

Algorithmic cooling in liquid-state nuclear magnetic resonance

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Algorithmic cooling is a method that employs thermalization to increase qubit purification level; namely, it reduces the qubit system's entropy. We utilized gradient ascent pulse engineering, an optimal control algorithm, to implement algorithmic cooling in liquid-state nuclear magnetic resonance. Various cooling algorithms were applied onto the three qubits of ¹³C₂-trichloroethylene, cooling the system beyond Shannon's entropy bound in several different ways. In particular, in one experiment a carbon qubit was cooled by a factor of 4.61. This work is a step towards potentially integrating tools of NMR quantum computing into *in vivo* magnetic-resonance spectroscopy.

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I. INTRODUCTION

The quantum computational model permits algorithms that provide significant—and sometimes even exponential—speedup over any known classical counterpart [1–4]. A rather different scope of that model is to enable improved quantum technologies, e.g., quantum repeaters for communicating secure data over longer distances [5]. Algorithmic cooling (AC), experimentally implemented in this work, is a method that might contribute to both scopes. On the one hand, it was originally suggested as a method for increasing the qubits' purification level [6–11], as qubits in a highly pure state are required both for initialization and for fault tolerant [12,13] quantum computing. On the other hand, the suggested usage of data compression may potentially be found useful for increasing the signal-to-noise ratio of liquid-state NMR and *in vivo* magnetic-resonance spectroscopy [7,14,15].

Nuclear magnetic-resonance quantum computing (NMR-QC) [16–20] commonly uses spin 1/2 nuclei (hereinafter *spins*) of molecules as qubits. Compared to other implementations of small quantum computing devices, liquid-state NMR has an advantage of relatively easy realization of quantum gates by applying rf fields and utilizing spin-spin interactions. However, NMR-QC also has some disadvantages due to working with an ensemble of spins in a mixed state [21,22]; e.g., it is not scalable. Algorithmic cooling, in theory, resolves that problem [6–8].

The thermal energy at room temperature is much higher than the magnetic potential energy of nuclear spins even in the most advanced NMR devices. Therefore, at equilibrium, the qubit ensemble is in a highly mixed state—the probability difference between the “up” and “down” states (hereinafter the *polarization*, denoted as ϵ) is very small. At thermal equilibrium

$$\epsilon = P_{\uparrow} - P_{\downarrow} = \tanh\left(\frac{\Delta E}{2k_{\text{B}}T}\right) \xrightarrow{\Delta E \ll k_{\text{B}}T} \frac{\Delta E}{2k_{\text{B}}T} = \frac{\hbar\gamma B_z}{2k_{\text{B}}T}. \quad (1)$$

Here γ is the gyromagnetic ratio of the spin, B_z is the intensity of the magnetic field, k_{B} is the Boltzmann constant, and T is the bath temperature. When outside of equilibrium, spins with higher polarization than their thermal equilibrium polarization

are considered “cool,” and we can use Eq. (1) to define the spin temperature as $T_{\text{spin}} = \frac{\hbar\gamma B_z}{2k_{\text{B}}\epsilon}$.

Upper bounds on spin cooling (i.e., on polarization enhancement) can be derived by interpreting the spin state in terms of information theory [23]. The information content (IC) of the spin was defined using the conventional notion of Shannon entropy H . The relation between a single spin's polarization and IC is given by the following equation [24,25]:

$$H_{1\text{qubit}} = \left[\frac{1-\epsilon}{2} \ln\left(\frac{1-\epsilon}{2}\right) + \frac{1+\epsilon}{2} \ln\left(\frac{1+\epsilon}{2}\right) \right], \quad (2)$$

$$\text{IC}_{1\text{qubit}} = 1 - H_{1\text{qubit}} = \frac{\epsilon^2}{\ln 4} + O(\epsilon^4).$$

The information content of a spin system is invariant to reversible operations, and therefore bounds the maximal IC a single spin can reach by lossless manipulations, such as quantum gates. This entropy bound, also often called *Shannon's bound*, limits the maximal polarization of a single spin, given an initial thermal state of the spin system.

In our spin system, ¹³C₂-trichloroethylene (TCE, see Fig. 1), consisting of a proton and two ¹³C, the IC at thermal equilibrium is

$$\text{IC}_{\text{eq}} = \frac{\epsilon_{H,\text{eq}}^2}{\ln 4} + 2 \frac{\epsilon_{C,\text{eq}}^2}{\ln 4} = \left[\frac{\gamma_H^2}{\gamma_C^2} + 2 \right] \frac{\epsilon_{C,\text{eq}}^2}{\ln 4} = 17.84 \frac{\epsilon_{C,\text{eq}}^2}{\ln 4}. \quad (3)$$

Shannon's bound dictates that a single spin cannot hold more than the equilibrium information content of the entire spin system:

$$\text{IC}_{\text{eq}} = 17.84 \frac{\epsilon_{C,\text{eq}}^2}{\ln 4} = \frac{\epsilon_{\text{max}}^2}{\ln 4} \Rightarrow \epsilon_{\text{max}} = 4.224\epsilon_{C,\text{eq}}. \quad (4)$$

For convenience, we approximate $\gamma_H/\gamma_C = 4$, and then $\tilde{\text{IC}}_{\text{eq}} = 18$ and $\tilde{\epsilon}_{\text{max}} = 4.24\epsilon_{C,\text{eq}}$.

Algorithmic cooling of spins counterintuitively utilizes the heat bath, that decays polarizations to thermal equilibrium, to cool spins beyond Shannon's bound. AC requires a spin system where some spins, called *reset spins*, thermalize significantly faster than other spins, called *computation spins*. Reversible polarization compression (hereinafter *compression*) is applied

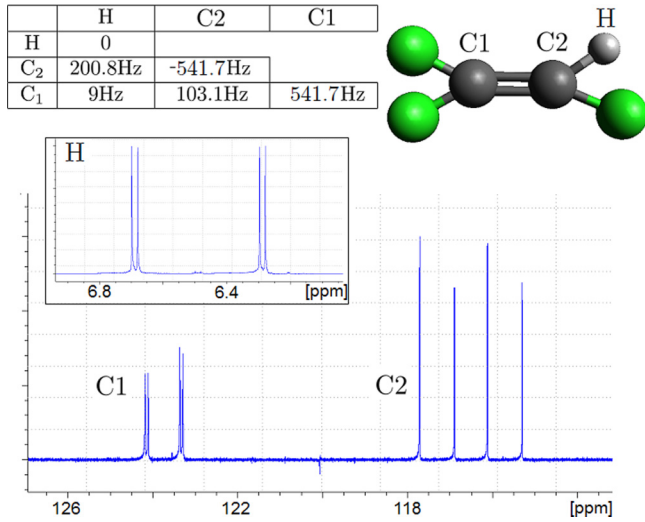


FIG. 1. $^{13}\text{C}_2$ TCE with paramagnetic reagent $\text{Cr}(\text{acac})_3$, in CDCl_3 (chloroform-d) solution. The experiments were performed on a Bruker Avance III 600 spectrometer using a standard 5-mm double resonance probe with a broadband inner coil tuned to ^{13}C and an outer ^1H coil probe. This sample has three active spins marked H, C2, and C1. In the table, the chemical shifts relative to the transmitter frequency are in the diagonal cells, and the J couplings are in the off-diagonal cells. The carbon spectrum is at the bottom, and the proton spectrum is in the small frame. The units of the x axis are parts per million.

to the spin system, transferring some of the computation spins' entropy to the reset spins, which quickly lose some of it to the environment. The process can be repeated, converging the system to a stable trajectory (limit cycle) in the thermodynamic diagram. The efficiency and the cooling limit of AC are (ideally) dependent on the unitary restriction of processes between reset steps, and on the ratio between the relaxation times of the cooled spins and the reset spins.

Various cooling algorithms were developed, following the basic principle presented in [6]. For example, in a three-qubit system with uniform equilibrium polarization ϵ , the initial information content is $\text{IC}_{\text{eq}} = 3\epsilon^2/\ln 4$, the maximum polarization of a single spin that can be reached using unitary transformations [26,27] is 1.5ϵ , and Shannon's bound for the maximal polarization of a single spin is $\sqrt{3}\epsilon = 1.732\epsilon$. However, if one spin has a much shorter thermalization time than the other spins, it will reset while the others retain most of their polarization, so that the entire spin system is cooled. Ideally, iterating the compression process twice leads to a bias of 1.75ϵ , bypassing the result obtained by unitary transformations and even bypassing Shannon's bound [7]. Repeating the process while assuming infinite relaxation time ratios allows enhancing the polarization of one spin asymptotically to 2ϵ [28]. Algorithms applying these processes to n qubits ideally cool exponentially beyond the unitary cooling [6,7], and can be practicable or optimal (see [7,8,25,29–31]).

In TCE, the proton reset spin has higher equilibrium polarization than C1 and C2, the ^{13}C computation spins. In such scenarios, even a special case—AC without compression (called heat bath cooling [15,24]), can cool the spin system

beyond Shannon's bound. This can be done by applying a polarization transfer [32] from the proton to C1 or, alternatively, by swapping the two polarizations via a polarization exchange (PE) gate, and waiting for the proton to regain some of its polarization (while the carbon is still cool). A successive PE from the proton to C2 followed by another waiting period yields polarization of approximately 4 on all three spins, in units of carbon equilibrium polarization. If the relaxation time ratio is sufficiently large and all gates are perfect then $\tilde{\text{IC}}_{\text{total}} \rightarrow 48$, far above the initial approximate value of 18. Further cooling can be achieved using compression.

In practice, heat bath cooling of TCE [24] yielded polarizations {1.74, 1.86, 3.77} for C1, C2, and the proton, respectively, well below the ideal {4,4,4}. Yet, the resulting total IC is 20.70 (± 0.06), which is beyond (and statistically significant) the experimental initial IC (of 17.84) at equilibrium, hence showing for the first time that the Shannon bound can be experimentally bypassed. Heat bath cooling on two amino acids [15] also successfully bypassed Shannon's bound later on. On the other front, experimental work cooling solely by compression was done by Sørensen [26] on methylene chloride and by Chang, Vandersypen, and Steffen [33] on three fluorines in $\text{C}_2\text{F}_3\text{Br}$. Full AC [34] and multicycle AC [35] using solid-state NMR was successfully done at the University of Waterloo.

II. MATERIALS AND METHODS

In order to implement AC and multiple-cycle AC on liquid-state TCE we utilized (following [35]) gradient ascent pulse engineering (GRAPE) [36], an optimal control algorithm, to generate high-fidelity pulses for obtaining the compression gate and the PE gate [37]. Here we present various algorithms for cooling liquid TCE. Process 1 (see Fig. 2), aimed to maximize IC_{C1} , is as follows.

- (1) Wait for duration D2 (H regains polarization).
- (2) PE(H \rightarrow C2).
- (3) Wait for duration D3 (H regains polarization).
- (4) Compression of C1, C2, and H onto C1.
- (5) Return to step 1, unless C1 is saturated.

Ideally, the polarization of C1 is saturated at $\tilde{\text{IC}}_{\text{C1}} \rightarrow 64$. Process 2, aimed to maximize $\text{IC}_{\text{C1,C2}}$, is composed of process 1 followed by a wait step for duration D4, and by PE(H \rightarrow C2) to cool C2, ideally reaching $\tilde{\text{IC}}_{\text{C1,C2}} = 80$ (see Fig. 3).

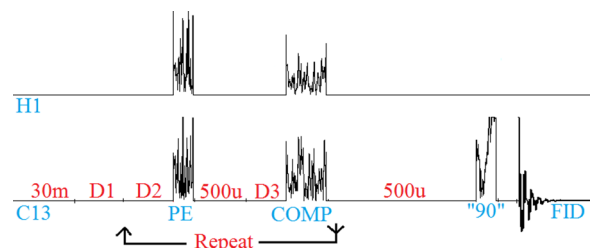


FIG. 2. Visualization of process 1, based on the TopSpin output, showing the rf power vs time in both channels, followed by an acquisition of the free induction decay (FID). The PE and COMP pulses are 6.5 and 13 ms long, respectively, and the delays maximizing the polarization of C1 are $D2 = 5$ s and $D3 = 3$ s. The delay D1 is set to 150 s to equilibrate the system.

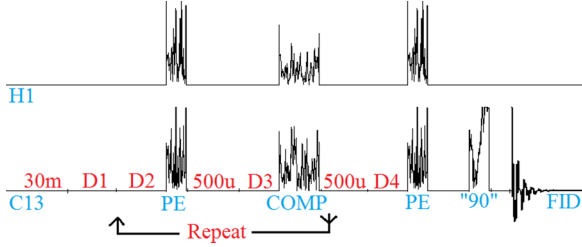


FIG. 3. Visualization of process 2, based on the TopSpin output, showing the rf power vs time in both channels, followed by an acquisition of the free induction decay (FID). The delays maximizing the polarization of C1 are D2 = 5 s, D3 = 3 s, and D4 = 5 s.

The goal of process 3 is to maximize IC_{total} , hence we apply process 2, followed by a wait step for duration D5, ideally reaching $\tilde{IC}_{\text{total}} = 96$ (see Fig. 4). In all cases, a readout pulse was applied on the spin of interest prior to acquisition.

In the experiment, the measured relaxation times (see Table I) were obtained by inversion recovery as in [24,37]. Adding a paramagnetic reagent to the TCE improved the relaxation time ratios as suggested in [38]. We simulated the three processes using the experimental delays and measured relaxation times, while assuming perfect pulses. According to the simulation, the polarization of C1 could be enhanced by a factor of 5.49 after seven rounds ($IC_{C1} = 30.13$, see also Fig. 6) via process 1, and the polarization of the two carbon spins could reach 4.78 and 3.70 ($IC_{C1,C2} = 36.53$) via process 2. The polarization of three spins could reach {3.98, 2.97, 3.75} ($IC = 38.73$) via process 3.

The implemented PE and compression pulses were generated using SIMPSON version 3.0 [39,40], an open source program implementing GRAPE. The pulses were designed to be robust to deviations up to $\pm 15\%$ in rf power [37]. The pulses were not designed to apply a specific unitary gate, but to apply a less constraining state-to-state transformation. However, the state of the system changes with each cooling cycle. Therefore, among two pulses that apply PE, even though one pulse performs better in equilibrium [37], we used another pulse, which yielded better cooling for the entire process. Let $\{\{\dots\}\}$ stand for the diagonal of a density matrix in the computation basis. The PE pulse was designed to evolve the system from equilibrium,

$$4I_z^H + I_z^{C2} + I_z^{C1} \propto \{\{6, 4, 4, 2, -2, -4, -4, -6\}\}, \quad (5)$$

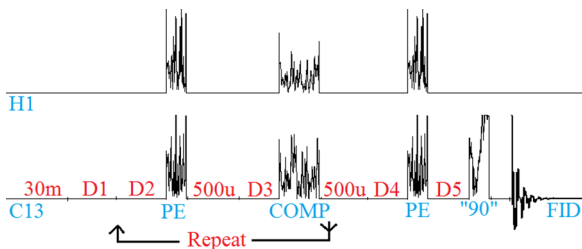


FIG. 4. Visualization of process 3, based on the TopSpin output, showing the rf power vs time in both channels, followed by an acquisition of the free induction decay (FID). The delays maximizing the polarization of C1 are D2 = 5 s, D3 = 3 s, D4 = 6 s, and D5 = 6 s.

TABLE I. Measured relaxation times of TCE in units of seconds.

	H	C2	C1
T_1	2.67 ± 0.03	17.3 ± 0.2	29.2 ± 0.1
T_2^*	0.2 ± 0.01	0.44 ± 0.03	0.23 ± 0.01

to a finite state,

$$I_z^H + 4I_z^{C2} + I_z^{C1} \propto \{\{6, 4, -2, -4, 4, 2, -4, -6\}\}, \quad (6)$$

where $I_z = \frac{1}{2}\sigma_z$. The two states are represented here [41] in product operator formalism as the reduced (shifted and scaled [29]) diagonal density operator [16,37]. The compression pulse (COMP) was designed to evolve the system from

$$I_z^H + I_z^{C2} + I_z^{C1} \propto \{\{3, 1, 1, -1, 1, -1, -1, -3\}\} \quad (7)$$

(a state of three spins with identical polarizations) to

$$\begin{aligned} & \frac{1}{2}I_z^H + \frac{1}{2}I_z^{C2} + \frac{3}{2}I_z^{C1} + 2I_z^H I_z^{C2} I_z^{C1} \\ & \propto \{\{3, 1, 1, 1, -1, -1, -1, -3\}\}. \end{aligned} \quad (8)$$

We chose this final state, as the four highest probabilities correspond to the four states where C1's spin is zero, namely, $|0ij\rangle, ij \in 0, 1$. Notice that the polarization-increase factor of C1 is 1.5, the maximum possible under unitary transformations, as mentioned above.

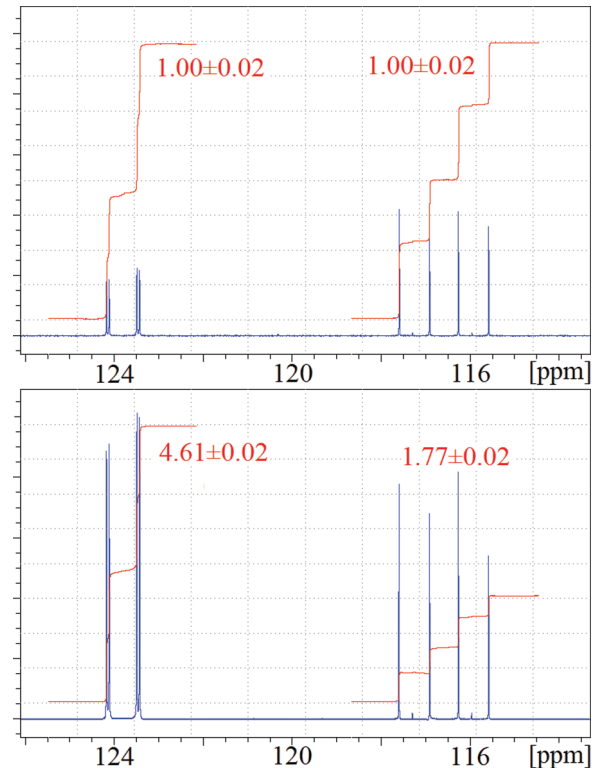
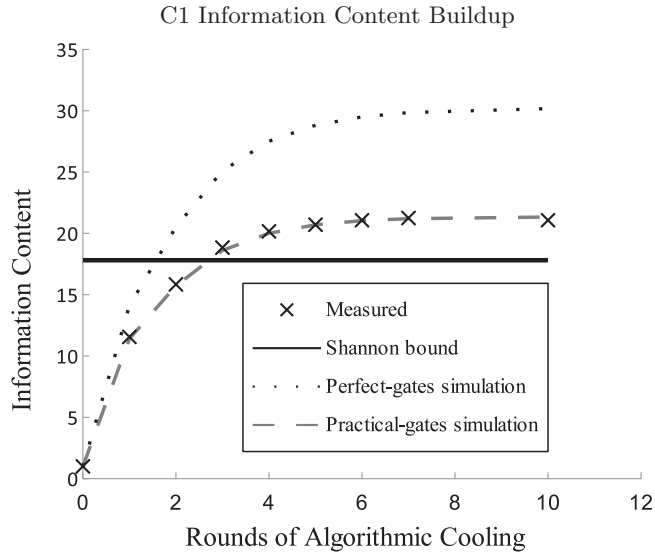


FIG. 5. ^{13}C carbon spectrum before and after seven cycles of algorithmic cooling to maximize the polarization of C1. The peak integrals are displayed in red (arbitrary units).



Round	Polarization	Carbon's IC
0	1.00	1.00 ± 0.04
1	3.40	11.56 ± 0.14
2	3.98	15.84 ± 0.16
3	4.34	18.84 ± 0.17
4	4.49	20.16 ± 0.18
5	4.55	20.70 ± 0.18
6	4.59	21.07 ± 0.18
7	4.61	21.25 ± 0.18
10	4.59	21.07 ± 0.18

FIG. 6. On the left, the measured IC vs the simulated IC of C1 at each cooling round of process 1, in units of $\frac{\epsilon_{C,\text{eq}}^2}{\ln(4)}$ (see [24]), where $\epsilon_{C,\text{eq}}$ is the carbons' equilibrium polarization. On the right, the measured polarization and IC of C1 in each round is shown. The measured error of all the polarizations is 0.02.

III. RESULTS

After seven rounds (see Fig. 6), the system reached its limit cycle and no more improvement could be expected. In process 1, C1 was cooled by a factor of 4.61 ± 0.02 , with $IC_{C1} = 21.25 \pm 0.18$, significantly higher than 17.84, the IC of the entire spin system at equilibrium (see Figs. 5 and 6). Alternatively, we see that the polarization bypassed the information theoretical bound of $\sqrt{17.84} = 4.22$. In process 2 we maximized $IC_{C1,C2}$, by adding another delay, D4 (that happened to be equal to D2=5 s in the optimal case), followed by PE. We obtained polarizations of 3.78 ± 0.02

and 3.4 ± 0.02 (of C1 and C2, respectively), with $IC_{C1,C2} = 25.9 \pm 0.2$. In process 3 we maximized the total IC, using an additional delay D5 = 6 s before the measurement (in addition, D4 was modified to 6 s). The measured polarizations were 2.87 ± 0.02 , 2.64 ± 0.02 and 3.58 ± 0.02 (for C1, C2, and H, respectively), with IC of 28.0 ± 0.20 .

There is a gap between SIMPSON's very high predicted pulse efficiency and the laboratory results (see [37]). The polarization of C2 following a PE pulse was ≈ 3.8 (0.95 efficiency), and a COMP pulse applied on the equilibrium state resulted in $\epsilon_{C1} \approx 2.8$ (0.92 efficiency). The main error factors are the hardware's imperfection distorting the pulses [42] and the pulses themselves being prepared without taking into account the T_2^* relaxation (see Table I) during the system evolution. The first factor could be negated using a technique mentioned in [35]. Once the measured gate efficiencies of 0.95 and 0.92 are added into the simulation, the simulated IC per round fit near perfectly the measured IC (see Figs. 6 and 7).

IV. SUMMARY

Using optimal control, we demonstrated a single-round and multiple-round AC applied on liquid-state NMR. We bypassed Shannon's bound in three different processes. The current optimal control methods (GRAPE), and better ones such as a second-order GRAPE [43] and Krotov based optimization [44], could enable various applications of AC in magnetic-resonance spectroscopy [14,15,45] and maybe also other potential applications [46–55].

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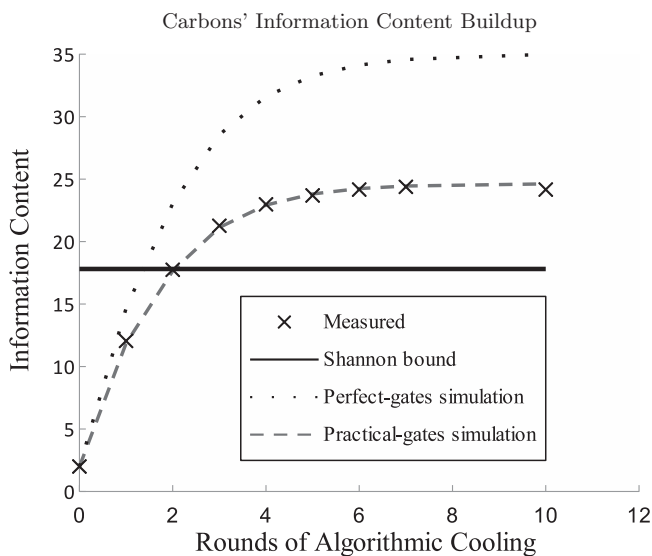


FIG. 7. The carbons' information content at each cooling round of process 2 in units of $\frac{\epsilon_{C,\text{eq}}^2}{\ln(4)}$ (see [24]), where $\epsilon_{C,\text{eq}}$ is the equilibrium polarization of the carbons. The measured error of all polarizations is 0.02.

- [1] D. Simon, On the power of quantum computation, in *Foundations of Computer Science, 1994 Proceedings, 35th Annual Symposium on the Foundations of Computer Science* (IEEE Computer Society Press, Los Alamitos, CA, 1994), pp. 116–123.
- [2] D. R. Simon, On the power of quantum computation, *SIAM J. Comput.* **26**, 1474 (1997).
- [3] P. W. Shor, Polynomial-time algorithms for prime factorization and discrete logarithms on a quantum computer, *SIAM J. Comp.* **26**, 1484 (1997).
- [4] A. W. Harrow, A. Hassidim, and S. Lloyd, Quantum Algorithm for Linear Systems of Equations, *Phys. Rev. Lett.* **103**, 150502 (2009).
- [5] H.-J. Briegel, W. Dür, J. I. Cirac, and P. Zoller, Quantum Repeaters: The Role of Imperfect Local Operations in Quantum Communication, *Phys. Rev. Lett.* **81**, 5932 (1998).
- [6] P. O. Boykin, T. Mor, V. Roychowdhury, F. Vatan, and R. Vrijen, Algorithmic cooling and scalable NMR quantum computers, *Proc. Natl. Acad. Sci. USA* **99**, 3388 (2002).
- [7] J. M. Fernandez, S. Lloyd, T. Mor, and V. Roychowdhury, Algorithmic cooling of spins: A practicable method for increasing polarization, *Int. J. Quant. Inf.* **2**, 461 (2004).
- [8] L. J. Schulman, T. Mor, and Y. Weinstein, Physical Limits of Heat-Bath Algorithmic Cooling, *Phys. Rev. Lett.* **94**, 120501 (2005).
- [9] S. Raeisi and M. Mosca, Asymptotic Bound for Heat-Bath Algorithmic Cooling, *Phys. Rev. Lett.* **114**, 100404 (2015).
- [10] N. A. Rodriguez-Briones and R. Laflamme, Achievable polarization for heat-bath algorithmic cooling, [arXiv:1412.6637](https://arxiv.org/abs/1412.6637) (2014).
- [11] D. K. Park, N. A. Rodriguez-Briones, G. Feng, R. Rahimi-Darabad, J. Baugh, and R. Laflamme, Heat bath algorithmic cooling with spins: Review and prospects, [arXiv:1501.00952](https://arxiv.org/abs/1501.00952) (2015).
- [12] E. Knill, R. Laflamme, and W. H. Zurek, Resilient quantum computation: Error models and thresholds, *Proc. Roy. Soc. London Ser. A* **454**, 365 (1998).
- [13] D. Aharonov and M. Ben-Or, Fault-tolerant quantum computation with constant error, in *Proceedings of the 29th ACM Symposium on the Theory of Computing* (ACM, El Paso, TX, 1997), pp. 176–188.
- [14] T. Mor, V. Roychowdhury, S. Lloyd, J. M. Fernandez, and Y. Weinstein, US Patent No. 6,873,154 (2005).
- [15] Y. Elias, H. Gilboa, T. Mor, and Y. Weinstein, Heat-bath cooling of spins in two amino acids, *Chem. Phys. Lett.* **517**, 126 (2011).
- [16] D. G. Cory, M. D. Price, and T. F. Havel, Nuclear magnetic resonance spectroscopy: An experimentally accessible paradigm for quantum computing, *Physica D: Nonlinear Phenomena* **120**, 82 (1998).
- [17] D. G. Cory, R. Laflamme, E. Knill, L. Viola, T. F. Havel, N. Boulant, G. Boutis, E. Fortunato, S. Lloyd, R. Martinez *et al.*, NMR based quantum information processing: Achievements and prospects, *Fortschr. Phys.* **48**, 875 (2000).
- [18] S. J. Glaser, NMR quantum computing, *Angewandte Chemie* **40**, 147 (2001).
- [19] L. M. K. Vandersypen and I. L. Chuang, NMR techniques for quantum control and computation, *Rev. Mod. Phys.* **76**, 1037 (2005).
- [20] J. A. Jones, Quantum computing with NMR, *Prog. NMR Spectrosc.* **59**, 91 (2011).
- [21] N. A. Gershenfeld and I. L. Chuang, Bulk spin-resonance quantum computation, *Science* **275**, 350 (1997).
- [22] P. O. Boykin, T. Mor, V. Roychowdhury, and F. Vatan, Algorithms on ensemble quantum computers, *Nat. Comp.* **9**, 329 (2010).
- [23] T. Cover and J. Thomas, *Elements of Information Theory*, Wiley Series in Telecommunications and Signal Processing (Wiley-Interscience, Hoboken, NJ, 2006).
- [24] G. Brassard, Y. Elias, J. M. Fernandez, H. Gilboa, J. A. Jones, T. Mor, Y. Weinstein, and L. Xiao, Experimental heat-bath cooling of spins, [arXiv:quant-ph/0511156](https://arxiv.org/abs/quant-ph/0511156) (2005).
- [25] Y. Elias, J. M. Fernandez, T. Mor, and Y. Weinstein, Optimal algorithmic cooling of spins, in *Proceedings of Unconventional Computation* (Springer, New York, 2007), Vol. 4618, pp. 2–26.
- [26] O. W. Sørensen, Polarization transfer experiments in high-resolution NMR spectroscopy, *Prog. Nucl. Mag. Res. Spec.* **21**, 503 (1989).
- [27] L. J. Schulman and U. V. Vazirani, Scalable NMR quantum computation, in *Proceedings of the 31st ACM Symposium on the Theory of Computing (STOC)* (ACM, Atlanta, GA, 1999), pp. 322–329.
- [28] J. M. Fernandez, *De Computazione Quantica* Ph.D. thesis, University of Montreal, Montreal, Canada, 2003.
- [29] Y. Elias, J. M. Fernandez, T. Mor, and Y. Weinstein, Algorithmic cooling of spins, *Isr. J. Chem.* **46**, 371 (2006).
- [30] L. J. Schulman, T. Mor, and Y. Weinstein, Physical limits of heat-bath algorithmic cooling, *SIAM J. Comp.* **36**, 1729 (2007).
- [31] Y. Elias, T. Mor, and Y. Weinstein, Semioptimal practicable algorithmic cooling, *Phys. Rev. A* **83**, 042340 (2011).
- [32] G. A. Morris and R. Freeman, Enhancement of nuclear magnetic resonance signals by polarization transfer, *J. Am. Chem. Soc.* **101**, 760 (1979).
- [33] D. E. Chang, L. M. Vandersypen, and M. Steffen, NMR implementation of a building block for scalable quantum computation, *Chem. Phys. Lett.* **338**, 337 (2001).
- [34] J. Baugh, O. Moussa, C. A. Ryan, A. Nayak, and R. Laflamme, Experimental implementation of heat-bath algorithmic cooling using solid-state nuclear magnetic resonance, *Nature (London)* **438**, 470 (2005).
- [35] C. A. Ryan, O. Moussa, J. Baugh, and R. Laflamme, Spin Based Heat Engine: Demonstration of Multiple Rounds of Algorithmic Cooling, *Phys. Rev. Lett.* **100**, 140501 (2008).
- [36] N. Khaneja, T. Reiss, C. Kehlet, T. Schulte-Herbrüggen, and S. J. Glaser, Optimal control of coupled spin dynamics: Design of NMR pulse sequences by gradient ascent algorithms, *J. Mag. Reson.* **172**, 296 (2005).
- [37] Y. Atia, Y. Elias, T. Mor, and Y. Weinstein, Quantum computing gates via optimal control, *Int. J. Quant. Inf.* **12**, 1450031 (2014).
- [38] J. M. Fernandez, T. Mor, and Y. Weinstein, Paramagnetic materials and practical algorithmic cooling for NMR quantum computing, *Int. J. Quant. Inf.* **3**, 281 (2005).
- [39] M. Bak, J. T. Rasmussen, and N. C. Nielsen, SIMPSON: A general simulation program for solid-state NMR spectroscopy, *J. Mag. Reson.* **147**, 296 (2000).
- [40] Z. Tošner, T. Vosegaard, C. Kehlet, N. Khaneja, S. J. Glaser, and N. C. Nielsen, Optimal control in NMR spectroscopy: Numerical implementation in SIMPSON, *J. Mag. Reson.* **197**, 120 (2009).

- [41] In the actual design we used the more precise γ ratio of 3.98, but in the explanation here we use 4 for clarity.
- [42] T. M. Barbara, J. F. Martin, and J. G. Wurl, Phase transients in NMR probe circuits, *J. Mag. Reson.* (1969) **93**, 497 (1991).
- [43] P. de Fouquieres, S. Schirmer, S. Glaser, and I. Kuprov, Second order gradient ascent pulse engineering, *J. Mag. Reson.* **212**, 412 (2011).
- [44] I. I. Maximov, Z. Tošner, and N. C. Nielsen, Optimal control design of NMR and dynamic nuclear polarization experiments using monotonically convergent algorithms, *J. Chem. Phys.* **128**, 184505 (2008).
- [45] G. Brassard, Y. Elias, T. Mor, and Y. Weinstein, Prospects and limitations of algorithmic cooling, *Eur. Phys. J. Plus* **129**, 258 (2014).
- [46] F. Rempp, M. Michel, and G. Mahler, A cyclic cooling algorithm, *Phys. Rev. A* **76**, 032325 (2007).
- [47] M. J. Henrich, F. Rempp, and G. Mahler, Quantum thermodynamic Otto machines: A spin-system approach, *Eur. Phys. J. Spec. Top.* **151**, 157 (2007).
- [48] H. Weimer, M. J. Henrich, F. Rempp, H. Schröder, and G. Mahler, Local effective dynamics of quantum systems: A generalized approach to work and heat, *Europhys. Lett.* **83**, 30008 (2008).
- [49] H. J. Briegel and S. Popescu, Entanglement and intramolecular cooling in biological systems? - A quantum thermodynamic perspective, [arXiv:0806.4552](https://arxiv.org/abs/0806.4552) (2008).
- [50] N. Linden, S. Popescu, and P. Skrzypczyk, How small can thermal machines be? The smallest possible refrigerator, *Phys. Rev. Lett.* **105**, 130401 (2010).
- [51] B. Criger, O. Moussa, and R. Laflamme, Study of multiple rounds of error correction in solid state NMR QIP, [arXiv:1103.4396](https://arxiv.org/abs/1103.4396) (2011).
- [52] R. Renner, Thermodynamics: The fridge gate, *Nature (London)* **482**, 164 (2012).
- [53] A. Blank, Scheme for a spin-based quantum computer employing induction detection and imaging, *Quantum Inf. Processing* **12**, 2993 (2013).
- [54] J.-S. Xu, M.-H. Yung, X.-Y. Xu, S. Boixo, Z.-W. Zhou, C.-F. Li, A. Aspuru-Guzik, and G.-C. Guo, Demon-like algorithmic quantum cooling and its realization with quantum optics, *Nat. Photonics* **8**, 113 (2014).
- [55] S. Lloyd, Quantum optics: Cool computation, hot bits, *Nat. Photonics* **8**, 90 (2014).