Cooling down quantum bits on ultrashort time scales

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Abstract. Quantum two-state systems, known as quantum bits (qubits), are unavoidably in contact with their uncontrolled thermal environment, also known as a macroscopic ‘bath’. The higher the temperature of the qubits, the more impure their quantum state and the less useful they are for coherent control or quantum logic operations, hence the desirability of cooling down the qubits as much and as fast as possible, so as to purify their state prior to the desired operation. Yet, the limit on the speed of existing cooling schemes, which are all based on Markovian principles, is either the duration of the qubit equilibration with its bath or the decay time of an auxiliary state to one of the qubit states. Here we pose the conceptual question: can one bypass this existing Markovian limit? We show that highly frequent phase shifts or measurements of the state of thermalized qubits can lead to their ultrafast cooling, within the non-Markov time domain, well before they re-equilibrate with the bath and without resorting to auxiliary states. Alternatively, such operations may lead to the cooling down of the qubit to arbitrarily low temperatures at longer times. These anomalous non-Markov cooling processes stem from the hitherto unfamiliar coherent quantum dynamics of the qubit–bath interaction well within the bath memory time.
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### 1. Introduction

How can an open quantum system change its temperature and entropy (synonymous with its state purity) prior to reaching a new equilibrium with the bath that thermalizes it? A standard means of qubit cooling employs transitions from the upper state of the qubit to an auxiliary intermediate state, which then radiatively decays to both qubit states \([1]–[8]\). This cooling can be fast but is still much slower than the intermediate-state radiative lifetime (typically \(\mu s\) or longer). More dramatically, there may not always be an intermediate state appropriate for cooling. Thus, free spin-1/2 systems in magnetic fields have only two levels.

An alternative is provided by *selective* quantum measurements of the energy: their results may be read out and sorted by ‘Maxwell’s demon’ \([9, 10]\) into ground-\(|g\rangle\) and excited-\(|e\rangle\) state subensembles, and the \(|e\rangle\)-subensemble is then discarded. This process may be continued...
until enough qubits are accumulated in the \(|g\rangle\) state. Yet, the feasibility of such a purification depends on the setup, and so does its efficiency and speed.

Conceptually simpler suggestions for fast changes in system temperature forego the use of auxiliary states or Maxwell-demon sorting. Instead, it has been suggested that the system–bath coupling be switched off, then on again, impulsively \([11, 12]\). Yet, this suggestion needs to be clarified: if indeed this off–on sequence is so fast that the system–bath correlations (entanglement) remain intact, this succession of effecting and then reversing a unitary change in the system–bath coupling may have no net effect on the system. This prompts the basic question: can unitary, reversible, operations change the system temperature or entropy?

An attempt to answer this question within a conceptually simple framework for quantum cooling, avoiding the use of auxiliary states or Maxwell-demon selection, is the motivation for the present work. We show that quantum non-demolition (QND) unitary or non-unitary operations, i.e. operations that commute with the operator \(\sigma_z\), if performed at the right rate, enable gradual purification of qubits coupled to a non-Markov bath, although they are non-selective, i.e. their results are neither read out nor acted upon.

As a rule, the tools of quantum thermodynamics \([13]–[17]\) and their application to cooling \([18]–[22]\) are based on long-time Markovian (Lindblad) master equations (MEs) that describe convergence at a constant rate to equilibrium, i.e. to ‘detailed balance’ between the qubit levels. By contrast, the understanding of non-Markovian, short-time effects in quantum thermodynamics is scant at best.

We have recently explored one such non-Markovian process \([23]\): an impulsive (brief), QND non-selective measurement of the qubit energy disturbs its equilibrium with the bath, thereby abruptly changing its temperature. The subsequent evolution of the measured qubit in the presence of the bath alternates between heating and cooling at times comparable to the qubit oscillation period. Such effects are at odds with the standard (Markovian) notions of thermodynamics \([13]–[17], [22]\), whereby temperature and entropy must monotonically converge to their equilibrium values.

Here, we investigate cooling on arbitrary time scales, without the Markov or Lindblad constraint. Our goal is to explore the tradeoff between cooling ‘well’ (i.e. to ultralow temperatures) and ‘fast’ (i.e. well within the system–bath equilibration time). To this end, we undertake a comprehensive quest for an effective and fast cooling of an initially thermalized qubit by comparing unitary with non-unitary (projective) QND operations along the \(z\)-axis of the Bloch sphere \([24]\).

If the qubit were isolated, these operations would not change the excitation, by the definition of QND. Yet, since the qubit interacts with a bath via coupling that is necessarily perpendicular to \(z\), e.g. \(\sigma_x\) coupling (so as to allow system–bath excitation exchange), \(z\)-axis operations lead to alternation between heating and cooling, depending on the time between consecutive operations. This behaviour is shown to hold also in the presence of dephasing (\(\sigma_z\)-noise).

Our quest reveals a universal bound on the largest qubit cooling/purification achievable by any means of non-selective QND evolution control for a given (weak) coupling to the bath. This bound can be approached very fast, within the bath memory time, by repeated impulsive disturbances of the equilibrium, intermittently with either free evolution or continuous (driven) modulation of the qubit levels. Remarkably, we find that this non-Markov universal bound allows cooling down to ultralow temperatures in realistic scenarios.
The attainment of this bound requires that the time resolution of the disturbances be comparable to a period of the qubit natural oscillation. Such QND disturbances are already being implemented (but not for cooling): ps optical pulses at sub-ns intervals satisfy the required time resolution when acting on qubits with MHz or GHz transition frequencies. The qubit states must be discriminated by the pulsed-QND disturbances according to their distinct symmetries, but not their energies, which are ill-defined during the pulses. Hence, the present approach is applicable to atoms, molecules or quantum dots with microwave transitions that are coupled to phononic, plasmonic or photonic cavity reservoirs.

Although the present strategy may be compared with sideband cooling or subensemble selection on a case-by-case basis, there is a more fundamental reason for its analysis. It can be viewed as part of an effort to unravel quantum thermodynamic anomalies, i.e. deviations from standard thermodynamics [13]–[17], on short non-Markov time scales under the least possible intervention [23]: allegedly non-intrusive (QND) observations and other non-selective QND operations turn out to be an effective method of steering the system towards a desirable (here ground) state.

2. Concept and outline

Having motivated our choice of non-selective QND cooling, we now outline the principles and physical content of its analysis.

2.1. Non-selective QND measurements versus phase shifts

The effect of non-selective QND measurements is to erase the qubit–bath correlations (entanglement) that exist at thermal equilibrium, thereby transforming their joint density matrix into an approximately factorized form (Supplementary I, available from stacks.iop.org/NJP/11/123025/mmedia). Phase shifts retain the system–bath correlations but modify the off-diagonal elements of their joint density matrix (Supplementary I, appendix A). Either operation results in a non-equilibrium state that starts evolving (sections 3.1–3.3).

2.2. Alternation between heating and cooling

The reduced density matrix of the qubit remains diagonal (in the \(H_S\) eigenbasis) throughout the considered evolution (Supplementary I and II, available from stacks.iop.org/NJP/11/123025/mmedia) [23]. Hence, it can always be written in Gibbs form: \(\rho_S(t) = Z^{-1} e^{-\beta(t)H_S}\), where \(\beta(t)\) is the time-dependent effective inverse temperature. This allows us to consider the dynamics in terms of the qubit temperature, i.e. its ‘heating’ and ‘cooling’.

Shortly after a non-selective QND operation, the qubit is shown to be hotter, i.e. more impure than before the operation (appendix A). How does this trend reverse to allow cooling? The answer lies in the structure of the system–bath interaction Hamiltonian. It can always be decomposed into rotating-wave (RW) near-resonant and counter-rotating (CR) anti-resonant terms (section 3). RW terms oscillate much more slowly than CR terms (in the interaction picture); hence RW terms are effective at longer times. A basic insight that stems from our analysis is that CR terms lead to heating shortly after the disturbance, but once RW terms take over, cooling becomes possible (section 4, appendix B).
2.3. **Time scales for QND-induced cooling**

What are the relevant time scales of QND-induced cooling? Such cooling is shown to take place for QND disturbances separated by time scales ranging from times comparable to \(1/\omega_a\) (the qubit oscillation period) up to \(t_c\), the bath memory time, i.e. well within the non-Markov time domain (sections 3 and 4). Proper dephasing is therefore typically unimportant, since it occurs on much longer time scales, \(T_2 \gg t_c\) (section 4.3).

2.4. **Physical picture**

The underlying physics of QND-induced cooling is shown to be the following (section 4.4).

(i) At very short times (below the inverse qubit energy, \(t \lesssim 1/\omega_a\)) all the oscillators of the bath act in unison, and thus give rise to a much faster rate of change of the qubit population compared to the Markovian (Golden Rule) limit, \(1/T_1\), wherein only the bath oscillator resonant with the qubit determines the variation of qubit level populations. On non-Markov time scales in between these extreme limits, bath oscillators detuned from the qubit energy contribute as well.

(ii) The balance between the effects of near-resonant (rotating) and anti-resonant (CR) terms in the system–bath interaction Hamiltonian determines the non-Markov heating or cooling. As the rotating terms dominate the dynamics on time scales \(t \gtrsim 1/\omega_a\), this represents exchange (swap) of energy between the qubit and the many bath oscillators that contribute to the process, according to their coupling strength. The strongly coupled bath modes near \(\omega_a\) have larger energy (lower effective temperature) than the qubit. The rotating terms drive the qubit towards swapping its high-temperature state with the low-temperature state of the detuned bath oscillators. As time progresses, swaps with different bath modes add up destructively, thereby reducing the total exchange (cooling) rate until finally attaining the Markovian equilibration rate.

(iii) Since the interaction of the qubit with effectively cold bath modes is not resonant, there will not be a complete qubit–bath swap of temperature states in a given cycle. To attain complete swap, one needs to perform repeated disturbances (measurements or phase shifts), on time scales of the qubit–bath maximal exchange, finally leading to a temperature where the exchange can no longer cause any further cooling. It may then seem that a completely off-resonant bath (with spectrum centered at \(\omega_0 \gg \omega_a\)) can lead to stronger cooling of the qubit. Yet, with increasing detuning, \(\omega_0 - \omega_a\), the CR terms leading to heating become as important as the rotating terms leading to cooling, so that cooling and heating tend to cancel each other.

(iv) Consequently, to achieve a desired cold state while increasing the detuning one may have to perform an increasingly larger number of measurements, thereby slowing down the process (section 5).

Hence, the particular choice of scheme and parameters would depend on the tradeoff between two considerations: how fast and how pure one needs the qubit to be ‘initialized’ for the quantum operation at hand (sections 5.1–5.4).
2.5. Quantitative analysis

The analysis of the qubit evolution, either between QND disturbances or concurrently with continuous QND $\sigma_z$-driving, invokes the previously derived non-Markov ME [25]–[28]. For an initially thermalized qubit, this ME reduces to rate equations for the qubit-level populations, since no coherences arise under such disturbances. Their non-Markov nature is manifest from the oscillatory time dependence of the $|e\rangle \leftrightarrow |g\rangle$ transition rates that violate the Markovian detailed balance between the levels. Their time dependence reflects the temperature-dependent spectral distribution $G_T(\omega)$ of the bath oscillators about the mean frequency $\omega_0$ and the detuning of $\omega_0$ from the qubit resonance $\omega_a$, as explained above. These equations are accurate to second order in the system–bath coupling. They yield the full system dynamics, without resorting to the rotating-wave approximation (RWA) [29] (see below), within the ubiquitous weak-coupling regime. This regime is consistent with the Born approximation, whereby the bath is a true thermostat, unaffected by the system evolution [30].

As verified by our detailed perturbative analysis (Supplementary II) and supported by the excellent agreement with our ab-initio numerical results, deviations from the bound on cooling derived here are of fourth order in the system–bath coupling and thus are typically negligible.

3. QND disturbances of the qubit–bath equilibrium

Consider an ensemble of qubits, i.e. two-level systems, in thermal equilibrium with a bath. The total Hamiltonian has the form

$$H_{\text{tot}} = H_0 = H_S + H_B + H_{SB},$$

$$H_S = \frac{\omega_a}{2} \sigma_z, \quad H_B = \sum_k \omega_k b_k^\dagger b_k,$$

$$H_{SB} = \sigma_x \sum_k \eta_k f(b_k, b_k^\dagger).$$

Here ($\hbar = 1$) the qubit Hamiltonian $H_S$ is written in terms of the level separation $\omega_a$ and the Pauli-matrix $\sigma_z$; the bath Hamiltonian $H_B$ is a sum of $k$-mode excitation quanta with frequency $\omega_k$. The system–bath coupling Hamiltonian $H_{SB}$ is a $\sigma_x$ Pauli-matrix of the qubit coupled to the sum of given $k$-mode functions of the annihilation and creation operators of the bath, $b_k$ and $b_k^\dagger$, with coupling rates $\eta_k$. Our treatment, culminating in the universal cooling bound, does not depend on the bath commutator $[b_k, b_k^\dagger]$, nor on the spectra of $H_B$ and $H_{SB}$ (Supplementary II). Yet it is helpful to illustrate the treatment for the generic spin-boson model [30, 31], wherein the contribution of each mode in $H_{SB}$ is a sum of RW and CR terms with

$$H_{SB} = \sum_k \eta_k \left( \begin{array}{c}
(\text{RW}) b_k \sigma^+ + b_k^\dagger \sigma^- \\
(\text{CR}) b_k \sigma^- + b_k^\dagger \sigma^+
\end{array} \right).$$

Upon transforming (4) to the interaction picture, it is seen that the RW and CR terms oscillate as $\exp(+i(\omega_k - \omega_a)t)$ and $\exp(\pm i(\omega_k + \omega_a)t)$, respectively. The (slow) RW and (fast) CR
oscillations of the coupling energy have profoundly different effects on the dynamics, as shown below.

The initial, equilibrium, state of the total ensemble is the density matrix \( \rho_{\text{Eq}} \propto e^{-\beta H_{\text{tot}}} \) whose off-diagonal elements express quantum correlations between the system (qubit) and bath. These correlations are concealed when observing only the qubit state, \( \rho_S = \text{Tr}_B \rho_{\text{Eq}} \). Yet these correlations increase the temperature (mixedness) of \( \rho_S \) as the system–bath coupling grows (Supplementary I).

To purify the qubit state, one needs first to disturb this equilibrium state, by a Hamiltonian \( H_{\text{SD}} \) of a device briefly acting on the qubit. If the disturbance is to be a QND effect, so that \( \rho_S \) retains its \( \sigma_z \)-diagonal form as in equilibrium, one can choose between a unitary \( \sigma_z \)-rotation and a projective measurement in the \( \sigma_z \) (qubit energy)- basis. For both unitary and non-unitary QND disturbances, it is essential that \( H_{\text{SD}} \propto \sigma_z \) does not commute with \( H_{\text{SB}} \propto \sigma_x \), thus leading to non-classical thermodynamic effects. The alternative is \( H_{\text{SD}} \propto \sigma_x \), which exerts a classical-like force on \( \rho_S \), changing it in a non-QND fashion.

We prove in appendix A that an impulsive \( \sigma_z \)-disturbance always produces heating of the equilibrium state immediately thereafter. Numerical results [23] support this general conclusion for a disturbance of finite, albeit brief, duration. An impulsive QND disturbance triggers departure from equilibrium towards heating, but it is the subsequent (free or driven) evolution that may cause cooling through the system–bath energy exchange.

### 3.1. Projective measurements

A measurement of \( \sigma_z \) (figure 1(a)) is done upon projecting the total state onto the energy eigenstates, \( |e(g)\rangle \). The total state (of the system and bath combined) after such a measurement is changed from \( \rho_{\text{Eq}} \) to (appendix A)

\[
\rho_{\text{tot}} = \rho_{\text{Eq}} \rightarrow \rho_{\text{tot}}^M = \frac{1}{2} \left[ \rho_{\text{Eq}} + \sigma_z \rho_{\text{Eq}} \sigma_z \right].
\]

Physically, such measurements can be affected by a strong, pulsed probe that discriminates between \( |e\rangle \) and \( |g\rangle \) by their distinct symmetries (say, magnetic numbers) but not by their energies, which are ill-defined during the brief disturbance (figure 1(c)).

### 3.2. Unitary rotations (phase shifts)

The total state following a rotation of the qubit by phase \( \phi \) about the \( z \)-axis (figure 1(a)) is given by

\[
\rho_{\text{Eq}} \rightarrow \rho_{\text{tot}}^\phi = \exp(-i\phi\sigma_z/2)\rho_{\text{Eq}}\exp(i\phi\sigma_z/2).
\]

Physically, such a rotation corresponds to an ac Stark (or Raman) shift of the qubit level-separation imposed by an external field (figure 1(b)).

### 3.3. Unitary or projective disturbances?

Which of these two types of operation should be repeatedly used on the qubit state to maximize its purification? An impulsive measurement is tantamount (equations (5) and (6)) to the mean of an impulsive phase flip \( (\phi = \pi) \) and doing nothing to the equilibrium. Hence, a phase flip pumps into the system twice the energy of a projective measurement. More generally, the energy transfer to the system by an impulsive phase shift \( \phi \) scales as \( 1 - \cos \phi \) (appendix A). Does this
imply that (all else being similar) $\pi$-phase flips result in greater purification of the qubit than measurements or phase shifts (rotations) by smaller $\phi$?

On the other hand, in the case of rotation, the qubit–bath correlations are preserved, which makes the change reversible, whereas in a projection the correlations are irreversibly destroyed. How does this basic difference between measurements and phase shifts affect their respective cooling effects?

4. Qubit non-Markov evolution: heating or cooling?

To answer the foregoing questions, we shall next explore the free evolution of a qubit in the presence of the bath between impulsive $\sigma_z$ disturbances, be it measurements or phase shifts (the extension to continuously driven $\sigma_z$-modulation is outlined in appendix B).
We can then ask: when does the immediate heating caused by an impulsive disturbance stop and cooling (purification) begin? This question may be analyzed by resorting to the non-Markov ME [31] for the (σz-diagonal) qubit state $\rho_S(t) = \text{Tr}_B \rho_{\text{tot}}(t) = \rho_{gg}(t)\langle g|g \rangle + \rho_{ee}(t)\langle e|e \rangle$. This ME evolves $\rho_S(t)$ under the action of the general qubit–bath coupling Hamiltonian $H_{SB}$ in equation (3), to second order in the coupling strength, with any bath (Supplementary II).

As shown in Supplementary I, the purification estimated by the ME-based analysis can greatly exceed its inaccuracy, since this inaccuracy is of higher (fourth) order in the qubit–bath coupling strength than the predicted (second-order) purification/cooling. The ME adequacy is corroborated by its excellent agreement with ab-initio simulations in figures 2–5.

The equations of motion for the matrix elements of $\rho_S$ derived from the ME [23, 26] read

$$\dot{\rho}_{ee}(t) = -\dot{\rho}_{gg}(t) = R_g(t)\rho_{gg} - R_e(t)\rho_{ee},$$

(7)

where the transition rates that govern the free-evolution dynamics in the $n + 1$ interval after $n$ disturbances are $R_g(|e\to|g\rangle)$ and $R_e(|g\to|e\rangle)$, obtainable for different disturbances from the formula in [25].

Non-Markov cooling or heating are departures from Markovian detailed balance between $|e\rangle$ and $|g\rangle$ in (7), due to $R_{e(g)}(t)$ oscillations on the ultrashort time scales in question. These oscillations are partly lost in the standard RW approximation [31] whereby only the rotating (RW) terms are retained in the qubit–bath interaction Hamiltonian (4). We must account for both the RW and CR terms since they induce different oscillations of $R_{e(g)}(t)$. The role of these oscillations is revealed once we exactly integrate equations (7):

$$\rho_{ee}(t) = e^{-J(t)} \left[ \int_0^t dt' e^{J(t')} R_g(t') + \rho_{ee}(0) \right],$$

(8)

$$J(t) \equiv J_g(t) + J_e(t),$$

(9)

$$J_{e(g)}(t) = \int_0^t dt' R_{e(g)}(t')$$

(10)

$$= 2\pi t \int_{-\infty}^{\infty} d\omega G_T(\omega) F_t(\omega - t),$$

(11)

the upper (lower) sign standing for $e(g)$. The relaxation integrals $J_{e(g)}(t)$ in (10) are determined by the overlap of two functions:

(i) $F_t(\omega)$, the finite-time spectrum (finite-time Fourier transform) of the disturbances’ sequence, centered at $+\omega_0$ ($-\omega_0$) for $|e\rangle$ ($|g\rangle$) and characterized by its width $1/\tau$, the disturbances rate;

(ii) the temperature-dependent coupling spectrum (spectral response) of the bath [25, 31] $G_T(\omega)$. In the spin-boson model (4)

$$G_T(\omega) = (n_T(\omega) + 1)G_0(\omega) + n_T(-\omega)G_0(-\omega),$$

(12)

determined by $G_0(\omega)$, the zero-temperature coupling spectrum, with $G_0(\omega < 0) = 0$, and $n_T(\omega) = 1/(e^{\beta\omega} - 1)$, the temperature-dependent ($T = 1/\beta$) population of bath mode $\omega$. This spectrum can be crudely characterized by its mean frequency $\omega_0$ and width, $1/\tau$, the inverse correlation or memory time.

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Figure 2. Evolution of a qubit coupled to a bath consisting of $10^3$ oscillators under the Hamiltonian (4). Excited population evolution (as a function of $\omega_a t$) induced by only rotating (RW) terms (dash-dot), only CR terms (dashed) and the entire Hamiltonian (solid). The parameters are the for the Lorentzian coupling spectrum (30): $\eta = \omega_a/25$, $\beta \omega_a = 2$, $\Gamma = \omega_a/10$ and $\omega_0 = 2\omega_a$, where $\Gamma = 1/t_c$.

In what follows, we investigate the implications of (8)–(12) on cooling for repeated QND measurements and phase shifts. The off-resonant detuning $\omega_0 - \omega_a$ and width $1/t_c$ of $G_T(\omega)$ along with the disturbances rate $1/\tau$ will be shown to determine the cooling.

4.1. Measurements

During the $n+1$ interval after $n$ measurements separated by $\tau$, we have, in (7) and the integrand of (10), the transition rates

$$R_{\epsilon(g)}(t) = 2(t-n\tau) \int_{-\infty}^{\infty} d\omega G_T(\omega) \text{sinc} [(\omega \mp \omega_a)(t-n\tau)].$$

(13)

Here we used the approximation (Supplementary I) that each measurement decorrelates the qubit and the bath anew, and thus resets the time to $t-n\tau$; $\text{sinc}(\alpha) \equiv \sin(\alpha)/\alpha$, the upper (lower) sign standing for $R_{\epsilon}(R_g)$.

The spectral width of the sinc function is a measure of the energy uncertainty in the system at the short times considered. The overlap between the sinc function and the coupling spectrum (12), $G_T(\omega)$, oscillates with time, rendering the rates (13) strongly oscillatory at times $\sim 1/\omega_a$. These rates remain time dependent at longer but still non-Markovian times $1/\omega_a \ll t \lesssim t_c$, where $t_c$, the bath correlation (memory) time, is the inverse width of $G_0(\omega)$ [26], [31]–[34].

The corresponding spectral function in the relaxation integrals (10) is then

$$F^{M}_{\text{res}}(\omega) = \frac{\tau}{2\pi} \text{sinc}^2(\omega \tau/2).$$

(14)

The function $F_1(\omega + \omega_a)$ causes the faster, CR-induced, oscillation of $\rho_{ee}(t)$. It is peaked near the frequency $\omega_0 + \omega_a$, where $\omega_0$ is the peak of $G_T(\omega)$, and accounts for heating (figure 2). It is dominant only at very fast disturbance rates, or very short times, $\tau \ll 1/\omega_a$, consistently with the conclusion (discussed above) that heating prevails immediately after a disturbance.
As shown elsewhere [23], this time scale corresponds to the $t^2$-variation of $\rho^{(n)}_{ee}(t)$, the gist of the quantum Zeno effect [32]–[34].

As the faster oscillation averages out at lower disturbance rates or longer times, it is taken over by the RW-induced oscillation of $\rho_{ee}(t)$ near the low frequency $\omega_0 - \omega_a$. This oscillation is seen in figure 2 to be responsible for cooling. RW processes are resonant: any change in the qubit energy is compensated for by the bath. Hence we may conclude that non-Markov cooling of the qubit comes at the expense of the bath heating.

Although RW and CR oscillations occur on different time scales and are associated with cooling and heating, respectively, cooling can be properly analyzed only by allowing for both oscillations: the CR oscillation restricts the amount of cooling that might have occurred had only RW terms been in force (appendix B).

4.2. Phase shifts

Free evolution intermittent with $n$ impulsive phase shifts ($\phi$ rotations of $\sigma_z$) at intervals $\tau$ still conforms at the $n + 1$ interval to equations (7)–(12), but with transition rates that vary continuously, without being reset to zero every $\tau$. The resulting counterpart of (14), to be used in (10), is then

$$F_{\phi \tau}^\phi(\omega) = \frac{\tau}{2\pi} \sin^2(\omega \tau/2) \sin^2[n(\phi + \omega \tau)/2]/n \sin^2[(\phi + \omega \tau)/2].$$

The evolution is determined by the interplay between two effects, shown in figure 3(b). Namely, at short times, $t \lesssim 1/\omega_a$, $F_{\phi \tau}^\phi(\omega)$ causes $\sin^2$ oscillations of $J_{eg}(t)$. Upon comparing (14) with (15) we conclude that at such times, the detailed-balance modifications are rather similar for phase shifts and measurements. By contrast, at longer times, $t \gg 1/\omega_a$, $F_{\phi \tau}^\phi(\omega)$ in (15) becomes a weighted sum of $\delta(\omega + \phi/\tau)$ and $\delta(\omega - (2\pi - \phi)/\tau)$, i.e. of energy modulations caused by (ac-Stark) frequency shifts $-\phi/\tau$ and $(2\pi - \phi)/\tau$. For $\phi = \pi$, equation (15) becomes an equal sum of opposite shifts, $\omega \pm \pi/\tau$, whereas for $\phi \ll 1$ it then becomes a unidirectional shift, $\omega + \phi/\tau$ [26].

4.3. Concurrent dephasing

In most experimental scenarios the dephasing time, $T_2$, is much shorter than the Markovian decay time, $T_1$. Since the qubit density matrix remains diagonal throughout the evolution, the effect of proper dephasing is here rather unusual: it does not act on the qubit coherences, $\rho_{eg}$, but only on qubit–bath correlations that are destroyed anyhow by QND measurements (section 3, Supplementary I). This incremental effect of proper dephasing on the evolution of the diagonal qubit state is unimportant, as we now show.

Consider proper dephasing, i.e. a zero-mean time-dependent Gaussian stochastic fluctuation of the phase, $\delta_r(t)$, described by the following addition to the Hamiltonian:

$$H_D = \delta_r(t)\sigma_z,$$

with the first and second ensemble average (denoted by an overbar) moments given by

$$\overline{\delta_r(t)} = 0, \quad \overline{\delta_r(t)\delta_r(t')} = \frac{1}{T_2 t_0^d} e^{-(t-t')/t_0^d}.$$
Figure 3. (a) and (b) Overlap of the $F_t(\omega)$ functions and $G_T(\omega)$ in $J_e(t)$ and $J_g(t)$, for (a) measurements; (b) $\pi$ phase flips; (c) evolution of $\langle H_S(t) \rangle$ (green, solid), $\langle H_B(t) \rangle$ (red, dash-dot), $\langle H_{SB}(t) \rangle$ (blue, dashed) and $\langle H_{tot}(t) \rangle$ (black, dotted) under repeated measurements at intervals $\omega_a t = 2$. (d) Likewise, under repeated phase flips at intervals $\omega_a t = 2$. The energy was shifted such that $\langle H_S(0) \rangle = 0$ and $\langle H_B(0) \rangle = 0$, for convenience. Parameters are identical to figure 2.

where $t^d_c$ is the dephasing correlation time. The disturbance time scales that lead to non-Markov cooling are much faster than the dephasing time scales,

$$\tau \approx 1/\omega_a \ll t^d_c \ll T^2,$$

leading to the phase factor associated with $\delta_r(t)$:

$$e^{i \int_0^t dt' \delta_r(t')} = e^{-(t-t')/T^2}. \quad (19)$$

This exponential decay of the phase factor results in a Lorentzian spread of $1/T^2$ in the frequency domain. Hence, the spectral functions in the convolution (11) (equation (14) for measurements or equation (15) for phase shifts) become convolutions with the dephasing Lorentzian

$$F_{t}^{M(\phi)}(\omega) \rightarrow F_{t}^{M(\phi)}(\omega) \otimes \frac{1}{1 + (\omega T^2)^2}, \quad (20)$$

where $\otimes$ denote a spectral convolution. It is evident that the spectral spread due to either measurements, equation (14), or phase shifts, equation (15), at the rates or time scales required for cooling, is negligibly affected by the dephasing $1/T^2$. 

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4.4. How does non-Markov cooling occur?

We are now in a position to simply interpret and set the rules for non-Markovian cooling by various QND disturbances. We may depict the cooling process as the time-dependent redistribution of the excess energy imparted to the system plus bath by the impulsive disturbance. This redistribution causes the cooling of $\langle H_S \rangle$ to be the deficit between the heating of $\langle H_B \rangle$ and the cooldown of $\langle H_{SB} \rangle$, expressed by the following conservation law for their respective changes ($\delta$):

$$\delta\langle H_S(t) \rangle = - [\delta\langle H_B(t) \rangle + \delta\langle H_{SB}(t) \rangle].$$ (21)

The competing RW- and CR-induced oscillations affect this redistribution: RW-induced oscillations amount to $\delta\langle H_S \rangle = -\delta\langle H_B \rangle$, i.e. energy exchange between the system and bath only. By contrast, CR-induced oscillations cause rapid variation of $-\delta\langle H_{SB} \rangle$, resulting in heating, $\delta\langle H_S \rangle > 0$.

More insight is obtained into this exchange when considering free evolution between measurements. The relaxation integrals in (10) oscillate as $\text{sinc}^2[(\omega_a \pm \omega_0)\tau/2]$ at $\tau \leq 1/\omega_a$, $\omega_0$ being the peak frequency of the bath response. These underdamped frequency beats govern the alternating heating and cooling. They attest to the remarkably simple quasi-reversibility of the system–bath energy exchange, on the time scale of $\omega_a^{-1}$, irrespective of the bath complexity: all bath oscillators oscillate in unison in this time scale that is much shorter than the non-Markovian bath correlation (memory) time $c$ (figure 3(c)). Hence, their energy exchange with the system is much more effective than at longer (Markovian) times when the bath oscillators are out of tune. Although maximal cooling occurs at $t \simeq \pi/(\omega_a - \omega_0)$, poorer time resolution does not invalidate this cooling procedure, attesting to its robustness.

If instead of impulsive measurements one uses impulsive phase shifts, the resulting redistribution is determined by the level modulation of the qubit by spectral shifts $\pi/\tau$ (figure 3(d)): $\delta\langle H_S \rangle$ then undergoes parametric modulation, during its exchange with $\delta\langle H_B(t) \rangle + \delta\langle H_{SB}(t) \rangle$.

It is clear from figures 3(c) and (d) that a phase flip allows for more pronounced cooling than a measurement under similar conditions. If the interval $\tau$ is kept fixed, the amplitude of $\delta\langle H_S(t) \rangle$ oscillations is indeed maximized for phase flips compared to measurements or smaller phase shifts, as anticipated from the fact that the initial energy transfer is the largest for $\phi = \pi$. Yet, does this imply that the cumulative cooling following many disturbances is likewise maximized for phase flips? As shown below, this is not necessarily the case.

5. Cooling conditions

5.1. Universal bound

Applying $n$ repeated disturbances at constant intervals, $\tau$, and taking the limit $n \rightarrow \infty$ result in the asymptotic approach to a fixed point of the evolution, given by

$$\rho_{ee}(t = n\tau) \xrightarrow{n \rightarrow \infty} \frac{\int_0^\tau dt' e^{J(t')} R_e(t')} {\int_0^\tau dt' e^{J(t')} [R_e(t') + R_e(t')]} = \rho_{ee}^{(\infty)}(\tau).$$ (22)

Here $R_e(t)$, $R_e(t)$ and $J(t)$ are defined as in (10); for measurements and phase shifts we use (14) and (15), respectively. The extension to continuous modulations (appendix B) yields a similar expression.

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Thus, one obtains the following universal condition for qubit purification (cooling) that increases with $n$:

$$\rho_{ee}(n\tau) > \chi \equiv \min_{\tau} \left( \rho_{ee}^{(\infty)}(\tau) \right) \quad \forall n,$$

(23)

where the minimal achievable excitation, $\chi$ (corresponding to the coldest achievable temperature), is found upon varying $\tau$ (figure 4(a)).

Provided that the qubit–bath coupling is weak enough, $\epsilon = (\eta_k)_{\text{max}}/\omega_a \ll 1$ (cf (3)), equation (23) is simplified to

$$\rho_{ee}(n\tau) > \chi \approx \min_{\tau} \left( \frac{J_{\text{H}}(\tau)}{J(\tau)} \right) \quad \forall n.$$

(24)

The universal cooling bound expressed by (24) is the central general result of this paper. Remarkably, the coupling strength $\epsilon^2$ does not affect the cooling condition or the bound $\chi$, as both the numerator and denominator of (24) are proportional to it.

We now explicitly investigate this bound in the spin-boson model (4) for several limiting cases (for details cf appendix B).

(i) For repeated measurements, $n \gg 1$

$$\chi^M = \min_{\tau} \left( \int_{-\infty}^{\infty} d\omega G_T(\omega) \sin^2 \left( \frac{((\omega + \omega_a)\tau)}{2} \right) \right).$$

(25)

Figure 4. Main panel: excitation falloff to its minimum (cooling bound) $\chi^M$ (solid) for measurements, $\chi^n$ (dash-dot) for phase flips and $\chi^{n/10}$ (dashed) for small phase flips. Parameters are identical to figure 2. (a) Asymptotic fixed point, $\rho_{ee}^{(\infty)}(\tau)$, as a function of measurement intervals, $\tau$, with $\chi$ marked. (b) Excitation as a function of time for repeated measurements (blue) and phase flips (red), the same parameters except for $\eta = \omega_a/100$, $\Gamma = \omega_a/2$. Numerical simulations (dashed) for a 28-mode bath fully agree with the ME solutions (solid).
(ii) For π-phase flips after $n\tau \gg 1/\omega_a$, but assuming $\pi/\tau > \omega_a$, and taking the low-temperature limit,

$$\chi^\pi = \min_\tau \left( \frac{G_-(n_- + 1) + G_+ n_+}{G_+(2n_+ + 1) + G_-(2n_- + 1)} \right)$$

(26)

$$\beta \to \infty \min_\tau \left( \frac{G_+ + G_+ n_+}{G_+ + G_-} \right),$$

(27)

where $G_{\pm} \equiv G_0(\pi/\tau \pm \omega_a)$ and $n_{\pm} \equiv n_T(\pi/\tau \pm \omega_a)$. This result is an average between $n_- + 1 \approx 1$ and $n_+$, weighted by $G_-$ and $G_+$, respectively.

(iii) For repeated small phase shifts $\phi \ll 1$, after $n\tau \gg 1/\omega_a$, upon taking the low-temperature limit,

$$\chi^{\phi \ll 1} = \frac{n_T(\omega_a + \phi/\tau)}{2n_T(\omega_a + \phi/\tau) + 1} \beta \to \infty \to \left(1 + e^{\beta(\omega_a + \phi/\tau)}\right)^{-1}.$$ 

(28)

The minimal excitation $\chi$ (its general form (23) or its limits (24)–(28)) sets an upper bound on the achievable purity, $1 - \chi$, by any QND means, for a given bath temperature and coupling spectrum in the spin-boson model. It is to be contrasted with the Markovian cooling bound, set by the $|e\rangle \leftrightarrow |g\rangle$ transition rates $R_{e(g)}(t \to \infty)$:

$$\chi^{\text{Markov}} = \frac{n_T(\omega_a)}{2n_T(\omega_a) + 1} \beta \to \infty \to \left(1 + e^{\beta \omega_0}\right)^{-1}. $$

(29)

5.2. Spectral dependence and limit of the cooling

The tradeoff between the fastest cooling rate and the lowest temperature is now clear: whereas both $\chi^M$ and $\chi^\pi$ can yield ultrafast cooling, it is in fact $\chi^{\phi \ll 1}$ that is able to reduce the temperature at $t > t_c$ to the lowest limit. The small-φ limit (28) may allow cooling down to any temperature, provided $\beta \phi/\tau \gg 1$. Whereas the convergence to $\chi^M$ and $\chi^\pi$ is through oscillatory falloff of the excitation, $\chi^{\phi \ll 1}$ is approached through stepwise falloff by $\phi/\tau$ (figure 4).

The spectral requirements for cooling can be summarized as follows:

(i) Cooling below the bath temperature by frequent measurements (appendix B) requires that the bath coupling spectrum $G_0(\omega)$ have higher energy components than the level separation, i.e. $G_0(\omega > \omega_a)$ must be significant in order to allow cooling. For a bell-shaped $G_0(\omega)$ this means that the peak of $G_0$ at $\omega = \omega_0$ must be detuned above $\omega_a$ (figure 3(a)), so as to maximize $R_e(t)$ while minimizing $R_g(t)$.

(ii) For phase flips (equation (26)), because of the exponential nature of $n_T(\omega)$, we get cooling only if $G_+/G_- \gtrsim \exp(\beta \omega_0)$.

(iii) For cooling by small phase shifts (equation (28)), we must wait longer than $t \gg 1/|\omega_0 - \omega_a|$ so that (15) is narrow enough to avoid overlap with $G_T(\omega)$. Otherwise, equation (28) shows that $\chi^{\phi \ll 1}$ is insensitive to the coupling spectrum $G_0(\omega)$, as opposed to bounds (i), (ii), achievable by phase flips or measurements, respectively.
5.3. Temperature dependence of cooling

The qubit temperature at equilibrium is higher than that of the bath (Supplementary I) for appreciable coupling strength. We wish to not only undo the increase in qubit temperature caused by its coupling to the bath (Supplementary II), but also cool it below the bath temperature $T = 1/\beta$. As can be seen from figure 5, there is a temperature above which non-Markovian cooling (by phase flips more than by measurements) can take the qubit to a temperature lower than that of the bath. On the other hand, as the temperature approaches zero ($\beta \to \infty$), only the extra temperature of the qubit due to the coupling may be overcome by phase flips or measurements, but not the bath temperature: the net cooling that one obtains by these means is a non-monotonic function of temperature and coupling spectrum (figure 5, inset). By contrast, cooling by small phase shifts is a monotonic function of temperature: if $\beta \phi/\tau$ is large enough, an arbitrarily low temperature is attainable by the qubit.

5.4. Numerical estimates

For comparison of (22) and (23) with \textit{ab initio} numerical simulations (by the multiconfiguration time-dependent Hartree method [35]) (figures 2–5), we have taken a (modified) Lorentzian
coupling spectrum in the spin-boson model

\[ G_0(\omega) = \eta \frac{\omega}{\omega_0} \frac{\Gamma^2}{\omega^2 + (\omega - \omega_0)^2}, \]  

where \( \eta \) is the coupling strength, \( \omega_0 \) is the Lorentzian peak and \( t_c = 1/\Gamma \) is the memory time of the bath. The agreement is excellent indeed.

We have numerically verified (appendix B, figure B.1) the robustness of our results, which are valid for arbitrary control pulses, to variations in the pulse duration \( t_p \) and shape affecting the measurements and phase shifts, provided they adhere to the bound \( t_p \ll \tau \), \( \tau \) being the interval between successive operations in equations (13)–(28).

6. Experimental scenarios

Consider atoms, molecules or quantum dots in a bath with the finite-temperature coupling spectrum \( G_T(\omega) \) centered at \( \omega_0 \). Measurements or phase flips can be affected on such a qubit ensemble with resonance frequency \( \omega_a \) in the microwave domain, at time intervals \( \tau \) chosen according to equations (24)–(28), using optical Raman pulses. The QND probe pulses undergo different phase shifts \( \Delta \phi_e \) or \( \Delta \phi_g \) depending on the different symmetries of \( |e\rangle \) and \( |g\rangle \). The relative abundance of \( \Delta \phi_e \) and \( \Delta \phi_g \) after \( n \) disturbances would then reflect the ratio \( \rho_{ee}(t_n)/\rho_{gg}(t_n) \). QND probing or phase flips may be performed with time duration much shorter than \( \omega_a^{-1} \), on ps or fs time scales, without resolving the energies of \( |e\rangle \) and \( |g\rangle \). Appreciable cooling is then obtainable well within the equilibrium time, on the ps scale. By comparison, ordinary (optical) cooling [5]–[8] takes longer than the lifetime of the populated state decaying to \( |g\rangle \), i.e. \( \mu s \) at the shortest.

The lowest temperature is achievable for any bath by small phase shifts according to (28): if \( \beta \phi/\tau \gg 2 \), we may reduce the equilibrium excitation \( e^{-\beta \omega_a} \) by more than an order of magnitude.

6.1. Microcavities

A possible experimental scenario involves an atomic or molecular transition at frequency \( \omega_a \) (in the MHz range) that is weakly coupled to a near-resonant microwave cavity whose finite-temperature radiation constitutes the bath. The coupling of \( N \) atoms or molecules at a cavity antinode is \( N^{1/2} \eta_{\text{max}} / 2\pi \). An optical beam will rotate in polarization (figure 1(c)), thus performing an ultrafast QND measurement that resolves \( |g\rangle \) and \( |e\rangle \), which depends on their contrasting magnetic (m-) numbers, whereas their energies are unresolved. The optical Raman-induced Rabi frequency that can induce phase shifts or flips between \( |g\rangle \) and \( |e\rangle \) (figure 1(b)) satisfies \( \Omega_{\text{probe}} \gg \omega_a > N^{1/2} \eta_{\text{max}} \). A ps pulse train with such a Rabi frequency can drive the cooling at the optimal rate, as discussed above.

6.2. Quantum dots

A single-electron quantum dot may be coupled to a phonon bath. In an applied magnetic field of 7 T along the z-axis, the Larmor precession frequency is \( \omega_a = 26.3 \text{ GHz, } \hbar \omega_a = 0.2 \text{ meV,} \) so that we require probing pulses with \( \Omega_{\text{probe}} \gg 1 \text{ meV.} \) For ultrafast phase modulation in these systems, one can use the experimentally achievable pulses of 100 fs duration at 1 ps intervals [36, 37]. Since the resonant qubit separation \( \omega_a \) is of the order of a few GHz, one can achieve considerable cooling even within a \( \omega_a \)-period, i.e. on a ps time scale.
In quantum dots the relaxation time scale $T_1 \sim 1 \text{ ms}$ [38] is much longer than the decoherence time $T_2 \approx 10–100 \text{ ns}$ [39]. Yet, since qubit manipulations described above are on a ps time scale, $T_2$ processes do not affect the cooling.

7. Discussion

Our analysis reveals hitherto unknown aspects of qubit entanglement with the bath at equilibrium (Supplementary I) and of the short-time quantum dynamics induced upon disturbing this equilibrium (appendix A). On the fundamental side, we may conclude the following:

(i) The differences between unitary phase shifts or non-unitary (measurement) QND disturbances as concerns cooling have been elucidated (section 3).

(ii) The qubit dynamics induced by either projective measurements or phase shifts at ultrashort times comparable to the qubit period has been shown to be remarkably coherent, or quasi-reversible, and hence amenable to control of qubit temperature (section 4).

(iii) The universal bound on non-Markov cooling derived here and its corollaries (section 5) demonstrates the importance of the spectrum of the qubit coupling to the bath and the limitations of the RW approximation when designing efficient cooling/purification.

Several technical comments are in order:

(a) The analytical expressions for the cooling bound, equations (22)–(24), were derived via the second-order MEs [25, 26] (Supplementary II). While fourth-order MEs are known for the spin-boson model, the formulation of the time-dependent transition rates and the cooling bounds to fourth order is a further complication beyond the scope of this paper. Nevertheless, the excellent agreement with ab-initio numerical simulations [35] reinforce the validity of our results in the regime of system–bath weak coupling [26, 31].

(b) The role of the RWA was elucidated. While it is not valid for ultrashort-time disturbances, $t \leq 1/\omega_a$, it may still be valid for intermediate-time non-Markovian perturbations, $1/\omega_a \ll \tau \leq t_c$. The cooling bound retains its form within the RWA, albeit with quantitative modifications (appendix B).

(c) Proper dephasing was shown to be typically much slower than the time scales relevant to non-Markov cooling (section 4.3).

On the applied side, the following conclusions have been reached:

(i) The practical advantage of the predicted non-Markov effects is the previously unexplored tradeoff between ultrafast cooling or purification of qubits, which may be attained after several QND disturbances (section 5.1), and their cooling down to arbitrarily low temperatures at longer times (section 5.2). This tradeoff is shown to be strongly dependent on whether we use measurements, phase flips or small phase shifts (section 5.3).

(ii) Small but frequent phase shifts have been found to be advantageous, yielding an arbitrarily deep cooling bound and the least sensitivity to the coupling spectrum of the bath (sections 5.1 and 5.2).

(iii) The ability (section 6) to achieve very fast (ps scale) cooling without resorting to auxiliary states or equilibration with the bath is a principal innovation. It may pave the way to better performance of coherent control of logic operations.
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Appendix A. Impulsive QND disturbances

Measurement

Following a projective measurement:

$$\langle H_{SB} \rangle^M = \frac{1}{2} \langle H_{SB} \rangle_{\text{Eq}} + \frac{1}{2} \sum_k \eta_k \text{Tr}[ (b_k + b_k^\dagger) \sigma_\epsilon \sigma_\sigma \rho_{\text{Eq}} ]$$

$$= \frac{1}{2} \langle H_{SB} \rangle_{\text{Eq}} - \frac{1}{2} \langle H_{SB} \rangle_{\text{Eq}} = 0,$$

where we have used the identity $\sigma_\epsilon \sigma_\sigma \sigma_\epsilon = -\sigma_s$.

Any non-selective measurement must increase the entropy of $\rho_{\text{tot}}$. Since prior to the measurement, $\rho_{\text{tot}}$ was in a Gibbs state, which is the maximal entropy state among all the states with the same mean energy $\langle H_{\text{tot}} \rangle$, the entropy increase implies an increase of $\langle H_{\text{tot}} \rangle$, i.e. heating.

A QND measurement changes $\rho_{\text{tot}}$, yet leaves $\rho_S$ and $\rho_B$ unchanged; hence it changes neither $\langle H_S \rangle$ nor $\langle H_B \rangle$. Therefore, the increase in $\langle H_{\text{tot}} \rangle$ must reflect an equal increase in $\langle H_{SB} \rangle$. Since the post-disturbance $\rho_{\text{tot}}^M = \text{Tr}_B \rho_{\text{tot}}^M$ is diagonal in the $H_S$ or $\sigma_\sigma$ eigenbasis, we have $\langle H_{SB} \rangle^M = 0$ immediately after the measurement. Thus, $\langle H_{SB} \rangle$ must have been negative at equilibrium, before the disturbance. This indeed follows from perturbation theory (Supplementary I). Hence, the projective measurement pumps into the system + bath an energy of

$$\delta \langle H_{\text{tot}} \rangle^M = -\langle H_{SB} \rangle_{\text{Eq}}.$$

This energy can be expressed as (Supplementary I) the bath-induced Lamb-shift, modified by population inversion factors, which are negative at positive temperatures. Owing to the non-unitary nature of the projection, the mixedness of the total state increases, whereas it remains unchanged under rotation:

$$\text{Tr}[ (\rho^\phi)^2 ] = \text{Tr}[ (\rho_{\text{Eq}})^2 ],$$

$$\text{Tr}[ (\rho^M)^2 ] = \frac{1}{2} \text{Tr}[ (\rho_{\text{Eq}})^2 ] + \frac{1}{2} \text{Tr}[ (\sigma_\epsilon \rho_{\text{Eq}} \sigma_\sigma \rho_{\text{Eq}}) ].$$

In the weak-coupling limit, the increase in mixedness due to measurement is $\approx O(\epsilon^4)$ and hence can be neglected.

Rotation (phase shift)

The total state following a rotation of the qubit by a phase $\phi$ about the $z$-axis (figure 1(a)) is given by

$$\rho_{\text{Eq}} \rightarrow \rho_{\text{tot}}^\phi = |e\rangle \langle e| \hat{B}_{ee} + |g\rangle \langle g| \hat{B}_{gg} + e^{-i\phi} |e\rangle \langle g| \hat{B}_{eg} + \text{h.c.}$$

For this state, $\langle H_S \rangle$ and $\langle H_B \rangle$ are the same before and after the rotation, while the mean energy changes.
Following a $\sigma_z$ rotation by angle $\phi$:

$$\langle H_{SB}\rangle_{\text{Eq}}^{\phi} \equiv \sum_k \eta_k \text{Tr}[\sigma_x (b_k \sigma_x + b_k^\dagger) e^{-i\phi \sigma_z}]$$

$$= \sum_k \eta_k \text{Tr}[(b_k \sigma_x + b_k^\dagger) e^{-i\phi \sigma_z} \rho_{\text{Eq}}]$$

$$= \cos \phi \langle H_{SB}\rangle_{\text{Eq}} + \sin \phi \sum_k \eta_k \text{Tr}[(b_k \sigma_x + b_k^\dagger) \rho_{\text{Eq}}]. \quad (A.6)$$

By rewriting the total Hamiltonian as

$$H_{\text{tot}} = \hat{A} \sigma_z + \hat{B}_1 + \hat{B}_2 \sigma_x, \quad (A.7)$$

where $[\hat{A}, \hat{B}_i] = 0$ and $[\hat{B}_1, \hat{B}_2] \neq 0$, one can show that to $O(\epsilon^2)$

$$\sum_k \eta_k \text{Tr}[(b_k \sigma_x + b_k^\dagger) \rho_{\text{Eq}}] = 0. \quad (A.8)$$

Hence, the mean interaction energy after a $\sigma_z$ rotation is given by

$$\langle H_{SB}\rangle_{\text{Eq}}^{\phi} \approx \cos \phi \langle H_{SB}\rangle_{\text{Eq}}, \quad (A.9)$$

and the change, which gives an estimate of the energy injected into the system due to the rotation, is given by

$$\delta \langle H \rangle_{\text{tot}}^{\phi} = -(1 - \cos \phi) \langle H_{SB}\rangle_{\text{Eq}}. \quad (A.10)$$

The energy pumped into the total system is maximal when the rotation angle is $\phi = \pi$, which corresponds to the phase-flip operation. Since (A.5) is no longer a Gibbs state, which is the minimal-energy state for that entropy, it must correspond to higher mean energy; hence

$$\delta \langle H_{\text{tot}} \rangle^{\phi} > 0, \quad (A.11)$$

thus completing the general proof of post-disturbance heating.

**Appendix B. Cooling conditions**

The excited population of the qubit after time $t$ is given by

$$\rho_{ee}(t) = e^{-J(t)} \rho_{ee}(0) + e^{-J(t)} \int_0^t dt' e^{J(t')} R_x(t'), \quad (B.1)$$

where both $R_x(t)$ and $J(t)$ depend on the specific disturbances and the interval $\tau$ between them. In the limit of $t \to \infty$, the first term in (B.1) vanishes. This gives the universal cooling bound

$$\rho_{ee}(t) \lim_{t \to \infty} \rho_{ee}(t') \equiv \rho_{ee}(\infty)(\tau). \quad (B.2)$$

In the limit $t \to \infty$, further simplification to the cooling condition, $\chi < \rho_{ee}(0)$, gives

$$\frac{J_e(t)}{J_g(t)} \lim_{t \to \infty} \frac{R_e}{R_g} \equiv \frac{n_T(\omega_a) + 1}{n_T(\omega_a)} = \frac{R_e}{R_g} \quad (B.3)$$
Impulsive measurements

The relaxation integrals, \( J_e(t = n\tau) = \int_0^\infty dt' R_e(t')(t') \), are explicitly given by

\[
J_e(t = n\tau) = n\tau^2 \int_0^\infty d\omega G_0(\omega) n_T(\omega) \text{sinc}^2((\omega - \omega_a)\tau/2) \\
+ n\tau^2 \int_0^\infty d\omega G_0(\omega) (n_T(\omega) + 1) \text{sinc}^2((\omega + \omega_a)\tau/2),
\]

(B.4)

\[
J_e(t = n\tau) = n\tau^2 \int_0^\infty d\omega G_0(\omega) n_T(\omega) \text{sinc}^2((\omega + \omega_a)\tau/2) \\
+ n\tau^2 \int_0^\infty d\omega G_0(\omega) (n_T(\omega) + 1) \text{sinc}^2((\omega - \omega_a)\tau/2).
\]

(B.5)

Rearranging the terms in equations (B.4) and (B.3) yields the cooling condition

\[
\int_0^\infty G_0(\omega) \text{sinc}^2\left(\frac{\omega - \omega_a}{2}\tau\right) (n_T(\omega_a) - n_T(\omega)) > \int_0^\infty G_0(\omega) \text{sinc}^2\left(\frac{\omega + \omega_a}{2}\tau\right) (n_T(\omega_a) + n_T(\omega) + 1).
\]

(B.6)

Here the LHS stems from RW terms of the interaction Hamiltonian \( H_{SB} \), whereas the RHS stems from CR terms therein. This inequality underscores the need to account for both RW and CR induced effects when studying cooling.

A quest for the spectral density function \( G_0(\omega) \), which satisfies condition (B.6) in a given time interval, may be undertaken. In the high-temperature limit, \( n_T(\omega) = 1/\beta\omega \gg 1 \), a necessary condition to satisfy the above inequality is that \( G_0(\omega) \) be concentrated in the frequency interval

\[
\omega_a < \omega < \Omega,
\]

(B.7)

where

\[
\Omega = \frac{1}{\beta} \left[ 1 + \frac{\beta\omega_a + \sqrt{4 + 12\beta\omega_a + \beta^2\omega_a^2}}{2} \right].
\]

(B.8)

This imposes a bound on the detuning. We should not detune the bath spectrum too far above \( \omega_a \) if we wish to get cooling. We have numerically verified these conditions for various bath coupling spectra. In the same vein, one can find frequency regions where in specific time intervals there is no cooling, regardless of the shape of \( G_0(\omega) \).

Impulsive phase shifts

Upon using the appropriate expressions for the relaxation integrals \( J_e(t) = \int dt' R_e(t')(t') \), we obtain the following cooling condition:

\[
\frac{\int_0^\infty d\omega G_0(\omega) F_1(\omega_a - \omega) [n_T(\omega_a) - n_T(\omega)]}{\int_0^\infty d\omega G_0(\omega) F_1(\omega_a + \omega) [n_T(\omega_a) + n_T(\omega) + 1]} > 1.
\]

(B.9)
For $\phi \ll 1$ this condition reduces to

$$\phi > 0.$$  \hspace{1cm} (B.10)

Remarkably, this cooling condition is completely insensitive to the coupling spectrum $G_0(\omega)$, in sharp contrast to measurements and $\pi$-phase flips.

**Continuous modulations**

One can maximize the purification by applying time-dependent modulation on the system levels either continuously or by impulsive phase shifts. By applying time-dependent unitary ac-Stark shift on the excited level, $\delta_a(t)$, one obtains from the Kofman–Kurizki formula

$$J_{e(g)}(t) = \int_{-\infty}^{\infty} d\omega G_T(\omega) F_t(\omega_a + \omega),$$  \hspace{1cm} (B.11)

$$F_t(\omega) = |\epsilon_t(\omega)|^2,$$  \hspace{1cm} (B.12)

$$\epsilon_t(\omega) = (2\pi)^{-1/2} \int_0^t dt' e^{i\omega t'} + \int_0^{t'} dt \delta_a(t).$$  \hspace{1cm} (B.13)

The modulation can optimize the cooling described in the main text.
Pulse shaping analysis

Our treatment of the cooling dynamics and universal cooling bounds is general: they hold for any form of phase shifts and pulse shaping, expressed explicitly by $F_t(\omega)$ (see equations (8)–(12) in the main text). We have numerically verified that variations of the pulse shaping of phase flips of duration $t_{\text{flip}}$, conforming to the requirement that $\omega_0 t_{\text{flip}} \ll 1$, do not have a strong effect on the cooling dynamics.

In figure B.1 we can see the cooling of a system by $\pi$-pulse modulation for different pulse durations. The system is the same as in figure 4 (main panel) in the main text, but for clarity only the minimum point of each oscillation was plotted. One can clearly see that changes in the pulse duration only weakly affect the cooling.

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