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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 sizable 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.

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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 [1]. The related problem of cooling spins is equally known; it originated as an attempt to improve the sensitivity of NMR/ESR spectroscopy [2–7], since in experiments the signal strength is proportional to the polarization. Recently it got renewed attention due to realizations of setups for quantum computers [8]. 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 biology. At temperature $T = 1$ K and magnetic field $B = 1$ T the equilibrium polarization of a proton is only $\tanh^{1/2} \frac{\mu B}{2} \approx 10^{-3}$ 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 $^{15}\text{N}$ it is $10$ times smaller. The weak polarization can be compensated by a large number of spins, but for some NMR isotopes the natural abundance is too low (0.36% for $^{15}\text{N}$).

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 [2–7]. Specially known are methods where a relatively high polarization is transferred from one place to another, e.g., from electronic to nuclear spins [2–6]. In this respect, electronic spins play the same role as the zero-temperature bath of vacuum modes employed for laser cooling of atoms [1] (this bath is typically inadequate for cooling nuclear spins, but was employed to study cooling of atomic few-level systems [9]). Polarization transfer was studied in various settings both theoretically and experimentally [2–6]. 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 of manipulating a set of $n$ spins, each having a small polarization, in such a way that the polarization of one spin is increased at the expense of decreasing the polarization of the remaining $n-1$ [7]. Since spins are cooled one by one, a long time is needed for cooling a large ensemble.

Here we propose a mechanism of cooling that uses only the most standard setting of NMR or ESR physics [2–4]: spins-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 [10]. However, we assume that the bath is not under any direct control nor are there 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. One can also cool at once a completely disordered ensemble of spins.

The model we study is well known [11–15]: a spin-1/2 with energy levels $\pm \hbar \Omega$ 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

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

Here $\omega_k$ are the bath frequencies, $\hat{\sigma}_{x,y,z}$ are the Pauli operators, and $\hat{X} = \sum_k \hbar g_k (\hat{a}_k^\dagger + \hat{a}_k)$ is the collective coordinate of the bath. The interaction is chosen assuming that the $T_1$ time, connected to relaxation of the average $\langle \hat{\sigma}_z \rangle$ is very large (infinite) [2–4]. The $g_k$ are couplings parameterized via the spectral density function $J(\omega)$:

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

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 (1) is solved [12]: \( \hat{\sigma}_z \) is conserved, while \( \hat{a}_k(t) = e^{-i\omega_k t}\hat{a}_k(0) + \frac{\hbar\omega_k}{2\omega_i}(e^{-i\omega_i t} - 1) \). This leads along with Eqs. (1) and (2) to

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

where \( \hat{\eta}(t) \) is the quantum noise operator, and where \( F(t) = \frac{\hbar}{2} F(t) \) quantifies the backreaction of the spin on the collective operator of the bath. This effect, not relevant for decoherence 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:

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

where \( T \equiv 1/\beta \) is the temperature. \( \rho(0) \) 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. As follows from Eq. (6), \( \hat{\eta}(t) \) is a Gaussian operator with \( \langle \hat{\eta}(t) \hat{\eta}(0) \rangle = \hat{\xi}(t) - i\dot{F}(t) \), \( t > 0 \), where \( \langle \cdot \cdot \rangle \) is taken over \( \rho(0) \), and where

\[
\hat{\xi}(t) = \int_0^\infty d\omega J(\omega) \frac{1 - \cos \omega t}{\omega^2} \coth \frac{\hbar \omega}{2T}.
\]

The Heisenberg equation of the spin, \( \hbar \dot{\hat{\sigma}}_z = i[\hat{\sigma}, \hat{H}]_z \), with \( \hat{\sigma}_z = \hat{\sigma}_x \pm i\hat{\sigma}_y, \hat{\sigma}_x \hat{\sigma}_z = \pm \hat{\sigma}_z \), is solved as

\[
\hat{\sigma}_z(t) = e^{\frac{1}{2}i\Omega(t-t_0)-iF(t-t_0)} e^{\frac{1}{2}i\int_{t_0}^t d\hat{s}(s) \hat{\sigma}_z(s_0)},
\]

where \( \hat{e} \) is the time-ordered exponent. Defining \( \hat{E}_i \hat{A} = \hat{E} e^{i\Omega t/\hbar} \hat{A} e^{-i\Omega t/\hbar} \) for any operator \( \hat{A} \), one derives

\[
\hat{E}_i \hat{e} = e^{\frac{1}{2}i\int_{t_0}^t d\hat{s}(s)} e^{\frac{1}{2}i\Omega(t-t_0)} e^{i\eta_{i}(t_0,t_0)},
\]

where \( \chi(t_1,t_2,t) = F(t_2) - F(t_1) + F(t_1 + t) - F(t_2 + t), \)

\[
\langle \hat{E}_i \hat{e} \rangle = e^{-\frac{1}{2}i\int_{t_0}^t d\hat{s}(s)} = e^{-\frac{1}{2}i\int_{t_0}^t d\hat{s}(s)} + i\int_{t_0}^t d\hat{s}(s),
\]

The factor \( e^{-\frac{1}{2}i\chi(t)} \) leads to decoherence [12], since due to Eqs. (8) and (11) \( \langle \hat{\sigma}_z(t) \rangle = e^{-\frac{1}{2}i\Omega t} \langle \hat{\sigma}_z(0) \rangle \) 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 \( \hat{H}(t) = \hat{H} + \hbar\hat{a}\hat{\sigma}_z \). In the pulsed regime [2–4,13–15] \( \hbar(t) \) differs from zero only for very short intervals of time \( \delta \) such that \( \hbar(t) \delta \sim 1 \), 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. We parametrize it as \( \hat{U} = e^{i\hbar(t/\delta)}\hat{\sigma} / \delta (0 \leq \phi, \psi \leq 2\pi, 0 \leq \theta \leq \pi / 2), \)

\[
\hat{U} = \left( \begin{array}{cc}
 e^{-i\phi} \sin \theta & e^{-i\phi} \cos \theta \\
 e^{i\phi} \sin \theta & e^{i\phi} \cos \theta
\end{array} \right).
\]

As a first example, we take Ohmic interaction [11]

\[
J(\omega) = \gamma \omega e^{-\omega / \Gamma}, \quad \Theta = \frac{T}{\hbar \Gamma},
\]

where \( \gamma \) is a dimensionless coupling constant, and where \( \Gamma \) (usually \( \gg \Omega \)) is the bath’s response frequency. Now

\[
\hat{\xi}(t) = \gamma \ln \left[ \frac{\Gamma(t + \theta)}{\Gamma(t + \theta) + \Gamma(t + \theta)} \right],
\]

where \( \Gamma \) is Euler’s function and \( \Theta \) is the dimensionless temperature. The time scale \( 1 / \Gamma \) of backreaction \( F(t) \) is \textit{temperature independent} as opposed to the decoherence time. For low temperatures \( \Theta \ll 1 \), \( e^{-\xi(t)} \) is practically \( 1 + (\Gamma^2/2)^{-}\gamma / 2 \), with \( \Theta \gg 1 \), \( e^{-\xi(t)} \) starts as a Gaussian, but continues as \( e^{-\xi(t)/\gamma} \) with \( \gamma = \hbar / (2\gamma T) \).

\textbf{Cooling} means to get a final polarization \( \langle \hat{\sigma}_z \rangle \) more negative than the initial one, \( \langle \hat{\sigma}_z \rangle = -\tanh^2(\beta \hbar \Omega) \) [Eq. (6)] implies \( \langle \hat{\sigma}_z \rangle = \langle \hat{\sigma}_z \rangle = 0 \). One pulse cannot achieve cooling since it sees the initial equilibrium state of the spin, and then according to the no-cooling principle [10] it can heat only the spin’s state: for an arbitrary pulse \( \mathcal{P}_1 \) applied at time \( t \), \( \langle \hat{\sigma}_z(t) \rangle = \langle \hat{\sigma}_z \rangle = \langle \hat{\sigma}_z \rangle \cos 2\theta_1 \geq \langle \hat{\sigma}_z \rangle \) (recall \( \langle \hat{\sigma}_z \rangle \leq 0 \)). Thus we need at least two pulses. The final polarization after one pulse at \( t \) and one at \( t + \tau \), \( \mathcal{P} = \mathcal{P}_1 \mathcal{P}_2 \mathcal{P}_1 \), reads from Eqs. (8)–(12)

\[
\langle \hat{\sigma}_z \rangle = \langle \hat{\sigma}_z \rangle \cos 2\theta_1 \cos 2\theta_2 + s_2 \sin 2\theta_1 \sin 2\theta_2,
\]

where \( s_2 = -e^{-\xi(t)} \Re \left[ e^{i\Omega t/\hbar} \langle \hat{\sigma}_z \rangle \cos \chi + i \sin \chi \right] \).

\textbf{For all results below we set} \( \Gamma = 1 \) (a mild condition, since \( \Gamma \ll 1 \) is typically the shortest time scale), since in this \textit{ergodic limit}, the initial condition (6) is equivalent to the overall equilibrium preparation \( \rho_{eq}(t) \propto e^{-\beta \hat{H}} \).

In Eq. (16), \( s_2 \) can always be made negative by tuning \( \alpha_2 \). Minimizing \( \langle \hat{\sigma}_z \rangle \) over \( \theta_1, \theta_2 \) produces \( \min \langle \hat{\sigma}_z \rangle, s_2 \). If the initial polarization is already high, \( \langle \hat{\sigma}_z \rangle \gg |s_2| \), no pulses should be applied, since they heat only the spin. However, in the relevant situation \( \langle \hat{\sigma}_z \rangle \approx 0 \), the minimum \( \langle \hat{\sigma}_z \rangle = s_2 \) is reached for \( \theta_1 = \theta_2 = \pi / 2 \).

Altogether, using Eq. (10) and \( \Omega \ll \Gamma \) yields
The collective variables are obtained by averaging the environment or due to inhomogeneous external field \[\{2,4\}\].

Transversal components generated by the two pulses decay after a time \(T_2\), and the spin is 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 \(\Omega\) by the factors \(F \sim \gamma \Gamma\) and \(\chi\) [recall Eqs. (8) and (10)], which arise from the (via the pulses) enhanced backreaction of the spin on the collective coordinate \(\hat{X}\) of the bath. The generation of coherences by the first \(\frac{\pi}{2}\) pulse is necessary to couple \(\hat{\sigma}_z\) to the bath, which “thermalizes” \(\langle \hat{\sigma}_z \rangle\) under the shifted frequency. Cooling is achieved via the proper pulses; still it decreases with \(\gamma\) (weaker backreaction) and with \(\beta\) (larger decoherence). The time \(\tau\) between two pulses can be neither too short \([\chi\text{ is visible on the time scale }1/(\gamma \Gamma)]\) 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 \(\approx T_2\), so that the transversal components decay, \(\langle \hat{\sigma}_z \rangle \to 0\), applying another two pulses, and so on \(n\) times. The final \(\langle \hat{\sigma}_z \rangle\) is maximized over all free parameters. It appears that this numerical maximization can be done locally, i.e., by maximizing the output \(\langle \hat{\sigma}_z \rangle\) after each pair of pulses. This “greedy” optimization shows up also in [9].

Inhomogeneous broadening.—Many experiments in NMR/ESR are done with an ensemble of noninteracting spins having random frequencies \(\Omega\) 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: \(m_i = \int d\Omega P(\Omega)\langle \hat{\sigma}_z \rangle_i\). Assume that the distribution of \(\Omega\) is Gaussian with average \(\bar{\Omega}\) and dispersion \(d:\ P(\Omega) \propto e^{-(\Omega-\bar{\Omega})^2/(2d)}\). Averaging over \(P(\Omega)\) the term \(e^{i\Omega \tau}\) in Eq. (16) produces a factor \(e^{-d\tau^2/2}\), a strong decay on times \(T^*_2 \approx 1/\sqrt{d}\). After this decay, \(s_2 \to 0\) and no cooling is possible as seen from Eq. (15).

It is possible to use the spin-echo effect and cool, i.e., increase the collective final polarization \(|m_i|\) as compared to the initial \(|m_i|\), even for a completely disordered ensemble with \(T_2^*\) being very short: Apply precisely in the middle of the two pulses an extra \(\pi\) pulse in the \(x\) direction: \(P_\pi \hat{\sigma}_x = -\hat{\sigma}_x\), \(P_\pi \hat{\sigma}_x = \hat{\sigma}_x\), and work out \(m_i = \int d\Omega P(\Omega)(\xi, P_\pi, \xi, P_\pi \hat{\sigma}_x)\):

\[
m_i = -m_i \cos 2\theta_1 \cos 2\theta_2 + s_3 \sin 2\theta_1 \sin 2\theta_2,
\]

where \(s_3 = -e^{-4\xi(t)/2} \Re\{e^{i\alpha_3}(m_i \cos \chi_3 - i \sin \chi_3)\}\),

where \(\alpha_3 = \phi_1 - \psi_1 - \phi_2 - \psi_2\) and \(\chi_3 = \chi(0, \tau, t) - \chi(\tau, 2\tau, t).\) As compared to (16), both the decoherence \(e^{-4\xi(t)/2}\) and the backreaction \(\chi_3\) term are different. In the Gaussian regime \(\xi \approx \beta^2\) decoherence is absent [14,15,17]. Because of the \(P_\pi\) pulse, the \(T_2\) decay has been eliminated, no term like \(e^{-\Delta \tau}\) in Eq. (16) appears here. Now \(\Omega_0\) and \(d\) enter only via \(m_i\). The structure of Eqs. (18) and (19) is close to the one of Eqs. (15) and (16), and the optimization over \(\theta_1, \theta_2\) goes in the same way. To facilitate comparison, we take \(\Omega_0 = 0\); thus \(m_i = 0\), and disorder strength \(d\) is arbitrarily large. Using Eqs. (10) and (13) and the ergodic condition \(\Delta \gg 1\), we get

\[|m_i| = e^{-4\xi(t)/2} \sin[2\arctan(\gamma \Gamma) - \arctan(2\pi \Gamma)],\]

where we already inserted the optimal values \(\theta_1 = \theta_2 = -\frac{\pi}{4}\). The choice of optimal pulses can be the same as for the two-pulse scenario. The maximal \(|m_i|\) 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 \(2T_2\); 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) [17]. The spin’s interaction with the bath of background charges is modeled via [15,17]

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

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 the bath’s response. The upper frequency \(\Gamma\) is not relevant: the integrals in Eqs. (5) and (7) converge for \(\Gamma \to \infty\) [15]. Within the realization of [17]: \(\Gamma \to \infty(\gg 10^{11}\text{ Hz})\), \(\Lambda = 3144\text{ Hz}, \hbar \Omega = 34 \mu\text{eV},\) while the relevant dimensionless coupling constant is very large \(\gamma_f = b_f/\Lambda^2 = 2.3 \times 10^{11}\). Thus the cooling effect is especially visible here. The spin-phonon model does not describe all the aspects of the decoherence in the Cooper-pair box; see [18] for more elaborate approaches. Still, some important features are reproduced adequately [15,17]. We work out Eqs. (5), (7), (15), and (16) under the following conditions: (i) high
The cooling process takes a time shorter than \( T/(\hbar \Lambda) \gg 1 \) (pessimistic case); (ii) the temperature \( \Theta_f = T/(\hbar \Lambda) \approx 1 \) (experimentally relevant regimes [17]); (iii) \( \langle \sigma_z \rangle_t \approx 0 \) due to large temperatures; (iv) \( \Omega \tau \ll \gamma_f / \Lambda \) due to large \( \gamma_f \). In analogy to Eq. (17), the result is \( \langle \sigma_z \rangle_t = e^{-\theta} y^{3/2} \sin(y - \pi y'/4y) \), where \( y = \gamma_f \Lambda \tau \) is a dimensionless time, and where we omitted terms \( O(y^3/y^2) \). As seen in Fig. 1, the polarization can increase from its original value zero to 0.997 for \( \Theta_f / \gamma_f = 0.01 \), which corresponds to \( T = 15 \text{ K} \) [17].

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 [10], while the bath alone can generate only the standard decoherence [12]. As compared to existing methods [2–7,9], the present one does not assume already existing high polarization [2–6] nor controlled spin-spin or bath-spin interactions [7] nor a low-temperature bath [9]. 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 \( T_2^{\perp} \). 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 \( T_2^{\perp} \) much smaller than the energy relaxation time \( T_1^{\perp} \), and the availability of sharp and strong pulses acting on the spin. A long \( T_1^{\perp} \) time characterizes other methods [2,3], while short and strong 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 nonperturbative treatment of the bath-spin interaction and is usually missed by Markovian approaches [19]. It operates on a specific time scale and allows the spin to cool 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 [17] the cooling mechanism is expected to be feasible.

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### Table 1

Maximum value of \(|\langle \sigma_z \rangle_t \rangle/(2, 3, \text{ 50 } \times \text{ 2 pulses})

| \( \gamma \) | 0.1 | 0.1 | 1 | 1 | 10 | 10 |
|---|---|---|---|---|---|---|
| \( \Theta_f \) | 0.1 | 0.1 | 1 | 1 | 1 | 1 |
| 2 | 0.1126 | 0.0826 | 0.4930 | 0.3516 | 0.8910 | 0.7931 |
| 3 | 0.2036 | 0.1369 | 0.7250 | 0.4732 | 0.9550 | 0.9025 |
| \( 50 \times 2 \) | 0.2779 | 0.1895 | 0.6948 | 0.5430 | 0.9509 | 0.8995 |
| \( 2 + \pi \) | 0.0834 | 0.0604 | 0.2498 | 0.2026 | 0.4634 | 0.4155 |
| \( 50 \times [2 + \pi] \) | 0.1396 | 0.1055 | 0.3119 | 0.2639 | 0.5214 | 0.4834 |

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