Experimental Saturation of the Heat-Bath Algorithmic Cooling bound

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Heat-Bath Algorithmic cooling (HBAC) techniques provide ways to selectively enhance the polarization of target quantum subsystems. However, the cooling in these techniques are bounded. Here we report the first experimental observation of the HBAC cooling bound. We use HBAC to hyperpolarize nuclear spins in diamond. Using two carbon nuclear spins as the source of polarization (reset) and the 14N nuclear spin as the computation bit, we demonstrate that repeating a single cooling step increases the polarization beyond the initial reset polarization and reaches the cooling limit of HBAC. We benchmark the performance of our experiment over a range of variable reset polarization. With the ability to polarize the reset spins to different initial polarizations, we envisage that the proposed model could serve as a test bed for studies on Quantum Thermodynamics.

There is a rapid growth in the scope and sophistication of spin-based quantum technologies ranging from quantum-information applications to quantum sensing[1]. In a large number of cases the quantum spin system represent a central spin system, i.e. a central well controllable spin, which in most cases is an electron spin that dominates the dynamics of its environment. Irrespective of the particular application, controlling this environment is key for proper function of the quantum system [2]. Essentially one can try to eliminate all spins from the environment to enhance the central spin coherence time by dedicated material growth, e.g. isotope purification [3]. Alternatively the spin environment can be polarized as to not decohere the central spin [4].

Over the past decade single nitrogen-vacancy (NV) centers in diamond have emerged as a novel atomic-size magnetometer for detecting nuclear spin ensembles or even single nuclear spins with high sensitivity [5]. Essentially it is a prototype central spin system, consisting of a substitutional nitrogen atom and an adjacent vacancy. Its spin triplet (S = 1) ground state can be polarized and read out optically, so that electron spin resonance experiments can be performed on spin ensembles or single spins [6]. The electron spin associated with the NV center is a very good qubit and at the same time allows control over its immediate nuclear spin environment, e.g. the associated 14N or the 13C nuclear spins in the proximity of the NV center [2]. In this work we use these nuclear spins, and a combined usage of the HH and HBAC cooling schemes to demonstrate the hyperpolarization (see Fig. 1).

Polarization of spin environments has been largely dealt using resonant exchange interactions with highly polarized spins, the Hartmann-Hahn (HH) exchange method [7]. When this transfer happens from the highly polarized electron spin (due to its g-factor) to the nuclear spin, it is often dubbed dynamic nuclear polarization (DNP) [4]. Heat-Bath algorithmic cooling (HBAC), provides an alternative for cooling a target subset of spins [8, 9]. These techniques use quantum operations to compress the entropy away from a target element in an ensemble of the spins to the rest of the ensemble and then through that to the heat-bath.

For HBAC, it is assumed that there is an ensemble of spins or qubits and that universal quantum operations can be applied to them. The goal is to increase the polarization of a subset of spins in the ensemble, which are referred to as the “computation” qubits or spins. This idea was first introduced in [8] for closed systems and it was proposed to use entropy compression algorithms to push the entropy away from the target elements to the rest of spins which are known as the “reset” spins. However, the performance of the algorithm was limited to Shannon bound for compression. To circumvent the Shannon bound, Boykin et al. proposed to use a heat-bath to transfer the compressed entropy out of system [10]. In this setting, after the compression of the entropy, the reset spins, which are now heated up, are cooled down in interaction with a heat-bath. Often this is the natural spin relaxation process that equilibrates the reset spins, but it can also be actively done through optical pumping or similar techniques in some systems. After the reset spins are cooled down, the process of compression is repeated and this makes an iterative process where in each iteration, first the entropy is transferred away from the computation spins and compressed to the reset spins and then the reset spins are reset back to their initial state. However, even in the interaction with the heat-bath, HBAC techniques cannot always fully polarize the target spins. In [11], Schulman et al. showed that it is not always possible to completely purify the target spins and that the achievable cooling is limited. They introduced the Partner-Pairing algorithm (PPA), which, for compression, sorts the diagonal elements of the density matrix of the computation and reset spins decreasingly. They showed that this is the optimal HBAC technique and also found that it still cannot always reach full polarization for the target spins. Later, Raeisi and Mosca
Consisting of three nuclear spins (14N, 13C, 13C) and a central electron spin (e) to which they are coupled. Various quantum gates among the n-spins are mediated by the electron. (b) The quantum circuit implemented for HBAC. The power indicates that this is an iteration and is repeated M times. (c)-(d) Experimental Quantum circuits for implementing the reset operation, R and \( U_e \) in (b).

We start by describing our method of cooling and then present our results and compare them with PPA and the cooling limit of HBAC techniques \[12\].

Here we report the first experimental realization of reaching the cooling limit of HBAC. We use a system of three spins to polarize a Nitrogen nuclear spin in a nitrogen vacancy centre in diamond. Since we can easily polarize the two carbon spins, we use both of them for the reset and the Nitrogen as the single computation spin. We use a technique similar to the one in \[13\], which consists of repeating a fixed set of operations in subsequent iterations. We show numerically that this method converges to the HBAC limit and verify this in our experiment.

We implement the circuit shown in Fig. 1 (b) with three nuclear spins viz., one 14N spin and two 13C spins. We choose the 14N as our target nuclear spin to be hyper-polarized. Though the choice of the target qubit is not unique, a comparatively faster readout of the 14N allows us to reduce the overall experimental runtime \[2\]. The carbon nuclear spins chosen for reset are strongly coupled with the electron spin, allowing for a faster and efficient swap of their polarization with the electron spin.

The polarization of the two 13C spins are reset in each iteration of the algorithm, which is depicted here by the R box and its experimental implementation in Fig. 1 (c). These reset operations are non-unitary gates and are performed via the central NV spin, which we detail later. Mathematically, we take the reset state of the two carbon spins to be

\[
\rho_{C_{1,2}} = \frac{1}{z} \left( \begin{array}{cc} e^{\epsilon_{1,2}} & 0 \\ 0 & e^{-\epsilon_{1,2}} \end{array} \right),
\]

where \( z \) is normalization constant. The spin-polarization’s of the two carbon spins are then given by \( P_{C_{1,2}} = \tanh(\epsilon_{1,2}) \). The compression is done by the application of \( U_e \) (shown in Fig. 1(b)) which only swaps the \( |100\rangle \) and \( |011\rangle \) i.e., the target spin is flipped only when the states of the other two qubits are identical and dissimilar from the target spin state. When the circuit in Fig. 1(b) is repeated, the target spin polarization grows and saturates to a value higher than the reset polarizations of the other two spins. We represent the final asymptotic state of the target spin with

\[
\rho_f = \frac{1}{z} \left( \begin{array}{cc} e^{\epsilon_{\text{max}}} & 0 \\ 0 & e^{-\epsilon_{\text{max}}} \end{array} \right),
\]

with \( z \) the normalization factor. For the simple case of our three-qubit system, we can analytically calculate the asymptotic state of the 14N spin and show that it converges to the HBAC limit. The limit of HBAC with reset polarizations \( \epsilon_{1,2} \) is

\[
\epsilon_{\text{max}} \leq 2^{n-1}(\epsilon_1 + \epsilon_2),
\]

where \( n \) is the number of computation qubits and in our experiment, \( n = 1 \). We can express the final polarization, \( P_{\text{max}} \) in terms of \( \epsilon_{\text{max}} \) as

\[
P_{\text{max}} = p_0 - p_1 = \frac{e^{\epsilon_{\text{max}}} - e^{-\epsilon_{\text{max}}}}{e^{\epsilon_{\text{max}}} + e^{-\epsilon_{\text{max}}}} = \tanh(\epsilon_{\text{max}}),
\]

where \( p_0 \) and \( p_1 \) are the probability of the spin being in the up and down states correspondingly. This indicates that the polarizations should be limited by \[12\]

\[
P_{\text{max}} = p_0 - p_1 < \tanh(\epsilon_1 + \epsilon_2).
\]

We also use \( P_{C_1} \) and \( P_{C_2} \) to show the polarization of the two Carbon nuclear spins. We experimentally obtain this for a single Nitrogen spin by repeating the above circuit 25 times, with the first qubit in the circuit being the Nitrogen and the second and the third being the carbon spins. We also include this limit in our plots for comparison.

Figure 2 compares the polarization build-up of the target Nitrogen spin in our method to PPA with single and two-qubit reset. For the single-qubit-reset PPA, only one spin is reset and we assume that it is reset to \( \epsilon_1 > \epsilon_2 \). For the top plot, we take the reset polarizations of both carbons to be \( P_{C_1,C_2} = 0.2 \) whereas for the

\[\text{Figure 1. (a) Schematic representation of our physical system consisting of three nuclear spins (14N, 13C, 13C) and a central electron spin (e) to which they are coupled. Various quantum gates among the n-spins are mediated by the electron. (b) The quantum circuit implemented for HBAC. The power indicates that this is an iteration and is repeated } M \text{ times. (c)-(d) Experimental Quantum circuits for implementing the reset operation, R and } U_e \text{ in (b).}\]
An external static magnetic field of is fabricated onto the diamond via optical lithography. Microwave (MW) and RF excitation made from copper via focused ion beam milling. A coplanar waveguide for and a solid immersion lens has been carved around it and a 13C concentration of 0.2%.

After polarizing the two 13C spins using the SWAP gates shown in Fig. 1(c), we perform the $U_e$ gate shown in Fig. 1(d). This three-spin non-local gate swaps the population between the states $|00\rangle \leftrightarrow |11\rangle$, where the first spin state corresponds to the target 14N nuclear spin. We decompose $U_e$ to three Toffoli gates that are further decomposed into $C_n\text{NOT}_e$ gates. Such gate have been earlier used in [2] for entangling the nuclear spins.

We use optimal control platform DYNAMO [15] to realize fast electron spin (2π) rotations shown in Fig. 1(d). As the individual hyperfine transitions are inhomogeneously broadened to approximately 35kHz, it drastically complicates the implementation of electron spin gates required to have a spectral resolution better than 90kHz (the $A_{zz}$ coupling of the weakest coupled nuclear spin) [14]. Hence implementing electron spin rotations using shaped MW pulses, obtained from the optimization with DYNAMO, is key for our experiment [2]. The optimized pulses used here can avoid cross-talk between the individual electron hyperfine transitions even in a dense polarization.

The experiments were carried out at room temperature on a homebuilt confocal microscope setup using a type IIa CVD grown diamond crystal (layer) that has [100] surface orientation and a 13C concentration of 0.2%. The NVC is located $\sim 15\mu m$ below the diamond surface and a solid immersion lens has been carved around it via focused ion beam milling. A coplanar waveguide for microwave (MW) and RF excitation made from copper is fabricated onto the diamond via optical lithography. An external static magnetic field of $\sim 540 mT$ from a permanent magnet is aligned along the symmetry axis of the NV centre (z-axis). A 532 nm light is focused onto the NV by an oil immersion objective, which also collects light from the diamond. The fluorescence light of the NV is isolated by spatial and spectral filtering and finally detected with a single photon counting detector.

Optical excitation of the NV centre polarizes the electron spin triplet ($S=1$) into its $m_S = 0$ state, which forms the key element in the reset step shown in Fig. 1, and the source of spin polarization in the experiment.

Due to the weak coupling among the nuclear spins, no direct quantum gates among the nuclear spins, required for HBAC (Fig. 1(b)) can be performed. These gates are mediated by the electron spin, and hence is actively involved in the dynamics, (Fig. 1(d)), in addition to being the polarization source for the carbon spins. The coupling of the NV electron to the surrounding nuclear spins is dominant along its symmetry (z) axis [2]. The 14N spin and the two nearest 13C spins of the lattice chosen for our experiments, are coupled the NV spin with strengths $-2.16MHz$, 90kHz and 414kHz respectively. While the two 13C spins are spin-1/2 systems the 14N is a spin-1 system with population distributed among the triplet levels $m_I = +1, 0, -1$ respectively. Hence, we choose a two-level subspace (TLS) formed by the $|\pm 1\rangle$ states of the 14N for our analysis, and omit the state $|0\rangle$, and renormalize the spin-state population in the TLS accordingly. The spin-polarization is then directly measured by the population difference $P_N = P_+ - P_-$ within this effective TLS, as shown in Fig.3.

We transfer the optical spin polarization of the central (NV)-spin to the two 13C spins via SWAP gate which involves four conditional-NOT gates as shown in Fig. 1(c). In the experiments, a nuclear spin reset (repolarization) is performed with a fidelity that is much higher than the total fidelity of the quantum circuit shown in Fig. 1. This allows us to explore the whole polarization space ($P$) in HBAC.

The three nuclear spins used in the experiments do not interact and thus each non-local gate performed among them needs to be mediated by the electron spin. Thus, unlike the reset operation, nuclear - nuclear swap gates occur via involvement of the electron spin and hence, suffer from electron $T_2$ decay, which is on the order of 500μs for this specific NV center [14]. This lowers the fidelity of the swap operation and the required time is at least as long compared to a reset of any nuclear spin.
Figure 3. (a) The hyperpolarization of 14N spins for varying reset polarizations on the two 13C nuclear spins is shown here. The experimental data is compared with the theoretical simulations and the ideal HBAC saturation limit. (Inset) In the inset we plot the ratio of experimental polarization of the 14N, $P_N$, with the ideal HBAC limit $P_{\text{max}}$. (b) The initial polarization of two 13C spins obtained by varying the rotation angle $\theta$. (c) The quantum circuit employed for arbitrary initialization used in (a). (d) 3D plot showing the hyperpolarizaiton of the 14N over the polarization space of the two 13C spins.

spectrogram. The optimized $U_e$ gate is then iterated until the 14N polarization $P_N$ saturates, which we show in Fig. 3(a). With the limitation from the decoherence time of the electron spin, and the decoherence induced by the electron optical excitation on the nuclear spins, the fidelity of the $U_e$ gates will eventually deteriorate with increasing number of iterations. In our experiment one iteration i.e., a reset operation and the non-local gate $U_e$ would take $\sim 570\mu$s. Given the various life times of the electron and nuclear spins $T_{2e} \approx 500\mu$s, and $T_{2n} \approx 8.5\text{ms}$, and the relaxation time of the nuclear spins $T_{2n} \sim 1\text{s}$ we could safely perform $\sim 50$ iterations of the HBAC algorithm with high fidelity, which would allow the experimentally observed saturation of the polarization.

In the final part of our experiments, we use the ability to arbitrarily polarize the two reset spins to varying degree, and study the polarization gain of the 14N spin, as shown in Fig. 3(c). For this we first maximally polarize the two 13C spins via the SWAP gate with the electron spin, and then rotate the nuclear spins such that a superposition state, $\sim |0\rangle + \beta|1\rangle$, is formed. By (laser-induced) dephasing of the carbon spin, one would obtain an ideal (thermal-like) polarization $P_C = |\alpha|^2 - |\beta|^2$. We would like to note that the HBAC, ideally is not going to be affected by the off-diagonal elements of the density matrix. Hence, imperfect dephasing of the nuclear spins (Fig. 3(c)) here, would not be a source of error in our experiments. Polarization is only affected by the diagonal elements of the density matrix and the operation $U_e$ would only permute the diagonal elements.

In conclusion, we have experimentally implemented HBAC for three spins and reached the cooling limit of HBAC for this system. With 25 iterations, the Nitrogen spin reaches the $P_{\text{max}}$, the expected cooling limit of HBAC. The polarization of the 14N fluctuates around the cooling limit and in some cases, it even exceeds the limit. This may be attributed to correlations between spins in the bath and the system [16], due to imperfections in $U_e$. But further experiments are required to investigate this, and to benchmark the role of such correlations on the cooling limit of HBAC [16].

Further with the ability to polarize the reset spins to varying degree (a feature unique to our system), we benchmark our implementation of HBAC for purification and polarization of the nuclear spins. The main requirement for using this method is that the spins in the system can be individually controlled and that the relaxation time of the target elements is long enough compared to the HBAC process. While the NV-based electron-nuclear spin system employed here is a good model system accommodating both these requirements, it can also be implemented in other solid-state systems to boost the polarization of a target spin system. Further, the achievable polarization would increase exponentially with employing more computation and reset spins.

Combining our method with techniques like DNP would enhance the polarization of target spin(s) beyond that achievable directly with DNP.

The physical system employed here represents a generic heat engine model for a quantum refrigerator, where the working fluid (electron spin) under modulation (quantum control) could continuously extract heat from a cold bath and dump it into the heat bath [17].

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