Bath Assisted Cooling of Spins

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A suitable sequence of sharp pulses applied to a spin coupled to a bosonic bath can cool its state, i.e., increase its polarization or ground state occupation probability. Starting from an unpolarized state of the spin in equilibrium with the bath, one can reach very low temperatures or sizeable polarizations within a time shorter than the decoherence time. Both the bath and external fields are necessary for the effect which comes from the backreaction of the spin on the bath. This method can be applied to cool at once a disordered ensemble of spins. Since the bath is crucial for this mechanism, the cooling limits are set by the strength of its interaction with the spin(s).

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Cooling, i.e. obtaining relatively pure states from mixed ones, is of central importance in fields dealing with quantum features of matter. Laser cooling of motional states of atoms is nowadays a known achievement \cite{1}. The related problem of cooling spins is equally known: it originated as an attempt to improve the sensitivity of NMR/ESR spectroscopy \cite{2,3,4,5,6}, since in experiments the signal strength is proportional to the polarization. Recently it got renewed attention due to realizations of setups for quantum computers \cite{7}. The very problem arises since the most direct methods of cooling spins, such as lowering the temperature of the whole sample or applying strong dc fields, are not feasible or not desirable, e.g. in biological applications of NMR. Indeed, at temperature $T=1$K and magnetic field $B=1$T the equilibrium polarization of a proton is only $\tanh \left( \frac{\mu B T}{2k_B} \right) \approx 10^{-3}$ since the ratio $\mu = \frac{\text{frequency}}{\text{field}}$ is equal to 42 MHz/T. For an electron $\mu$ is $10^3$ times larger and for $^{15}$N it is 10 times smaller. The weak polarization can be often compensated by a large number of spins, but for some NMR-isotopes the natural abundance is too low (0.36\% for $^{15}$N).

Over the years, several methods were proposed to attack the problem of small polarizations. The polarization is generally increased via a dynamical process and it is used before relaxing back to equilibrium \cite{2,3,4,5,6}. Specially known are methods where a relatively high polarization is transferred from one place to another, e.g. from electronic to nuclear spins \cite{2,3,4,5,6}. In this respect electronic spins play the same role as the zero-energy states of atoms \cite{1}. Polarization transfer was studied for cooling nuclear spins, but can be employed to study cooling of atomic few-level systems in the context of optimal control theory \cite{6}. Polarization transfer was studied in various settings both theoretically and experimentally \cite{2,3,4,5,6}. However, this scheme is limited —besides requiring an already existing high polarization— by the availability and efficiency of the transfer interaction. A related method, polarization compression, consists in manipulating a set of $n$ spins in such a way that the polarization of one spin is increased at the expense of decreasing the polarization of the remaining $n-1$ spins. These spoiled ones can be recycled and used again \cite{7}. Since spins are cooled one by one, a long time and carefully designed inter-spin interactions are needed for cooling a large ensemble.

Here we propose a mechanism of cooling which only uses the most standard setting of NMR or ESR physics \cite{2,3,4}: spins-\textsuperscript{1/2} under the action of external field pulses coupled to a thermal bath at the same temperature. The bath is needed because external fields alone cannot achieve cooling \cite{10}. However, we assume neither that the bath is under any direct control, nor special constraints on the bath-spin interaction: it is the standard one, widely studied in the context of decoherence. We show that, rather than being a hindrance in quantum system manipulations, the bath is capable of producing ordered effects on the spin, which can cool it down to very low temperatures ($\sim 1\mu$K for a proton) in a finite time. Two factors are crucial: the backreaction of the spin on the bath and the generation of transversal components (coherences) during the cooling process. Since the effect is generated via the bath, one can cool \textit{at once} a completely disordered ensemble of spins.

The model we study is well known \cite{11,12,13,14,15}: a spin-\textsuperscript{1/2} with energy levels $\pm \frac{1}{2} \hbar \Omega$ couples to a bath, modeled by a set of harmonic oscillators with creation and annihilation operators $\hat{a}_k^\dagger$ and $\hat{a}_k$. The total Hamiltonian reads

$$\hat{H} = \frac{\hbar \Omega}{2} \sigma_z + \sum_k \hbar \omega_k \hat{a}_k^\dagger \hat{a}_k + \frac{\hbar \sigma_z}{2} \hat{X} \, , \quad [\hat{a}_l, \hat{a}_k^\dagger] = \delta_{kl}. \ (1)$$

Here $\omega_k$ are the bath frequencies, $\sigma_{x,y,z}$ the Pauli operators, and $\hat{X} = \sum_k g_k (\hat{a}_k^\dagger + \hat{a}_k)$ is the collective coordinate of the bath. The interaction is chosen assuming that the $T_1$-time, connected to relaxation of the average $\langle \sigma_z \rangle$ is very large (infinite) \cite{2,3,5}. The $g_k$ are couplings

\begin{align}
\hat{g}_k &= \frac{\hbar \sigma_x}{2} \left( \sum_l g_{kl} \hat{a}_l^\dagger \hat{a}_l \right), \\
\hat{g}_k &= \frac{\hbar \sigma_y}{2} \left( \sum_l g_{kl} \hat{a}_l^\dagger \hat{a}_l \right), \\
\hat{g}_k &= \frac{\hbar \sigma_z}{2} \left( \sum_l g_{kl} \hat{a}_l^\dagger \hat{a}_l \right),
\end{align}
parametrized via the spectral density function $J(\omega)$:

$$J(\omega) = \sum_k g_k^2 \delta(\omega - \omega_k).$$

(2)

In the thermodynamic limit the bath modes are dense and $J(\omega)$ becomes a smooth function determined by the physics of the system-bath interaction [11]. The oscillators can represent real phonons or stand for an effective description of a rather general class of thermal baths [11].

Let us recall how the model [11] is solved 12: $\hat{\sigma}_z$ is conserved, while $\hat{a}_k(t) = e^{-i\omega_k t/2} \hat{a}_k(0) + e^{i\omega_k t/2} (e^{-i\omega_k t} - 1)$.

This leads along with Eqs. 11 to 12 to

$$X(t) = \hat{\eta}(t) - \hat{\sigma}_z \hat{F}(t),$$

$$\hat{\eta}(t) = \sum_k g_k [\hat{a}_k^\dagger(0) e^{i\omega_k t} + \hat{a}_k(0) e^{-i\omega_k t}],$$

$$F(t) = \int_0^\infty \frac{d\omega}{\omega} J(\omega)(t - \sin\omega t),$$

(4)

where $\hat{\eta}(t)$ is the quantum noise operator, and where $\hat{F}(t) \equiv \hat{T}^\dagger(t)\hat{F}(t) \hat{T}(t)$ quantifies the backreaction of the spin on the collective operator of the bath. This effect, not relevant for coherence as such, is crucial for our purposes.

We assume that at the initial time $t = 0$ the common density matrix of the bath and the spin is factorized:

$$\rho(0) = \frac{e^{-\beta H_0}}{\text{tr} e^{-\beta H_0}}, \quad \hat{H}_0 = \frac{1}{2} \hat{H}_0 \hat{\sigma}_z + \sum_k h \omega_k \hat{a}_k^\dagger \hat{a}_k,$$

(6)

where $T = 1/\beta$ is the common temperature ($k_B = 1$). $\rho(0)$ describes the spin prepared independently from the bath and then brought in contact with it at $t = 0$, e.g., by injection of the spin into a quantum dot or by creation of an excited through external radiation.

As follows from Eq. 6, $\hat{\eta}(t)$ is a Gaussian operator with $\langle \hat{\eta}(t) \rangle = 0$ and time-ordered correlator $\langle \hat{\eta}(t) \hat{\eta}(0) \rangle = \xi(t) - i \hat{F}(t), \quad t > 0$, where $\langle (...) \rangle$ is taken over the initial state [13], and where

$$\xi(t) \equiv \int_0^\infty d\omega J(\omega) \frac{1 - \cos\omega t}{\omega^2} \coth\frac{\hbar\omega}{2T}.$$  

(7)

The Heisenberg equation of the spin, $\hat{h} \hat{\sigma}_\pm = i[\hat{H}, \hat{\sigma}_\pm]$ with $\hat{\sigma}_\pm = \hat{\sigma}_x \pm i \hat{\sigma}_y$, $\hat{\sigma}_\pm = \pm \hat{\sigma}_\pm$, is solved as

$$\hat{\sigma}_\pm(t) = e^{\pm \frac{i\hat{H}(t-t_0) - i\hat{F}(t-t_0)}{\hbar} \frac{\hbar}{2T}},$$

(8)

where $\tau$ is the time-ordered exponent. Defining $\xi t \hat{A} \equiv e^{it\hat{H}/\hbar} \hat{A} e^{-it\hat{H}/\hbar}$ one derives

$$\xi(t) \equiv \int_0^\infty d\omega J(\omega) \frac{1 - \cos\omega t}{\omega^2} \coth\frac{\hbar\omega}{2T},$$

$$\chi(t_1,t_2,t_3) \equiv \int_0^\infty \frac{d\omega}{\omega} J(\omega) e^{i\omega(t_1 + t_2 + t_3)} \cos\omega \chi(t_1,t_2,t_3),$$

$$\langle \hat{\sigma}_\pm(t_1) \hat{\sigma}_\pm(t_2) \hat{\sigma}_\pm(t_3) \rangle = e^{-\xi(t_1 + t_2 + t_3)} \chi(t_1,t_2,t_3).$$

(9)

Eq. 11 is the standard formula for the average of a Gaussian operator. The factor $e^{-\xi(t)}$ leads to decoherence since, due to Eqs. 8 11, $\langle \hat{\sigma}_\pm(0) \rangle = e^{-\xi(t_1) + \xi(t_2)} \langle \hat{\sigma}_\pm(0) \rangle$ for a general factorized initial state. In this simplest situation the backreaction factor $F$, properly obtained already in 12 14, cancels out. In general, $F$ can shift the spin’s frequency $\Omega$ as seen below.

The action of external fields on the spin amounts to a time-dependent Hamiltonian $\hat{H}(t) = \hat{H} + \hat{h}(t) \hat{\sigma}_z$. In the pulsed regime 2 4 13 14 15 $\hat{h}(t)$ differs from zero only for very short intervals of time $\delta$ being there very large, $\hat{h}(t)\delta \sim 1$, to achieve a finite effect. As a consequence, terms $\propto \hat{\sigma}_z$ in $\hat{H}$ can be neglected during the time-interval $\delta$. A single pulse can perform an arbitrary unitary transformation in the space of the spin (rotation of the Bloch vector $\langle \hat{\sigma}_z \rangle$). We parametrize it as

$$\hat{U} \equiv e^{i\hat{h}(t)\hat{\sigma}_z/\hbar}, \quad (0 \leq \phi, \psi \leq 2\pi, 0 \leq \theta \leq \pi/2),$$

$$\hat{F}(t) = \gamma [\Gamma t - \arctan(\Gamma t)] \equiv i \hat{F}(t), \quad \Gamma \equiv \frac{\hbar T}{\hbar} T.$$ 

(10)

where $\gamma$ is a dimensionless coupling constant, and where $\Gamma$ (usually $\gg \Omega$) is the bath’s response frequency. Eqs. 6 13 14 imply $\xi(t) = \gamma \ln \left[ \frac{e^{\frac{\pi^2 T^2}{\hbar^2} \phi^2}}{1 + \text{arctan}(\Gamma t)} \right].$

$$F(t) = \gamma [\Gamma t - \arctan(\Gamma t)] \quad \gamma = \frac{\hbar T}{\hbar} T.$$ 

(11)

As a first example, we take ohmic interaction 11:

$$J(\omega) = \gamma \omega e^{-\omega/\Gamma},$$

(12)

where $\gamma$ is a dimensionless coupling constant, and where $\Gamma$ (usually $\gg \Omega$) is the bath’s response frequency. For low temperatures $\Theta < 1$: $e^{-\xi(t)} = (1 + T^2 \Gamma^2)^{-\gamma/2}$, while for $\Theta \geq 1$, $e^{-\xi(t)}$ starts as a gaussian, but continues as $e^{-\xi/t^2}$ with $T_2 = \hbar/(2\gamma T)$.

Cooling amounts to make the final polarization $\langle \hat{\sigma}_z \rangle$ more negative than the initial one. $\langle \hat{\sigma}_z \rangle = \langle \hat{\sigma}_z(0) \rangle = 0$. A single pulse cannot achieve cooling since it sees the initial local equilibrium state of the spin, and then according to the no-cooling principle 11 it can only heat the spin’s state up: for an arbitrary pulse $P_1$ applied at time $t$, $\langle \hat{\sigma}_z(t) \rangle = \langle \hat{\sigma}_z(0) \rangle \cos 2\theta_2 \geq \langle \hat{\sigma}_z(0) \rangle \cos 2\theta_1 \geq \langle \hat{\sigma}_z(1) \rangle \equiv \langle \hat{\sigma}_z \rangle_1$ (recall $\langle \hat{\sigma}_z \rangle_1 \leq 0$). Thus we have to employ at least two pulses. The final polarization after one pulse at $t$ and one at $t + \tau$, $P = |\langle \hat{\sigma}_z \rangle_1| = |\langle \hat{\sigma}_z \rangle_2 \hat{P}_1 \hat{E}_2 \hat{P}_2 \hat{\sigma}_z \rangle|$, reads from Eqs. 8 12:

$$\langle \hat{\sigma}_z(t) \rangle = \langle \hat{\sigma}_z(0) \rangle \cos 2\theta_2 + s_2 \sin 2\theta_1 \sin 2\theta_2, \quad s_2 = -e^{-\xi(\tau)} \int e^{i\theta \tau + i\xi} \langle \hat{\sigma}_z \rangle \cos \chi \sin \chi \rangle.$$ 

(13)

where $\chi = \chi(0,t,t)$ was defined in Eq. 10, and

$$\omega_2 = \psi_1 - \psi_2 - \phi_1 - \phi_2$$

arises from Eq. 12. There are now two factors that come from the bath: $e^{-\xi(\tau)}$ in $s_2$ accounts for the decoherence in the time-interval $(t, t + \tau)$ of transversal terms generated by the first pulse, while $\chi$ is the backreaction factor from Eqs. 8 10.

Though the finite-$t$ situation can be of its own interest, for all results below we set $t \Gamma \gg 1$ (a mild condition, since
1/Γ is typically the shortest time-scale), since this makes the outcome independent on the details of the initial state preparation. In this ergodic limit, the initial condition ρ(0) ∝ e−βH0 defined by Eq. 10 is equivalent to the overall equilibrium preparation ρeq(0) ∝ e−βH 10.

In Eq. (10), s2 can always be made negative by tuning α2. Minimizing ⟨σ2⟩t over θ1, θ2 produces min[⟨σ2⟩t, s2]. If the initial polarization is already high, |⟨σ2⟩t| > s2, no pulses should be applied, since they only heat the spin up. However, in the relevant situation ⟨σ2⟩t ≈ 0, the minimum ⟨σ2⟩t = s2 is reached for θ1 = θ2 = π/2 = π/2. Altogether, using Eq. (10) and Ω ≪ Γ, yields

\[ ⟨σ2⟩t = s2 = -e^{-ξ(t)} \sin [γ \arctan (τΓ)] . \tag{17} \]

The choice of optimal pulses, which have to be coherence generating, can be a π/2-pulse along the x-axis followed by a π/2-pulse along the y-axis: \( P_1σ_{zx} = σ_{yx}, P_1σ_y = -σ_z, \) and \( P_2σ_{zy} = σ_{y}, P_2σ_y = -σ_z. \) Fig. 1 and Table I show that \( max_x[⟨σ2⟩t] \) can approach its maximal value 1.

Transversal components generated by the two pulses will decay after a time \( T_2^* \), and the spin will be described by a Gibbsian at a temperature lower than the initial T.

The origin of this cooling effect is in shifting the spin’s frequency Ω by the factors F ≈ Γγ and χ, recall Eqs. (3) [10], which arise from the (via the pulses) enhanced back-reaction of the spin on the collective coordinate \( \tilde{X} \) of the bath. The generation of coherences by the first \( \frac{π}{2} \)-pulse is necessary to couple \( \tilde{σ} \) to the bath, which so to say “thermalizes” \( \langle \tilde{σ} \rangle \) under the shifted frequency. Cooling is achieved via the proper pulses, still it decreases with \( γ \) (weaker backreaction) and with \( β \) (larger decoherence). The time τ between two pulses can be neither too short (\( χ \) is visible on the time-scale \( 1/(ΓΓ) \)), nor too long, since otherwise decoherence will diminish the influence of the first pulse. As shown in Table I, the cooling improves by i) applying three successive pulses and optimizing over their parameters; ii) applying two pulses, waiting for a time \( T_2^* \), so that the transversal components decay, \( ⟨σz⟩t \to 0 \), applying another two pulses, and so on \( n \) times. The final \( |⟨σ2⟩t| \) is maximized over all free parameters. It appears that this numerical maximization can be done locally, i.e., by maximizing the output \( |⟨σ2⟩t| \) after each pair of pulses. This “greedy” optimization shows up also in cooling via a zero-temperature bath 13.

Inhomogeneous broadening. Many experiments in NMR/ESR are not done with a single spin, but with an ensemble of non-interacting spins having random frequencies Ω due to action of their environment or due to inhomogeneous external field 2 4. The collective variables are obtained by averaging the corresponding expressions for a single spin: \( m_t = \int dΩ P(Ω)|⟨σ2⟩t| . \) Assume that the distribution of Ω is gaussian with average Ω0 and dispersion \( \Delta Ω = P(Ω) \propto e^{-(Ω−Ω_0)^2/2\Delta Ω^2} \). Averaging over \( P(Ω) \) the term \( e^{i\phi t} \) in Eq. (10) produces a factor \( \sim e^{-dΩ^2/2}, \) a strong decay on times \( T_2^* \propto 1/\sqrt{d} \). After this decay, \( s_2 \to 0 \) and any two pulses will only heat the ensemble up as seen from Eq. (10).

It is however possible to employ the spin-echo phenomenon and cool, i.e., increase the collective final polarization \( |m_t| \) as compared to the initial \( |m_t| \), even for a completely disordered ensemble with \( T_2^* \) being very short: Apply precisely in the middle of the two pulses an additional \( π \)-pulse in \( x \)-direction: \( P_πσ_y = -σ_y, P_πσ_y = σ_y, \) and work out \( m_t = \int dΩ P(Ω)|⟨σ⟩| \) for the \( π \)-pulse, the \( T_2^* \)-decay has been eliminated, no term like \( e^{-iθn} \) in Eq. (10) appears here. Now \( Ω_0 \) and \( d \) enter only via \( m_t \). The structure of Eqs. (10) [20] is close to the one of Eqs. (10) [10], and the optimization over \( θ_1, θ_2 \) goes in the same way. To facilitate comparison, we take \( Ω_0 = 0 \), thus \( m_t = 0 \), and disorder strength \( d \) arbitrary large. recalling in addition Eqs. (10) [10] and the ergodic
condition $\Omega \gg 1$, we get

$$|m_2| = e^{-4\xi(\tau) + \xi(2\tau)} \sin \left\{ \gamma \left[ 2 \text{arctan}(\tau \Gamma) - \text{arctan}(2\tau \Gamma) \right] \right\},$$

where we already inserted the optimal values $\vartheta_1 = \vartheta_2 = -\frac{\pi}{4} = \frac{\pi}{4}$. The choice of optimal pulses can be the same as for the two-pulse scenario. The maximal $|m_2|$ can exceed 0.4 for sufficiently strong coupling and/or low temperatures. The results improve by applying a sequence of three (spin-echo) pulses separated from each other by a time much larger than $T_2$, see Table I.

A $1/f$ spectrum is another relevant situation of the bath-spin interaction recently observed in a two-level system (spin) of charge states in Josephson-junction circuit (Cooper-pair box) [16]. The spin’s interaction with the bath of background charges is modelled via [15, 16]

$$J_f(\omega) = \frac{b_f}{\pi} e^{-\omega/\Gamma} \theta(\omega - \Lambda),$$

where $b_f$ is the coupling constant, $\theta$ is the step function, and where $\Gamma$ and $\Lambda$ are, respectively, the largest and smallest frequencies of bath’s response. The upper frequency $\Gamma$ is not relevant: the inhomogeneous broadening). We are not aware of other types of pulses.

The fields alone cannot cool [10], while the bath alone can prepare the optimal value zero to 0 for sufficiently strong coupling and/or low temperatures. The results improve by applying a sequence of three (spin-echo) pulses separated from each other by a time much larger than $T_2$, see Table I.

In conclusion, we described a new method for cooling spins due to common action of external fields and a bosonic bath, starting from the overall equilibrium state. The fields alone cannot cool, while the bath alone can generate only the standard decoherence [12]. As compared to existing methods [2, 3, 4, 5, 6, 7, 8, 9], the present one assumes neither already existing high polarization [2, 3, 4, 5, 6, 7, 8, 9], nor controlled spin-spin or bath-spin interactions [1], nor a low-temperature bath. It works even for very weak dc fields and applies to an ensemble of spins having completely random frequencies (strong inhomogeneous broadening). We are not aware of other methods achieving such a goal. The spins are cooled at once (not one by one) and the cooling process takes a time shorter than $T_2$. Together with the overall efficiency of the method, see the figures and the Table, these features are encouraging for applications, e.g., in NMR spectroscopy. Our basic assumptions are a decoherence time $T_2$ much smaller than the energy relaxation time $T_1$, and the availability of sharp and strong pulses acting on the spin. A long $T_1$ time characterizes other methods [2, 3], while strong and short pulses were used for a clean demonstration of the effect, which probably survives for other types of pulses.

The origin of the present mechanism lies in shifting the spin’s frequency due to backreaction of the spin on the bath. This dynamical effect requires a non-perturbative treatment of the bath-spin interaction and is usually missed by markovian approaches [15]. It operates on a specific time-scale and allows to cool the spin provided the proper, coherence generating, sequence of external pulses is chosen. The cooling is efficient already for small-to-moderate bath-spin couplings, and is especially visible for situations where a strong bath-spin coupling is inherent ($1/f$-noise). In this experimentally realized situation [10] the cooling mechanism is expected to be feasible.

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[18] G. Lindblad, Non-Equilibrium Entropy and Irreversibil-