system.

In the P-CNDR setup, properties of the thermal links between the NDS stages and the sample space (Fig. 4: wires 1d and 2d) determine the rate of maximum available cooling power at a given temperature. Generally, the sample space will have a significantly lower heat capacity than the demagnetization stages and hence the requirements on the equivalent electrical resistance of the thermal links may be expected to be less stringent than for S-CNDR. Nevertheless, thermal decoupling due to a finite thermal resistance between the sample and the stages will necessarily reduce the refrigeration efficiency, as any heat removed from the sample will generate additional entropy, in excess of the amount that would be generated if the stage and the sample were at the same temperature. This is fully taken into account in the numerical simulations presented below.

To directly compare the two configurations, we consider in each case two identical demagnetization stages, each containing 0.2 mol of PrNi$_5$. The thermal behaviour of the linking elements (wires) is modeled based on an equivalent electrical resistance from the Wiedemann-Franz law, assuming the wires would be made from a high-purity metal such as silver or copper. The same electrical resistance is then used for all the wires.
The heat capacity of the sample space is chosen so to approximate that of 10 g of high-purity Cu at 1 mK and zero magnetic field. Specific heat capacity of $1.15 \times 10^{-5}$ J kg$^{-1}$ is used, in close agreement with the electronic contribution to the heat capacity of Cu given in Ref. 4. We note that with the same heat load and thermal resistance, the simulation results do not change appreciably if a mass of 100 g or 1000 g is used for the sample space instead, as the heat capacity is still significantly lower than that of the nuclear stages. We note that the situation might be different if magnetic field were applied to the Cu sample space (such as for the purposes of implementing an additional demagnetization stage), as the heat capacity would be enhanced by the Schottky-like anomaly of Cu nuclear spins 4.

The thermal model of the heat switches and of the nuclear stages is given in Ref. 2. Additional heat leaks (vibrational heating $Q_{vibr} = 10^{-8}[\text{WT}^{-1}]|B|$, eddy current heating $Q_{eddy} = 0.03[\text{WT}^{-2}s^2]|B|^2$, plus an additional constant heat leak of 2 nW) are considered for each nuclear demagnetization stage in
the same way as in Ref. 2. The numerical computations are again described in detail in Ref. 2 as well as the model of entropy of PrNi$_5$ used throughout this work.

4 Results and Discussion

For the comparison of the performance of the two CNDR configurations, the data from Ref. 2 will be used for the S-CNDR setup, while new results will be presented here for the P-CNDR setup, which will be discussed in more detail.

First, a sample time-trace of the magnetic fields on the two stages and the temperatures of the P-CNDR components are illustrated in Fig. 5 for a 150 nΩ equivalent electrical resistance of the thermal links (modeling two bundles of copper wires and an aluminium heat switch in normal state) and 5 nW sample space heat leak, representing rather favourable conditions (see, e.g., Ref. 21 for comparison).

Similar numerical simulations as those shown in Fig. 5 were performed for the P-CNDR setup operating with different values of the equivalent electrical resistance of the thermal links and different values of sample space heat leak. For each case, steady-state operation was established and the maximum sample temperature within a cycle was obtained. These results are summarized in Fig. 6 and compared to the data of Ref. 2.

The results indicate that the rather stringent requirements for the operation of the S-CNDR setup (thermal links with equivalent electrical resistance comparable to 50 nΩ) are to some extent alleviated in the P-CNDR configuration. Temperatures close to 1 mK should be attainable with P-CNDR even with significantly worse thermal links of 500 nΩ equivalent resistance under a heat load of 20 nW.

5 Conclusions

While one must bear in mind that the simulations do not necessarily describe all experimental facts accurately, our results clearly show that, given the same quality of thermal links, the P-CNDR configuration has superior performance to the S-CNDR design in terms of the final temperature reached at a given heat leak, or in terms of the cooling power attained at a given temperature.

We note, however, that the superior cooling power of the P-CNDR comes at the cost of additional system complexity and reduced stability of sample space temperature, as switching the sample connection between the two stages may introduce significant temperature variations. An additional drawback of P-CNDR is the necessity to operate four heat switches together with their solenoids instead of just two such units in the S-CNDR design, again leading to additional heat leaks and delays.

Finally, specific applications may be better suited to the S-CNDR design. In particular, a very high sample temperature stability may be required, or
the sample could be an additional demagnetization stage with a large heat capacity. Aside from these special cases, we expect the P-CNDR to perform better than the S-CNDR for a given thermal link conductance and the P-CNDR clearly deserves experimental study.

Acknowledgements We acknowledge support from the ERC StG grant UNIGLASS No. 714692 and ERC CoG grant ULT-NEMS No. 647917. The research leading to these results has received funding from the European Unions Horizon 2020 Research and Innovation Programme, under Grant Agreement no 824109.

References

1. R. Toda, S. Murakawa, and H. Fukuyama, Design and expected performance of a compact and continuous nuclear demagnetization refrigerator for sub-mK applications, J. Phys. Conf. Series 969, 012093 (2018)
Fig. 6 Comparison of the final temperature (maximum sample space temperature during a cycle) vs. sample space heat leak for the serial (left) and parallel (right) CNDR setups. The data in the left panel are adapted from Ref. 2. It is clearly shown that the P-CNDR setup can achieve lower temperatures with the same quality of thermal links under the same heat load. Conversely, this implies that at a given temperature (in the mK range), the P-CNDR setup indeed has a higher cycle-averaged cooling power than the S-CNDR setup, as expected.

2. D. Schmoranzer, R. Gazizulin, S. Triqueneaux, E. Collin, A. Fefferman, Development of a sub-mK Continuous Nuclear Demagnetization Refrigerator, J. Low Temp. Phys. 196, 261-267 (2019)
3. O. V. Lounasmaa, Experimental Principles and Methods below 1 K. Academic Press, London and New York (1974)
4. F. Pobell, Matter and Methods at Low Temperatures, third edition. Springer-Verlag Berlin Heidelberg (2007).
5. Mkinen, J.T., Dmitriev, V.V., Nissinen, J. et al., Half-quantum vortices and walls bounded by strings in the polar-distorted phases of topological superfluid 3He., Nat Commun 10, 237 (2019)
6. J. T. Mkinen and V. B. Eltsov, Mutual friction in superfluid 3He-B in the low-temperature regime, Phys. Rev. B 97, 014527 (2018)
7. V. Tsepelin, A.W. Baggaley, Y.A. Sergeev, C.F. Barenghi, S.N. Fisher, G.R. Pickett, M.J. Jackson, and N. Suramishvili, Visualization of quantum turbulence in superfluid 3He-B: Combined numerical and experimental study of Andreev reflection, Phys. Rev. B 96, 054510 (2017)
8. E. E. Wollman, C. U. Lei, A. J. Weinstein, J. Suh, A. Kronwald, F. Marquardt, A. A. Clerk, and K. C. Schwab, Quantum squeezing of motion in a mechanical resonator, Science 349, 952 (2015).
9. C. F. Ockeloen-Korppi, E. Damskgg, J.-M. Pirkkalainen, M. Asjad, A. A. Clerk, F. Massel, M. J. Woolley, and M. A. Sillanp, Stabilized entanglement of massive mechanical oscillators, Nature 556, 478 (2018).
10. X. Liu, D.R. Queen, T.H. Metcalf, J.E. Karel, F. Hellman, Hydrogen-free amorphous silicon with no tunneling states. Phys. Rev. Lett. 113, 025503 (2014)
11. Rogge, S., et al., Nonlinear dielectric response of glasses at low temperature, Phys. Rev. B 55, 11256 (1997)
12. A. D. Fefferman, R. O. Pohl, A. T. Zehnder, and J. M. Parpia, Acoustic properties of amorphous silica between 1 and 500 mK, Phys. Rev. Lett. 100, 195501 (2008)
13. D. Schmoranzer, S. Kumar, A. Luck, E. Collin, A. Fefferman, X. Liu, T. Metcalf, G. Jernigan, Observations on Thermal Coupling of Silicon Oscillators in Cryogen-Free Dilution Refrigerators, J. Low Temp. Phys. 196, 268-274 (2019)
14. X. Liu, T.H. Metcalf, J.T. Robinson, B.H. Houston, F. Scarpa, Shear modulus of monolayer graphene prepared by chemical vapor deposition, Nano Lett. 12, 1013 (2012)
15. S. Riabzev, A. Veprik, H. Vilenchik, and N. Pundak, Vibration generation in a pulse tube refrigerator, Cryogenics 49 (1), 16 (2009)
16. J. Tuoriniemi, Physics at its coolest, Nat. Phys. 12 (1), 11-14 (2016)
17. P. J. Shirron, E. R. Canavan, M. J. DiPirro, J. G. Tuttle, C. J. Yeager, A Multi-Stage Continuous-Duty Adiabatic Demagnetization Refrigerator, in: Shu QS. (eds) Advances in Cryogenic Engineering. Advances in Cryogenic Engineering. Springer, Boston, MA (2000), DOI: 10.1007/978-1-4615-4215-5_86
18. P. Shirron, E. Canavan, M. DiPirro, J. Francis, M. Jackson, J. Tuttle, T. King, M. Grabowski, Development of a cryogen-free continuous ADR for the constellation-X mission, Cryogenics 44, 581-588 (2004)
19. P.J. Shirron, Applications of the magnetocaloric effect in single-stage, multi-stage and continuous adiabatic demagnetization refrigerators, Cryogenics 62, 130-139 (2014)
20. H. R. Folle, M. Kubota, Ch. Buchal, R. M. Mueller, F. Pobell, Nuclear refrigeration properties of PrNi5, Z. Physik B - Condensed Matter 41, 223 (1981)
21. J. M. Parpia, W. P. Kirk, P. S. Kobiela, T. L. Rhodes, Z. Olejniczak, and G. N. Parker, Optimization procedure for the cooling of liquid 3He by adiabatic demagnetization of praseodymium nickel, Rev. Sci.Instrum. 56 (3), 437 (1985)