Controlling a Mesoscopic Spin Environment by Quantum Bit Manipulation

J. M. Taylor,[1] A. Imamoglu,[2] and M. D. Lukin[1]
[1]*Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA*
[2]*Institute of Quantum Electronics, ETH-Hönggerberg, HPT G 12, Zürich, Switzerland*
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We present a unified description of cooling and manipulation of a mesoscopic bath of nuclear spins via coupling to a single quantum system of electronic spin (quantum bit). We show that a bath cooled by the quantum bit rapidly saturates. Although the resulting saturated states of the spin bath ("dark states") generally have low degrees of polarization and purity, their symmetry properties make them a valuable resource for the coherent manipulation of quantum bits. Specifically, we demonstrate that the dark states of nuclear ensembles can be used to coherently control the system-bath interaction and to provide a robust, long-lived quantum memory for qubit states.

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An intriguing challenge for modern science and technology is the coherent manipulation of quantum systems coupled to realistic environments. Interest in these problems is in part due to fundamental aspects of quantum control and decoherence, but this research has also been stimulated by recent developments in quantum information science [1]. Although over the past decades much progress has been made in the controlled manipulation of isolated atomic and optical systems [2], the complex environment of a solid-state system makes it significantly more challenging to achieve a similar degree of control.

This Letter demonstrates that a single quantum system (qubit) can be used to prepare and control a mesoscopic environment, turning the bath into a useful resource. We consider a system consisting of a single electronic spin in a semiconductor quantum dot interacting with a mesoscopic bath of nuclear spins within a confined volume. Recently it has been shown that cooling the spin bath to high values of polarization and purity greatly reduces the associated decoherence [3]. Furthermore, due to the bath's intrinsic memory, it can be used as a long-lived quantum memory for qubits and for quantum state engineering of collective nuclear states [4]. However, achieving a high degree of nuclear polarization in a quantum dot remains a major experimental challenge. Most ideas under exploration use the hyperfine contact interaction between electron spins and the bath. Some work *in situ,* using either spin-polarized currents [5] or optical pumping [6,7]. Other techniques use a different geometry for cooling, such as quantum Hall edge state tunneling near a quantum point contact [8].

We focus on *in situ* manipulation, when the qubit degrees of freedom are themselves used to cool nuclei. We show that such a qubit-based cooling process rapidly saturates resulting in nonthermal states of the nuclear bath with low polarization and purity. However, the symmetry properties of such dark states allow for complete control of the qubit's interaction with the environment. We illustrate this by showing that the mesoscopic bath prepared in saturated states can be used to provide a long-lived quantum memory for qubit states. A combination of adiabatic passage techniques and spin echo results in near unity storage fidelity even for a bath with vanishingly small polarization. Before proceeding we also note that the idea of using qubit states to cool the environment is now widely applied in atomic systems such as trapped ions [9] or microwave cavity QED [10].

A simple Hamiltonian can describe a single electron spin confined in a quantum dot interacting with an applied external magnetic field B_0 and with N spin I_0 surrounding nuclei via the hyperfine contact interaction:

$$\hat{H} = g^* \mu_B B_0 \hat{S}_z + g_n \mu_n B_0 \sum_j \hat{I}_z^j + a \sum_j \alpha_j \hat{\vec{S}} \cdot \hat{\vec{I}}^j, \quad (1)$$

where $\hat{I}^j_{z,\pm}$ and $\hat{S}_{z,\pm}$ are spin operators for nuclei and electrons, respectively. The hyperfine interaction is split into a field aligned (Overhauser) component $\hat{V}_{zz} = a\hat{A}_z\hat{S}_z$ and a Jaynes-Cummings–type component, $\hat{H}_{\mathrm{JC}} = a/2(\hat{A}_+\hat{S}_- + \hat{A}_-\hat{S}_+)$. We use collective operators $\hat{A}_{z,\pm} = \sum_j \alpha_j \hat{I}^j_{z,\pm}$, $\alpha_j = N v_0 |\psi(r_j)|^2$ is the weight of the electron wave function at the jth lattice site, and a is the average hyperfine interaction constant per nucleus.

Nuclear degrees of freedom are cooled by cycling spin polarization through the quantum dot. A spin-down electron is injected from leads connected to a polarized reservoir or by means of optical excitation. It interacts for some short time τ and then is ejected or recombined. Each iteration can cool the bath by flipping a nuclear spin through $\hat{H}_{\mathrm{JC}}$. If the energy difference of the injected electron spin and the flipped electron spin, $\langle\Delta\rangle = (g^*\mu_B - g_n\mu_n)B_0 + a\langle\hat{A}_z - 1\rangle$, is large compared to the inverse time of interaction, τ^{-1}, energy conservation considerations block the spin-flip process. However, changing the applied field to maintain $\langle\Delta\rangle\tau \ll 1$ allows cooling to continue efficiently [7].

Regardless of the exact details of the process, cooling *will* saturate. The system is driven into a statistical

mixture of dark states $|\mathcal{D}\rangle$, defined by [7]

$$\hat{A}_-|\mathcal{D}\rangle = 0. \tag{2}$$

To cool past this point of saturation, either dark states must couple to other states of the bath or the geometric coupling coefficients α_k must change [7]. When high polarization is achieved, previous results hold [3,4]. When these mechanisms are slow compared to the cooling rate, an appropriate mixture of dark states well approximates the steady state of the bath.

The homogeneous case illustrates the essential features of cooling. With $\alpha_k = 1$, we rewrite $\hat{A}_{z,\pm}$ as collective nuclear angular momentum operators $\hat{J}_{z,\pm}$; correspondingly, $\hat{J}^2$ becomes a conserved quantity. The Dicke basis, characterized by total (nuclear) angular momentum J $(0, 1/2 \le J \le N/2)$, its projection into the z axis m_J, and a permutation group quantum number β, is then appropriate [11]. $\hat{J}_-$ changes neither J nor β, but nuclei in a state $|J, m_J, \beta\rangle$ are cooled to the state with lowest m_J (dark state) $|J, -J, \beta\rangle$. For an initial thermal bath of nuclei with polarization P_0, the corresponding steady-state solution is found by summing over $-J \le m_J \le J$. Tracing over β, we find

$$\begin{aligned}\hat{\rho}_{ss} &= \sum_J \rho_n(J)|J, -J\rangle\langle J, -J| \\ &= \left(\frac{1-P_0^2}{4}\right)^{N/2} \frac{1}{2P_0} \sum_J D(J) \\ &\quad \times \left[\frac{(1+P_0)^{J+1}}{(1-P_0)^J} - \frac{(1-P_0)^{J+1}}{(1+P_0)^J}\right]|J, -J\rangle\langle J, -J|.\end{aligned} \tag{3}$$

$D(J)$ denotes the number of β quantum numbers allowed for a given J and is independent of m_J. In the case of spin-1/2 nuclei, $D(J) = \binom{N}{N/2-J} - \binom{N}{N/2-J-1}$. The resulting nuclear polarization P and von Neumann entropy associated with the "cooled" ensemble are shown in Fig. 1 as a function of initial thermal polarization P_0. The differences between the thermal and saturated baths become negligible for large N, but the dynamics of the two baths differ dramatically. In essence, even though the purity and polarization are low, the symmetry properties of dark states restrict the evolution of the combined electronic-nuclear system to two levels.

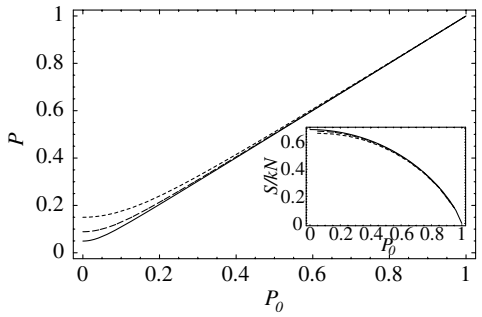

FIG. 1. Saturated state polarization P versus initial thermal polarization P_0 for $N = 100$ (dotted line), 300 (dashed line), and 10^3 (solid line). The inset shows entropy per spin.

We illustrate the reversible nature of the coupling between dark states and the single spin by showing how a quantum state can be stored into collective nuclear states. An arbitrary qubit state $|\Psi\rangle = u|\uparrow\rangle + v|\downarrow\rangle$ will be mapped into the bath states. With just a pure state $|J, -J\rangle$, spin-down is decoupled entirely, while spin-up couples nuclei to the collective state $|J, -J+1\rangle$ with a Rabi frequency $\Omega_J = a\sqrt{2|J|}$. As $\hat{H}_{\rm JC}^2|\uparrow\rangle|J, -J\rangle = \frac{a^2}{2}J|\uparrow\rangle|J, -J\rangle$, the motion is given by the cyclic dynamics of a two-level system. Near resonance ($|\langle\Delta^2\rangle| \lesssim |\Omega_J|^2$), the qubit will oscillate fully between electronic and nuclear states. For high polarization P this is in direct analogy to the case discussed in Ref. [4]. However, for low P, all states of the mixture must be in resonance; the range of J with significant probability goes as the width of the binomial distribution, $\sqrt{N}$, while the width of the resonance, given by $\langle\Omega_J\rangle/a$, goes as $\sqrt{PN}$. In this regime, the resonance is much narrower than the range of populated J states.

This problem can be solved with adiabatic passage [12]. By sweeping the detuning from far negative to far positive, the system passes through a series of avoided crossings and for each J

$$(u|\uparrow\rangle + v|\downarrow\rangle)|J, -J\rangle \rightarrow |\downarrow\rangle(ue^{i\phi_J}|J, -J+1\rangle + v|J, -J\rangle.$$

Adiabatic passage is not sensitive to the exact value of the coupling between individual pairs of levels and is robust provided the sweep rate of the detuning is sufficiently slow.

In general, the relative phase ϕ_J accumulated depends on the details of the detuning sweep. Sweeping the detuning back reverses storage, but the final state, $(ue^{2i\phi_J}|\uparrow\rangle + v|\downarrow\rangle)|J, -J\rangle$, has an additional nontrivial phase which reduces the final off-diagonal matrix element of electronic spin density matrix: $\rho_{\uparrow\downarrow} = uv^*[\sum_J \rho_n(J)e^{2i\phi_J}]$. Spin echo avoids this strong dephasing by exactly compensating the adiabatically acquired phase [13]. An example sequence is presented in Table I. The two waiting segments should be symmetric, to compensate for other

TABLE I. Adiabatic transfer with ESR spin echo.

State	Process	$\Delta(t)$
$(u\|\uparrow\rangle + v\|\downarrow\rangle)\|J, -J\rangle$	Start	Δ_i
$\|\downarrow\rangle(ue^{i\phi_J}\|J, -J+1\rangle + v\|J, -J\rangle)$	Store	$\rightarrow \Delta_f$
	Wait	Δ_f
$(ue^{2i\phi_J}\|\uparrow\rangle + v\|\downarrow\rangle)\|J, -J\rangle$	Retrieve	$\Delta_i \leftarrow$
$(iv\|\uparrow\rangle + ue^{2i\phi_J}\|\downarrow\rangle)\|J, -J\rangle$	π − pulse	Δ_i
$\|\downarrow\rangle(ive^{i\phi_J}\|J, -J+1\rangle + ue^{2i\phi_J}\|J, -J\rangle)$	Store	$\rightarrow \Delta_f$
	Wait	Δ_f
$(ive^{2i\phi_J}\|\uparrow\rangle + ue^{2i\phi_J}\|\downarrow\rangle)\|J, -J\rangle$	Retrieve	$\Delta_i \leftarrow$
$e^{2i\phi_J}(u\|\uparrow\rangle - v\|\downarrow\rangle)\|J, -J\rangle$	π − pulse	Δ_i

arbitrary J-dependent phases [14]. Thus a mixture of dark states can be used as an ideal quantum memory.

In practice, the electron-spin decoherence rate γ limits the minimum speed of the adiabatic sweep, the induced error scaling as $p_\gamma \simeq \gamma T$ with T as the characteristic duration of the storage procedure. For a saturated ensemble state, $T \simeq 4\langle\Omega_J\rangle/\xi$, where $\xi = \langle\dot{\Delta}\rangle$ is the rate of change of the detuning. Assuming a tangent-like pulse shape [15] we find the nonadiabatic flip probability

$$p_{\mathrm{na}} \simeq \xi^2\langle\Omega_J\rangle^4/32 = T^2\langle\Omega_J\rangle^2/2. \tag{4}$$

The total error probability is then $p_{\mathrm{tot}} = p_\gamma + p_{\mathrm{na}}$. Minimizing this for T gives $T_{\mathrm{min}} = (\gamma\langle\Omega_J\rangle^2)^{-1/3}$ and

$$p_{\mathrm{tot,min}} = 3/2(\gamma/\langle\Omega_J\rangle)^{2/3}. \tag{5}$$

We defer numerical analysis of this error to the end to allow inclusion of inhomogeneous corrections.

The saturated state lifetime, the storage lifetime, and the maximum polarization are limited by nuclear spin dephasing. Spin diffusion due to dipolar nuclear coupling is the dominant term for this dephasing in GaAs and is on the order of 6×10^4 s^{-1} [16]. This rate also provides an estimate for heating from proximal thermal spins. The competition between heating and cooling will likely determine the steady-state polarization of the dark-state mixture. Active correction pulse sequences such as WHH-4 (Waugh, Huber, and Haeberlen) can lead to sub-Hz decoherence rates and lowered spin diffusion [17], suggesting storage lifetimes on the order of seconds may be feasible. Finally, we note that these results generalize to higher spin by using the appropriate multinomial form of $D(J)$ [18].

We now extend these results to realistic inhomogeneous coupling between electrons and nuclei by developing a one-to-one mapping between the explicit homogeneous Dicke basis and its inhomogeneous equivalent. A Dicke state $|J, -J, \beta\rangle$ in the individual spin basis is

$$|J, -J = n - N/2, \beta\rangle = \sum_{\{j\}_n} c_{J,\beta}(\{j\})|\{j\}\rangle, \tag{6}$$

where the set $\{j\}_n$ labels n spins that are pointing up; the rest point down. As $\hat{J}_-|J, -J, \beta\rangle = 0$,

$$\sum_{l\notin\{i\}_{n-1}} c_{N/2-n,\beta}(\{i\} + l) = 0 \tag{7}$$

for all $\{i\}_{n-1}$. Furthermore, $\hat{J}_-$ is invariant under permutation so there exists a representation for dark states where every individual spin configuration is equally probable, i.e., $|c_{J,\beta}(\{i\})|^2 = |c_{J,\beta}(\{j\})|^2 = \binom{N}{N/2-J}^{-1}$. Using this explicit representation for homogeneous dark states, we construct a mapping to the more general inhomogeneous case $[\hat{A}_-|\mathcal{D}(n, \beta)\rangle = 0]$. For each dark state $|J, -J = n - N/2, \beta\rangle$, its inhomogeneous counterpart is

$$|\mathcal{D}(n, \beta)\rangle = \mathcal{N}_{0,0}^{-1/2}\sum_{\{j\}_n}\left(\prod_{k\in\{j\}}\frac{1}{\alpha_k}\right)c_{N/2-n,\beta}(\{j\})|\{j\}\rangle, \tag{8}$$

as can be checked by direct calculation. The exact form of the normalization constant $\mathcal{N}_{0,0}$ is defined below.

To quantify inhomogeneous effects, first we note that $\hat{A}_-\hat{A}_+$ maps $|\mathcal{D}(n, \beta)\rangle$ into an orthogonal state $|\mathcal{O}(n, \beta)\rangle$, $\hat{A}_-\hat{A}_+|\mathcal{D}(n, \beta)\rangle = |\Omega_n|^2|\mathcal{D}(n, \beta)\rangle + |\chi_n|^2|\mathcal{O}(n, \beta)\rangle$ with

$$\Omega_n = a\sqrt{\langle\mathcal{D}(n, \beta)|\hat{A}_-\hat{A}_+|\mathcal{D}(n, \beta)\rangle},$$

$$\chi_n = a[\langle\mathcal{D}(n, \beta)|\hat{A}_-\hat{A}_+\hat{A}_-\hat{A}_+|\mathcal{D}(n, \beta)\rangle - |\Omega_n|^4]^{1/4}.$$

Nonzero χ_n indicates that an inhomogeneous equivalent of $\hat{J}^2$ is not conserved under $\hat{A}_\pm$. Second, inhomogeneous dark states are also not eigenstates of $\hat{A}_z$, i.e., $\hat{V}_{zz}|\mathcal{D}(n, \beta)\rangle = \delta_n|\mathcal{D}(n, \beta)\rangle + \omega_n|\mathcal{B}(n, \beta)\rangle$, where

$$\omega_n = \sqrt{\langle\mathcal{D}(n, \beta)|\hat{V}_{zz}^2|\mathcal{D}(n, \beta)\rangle - \langle\mathcal{D}(n, \beta)|\hat{V}_{zz}|\mathcal{D}(n, \beta)\rangle^2}.$$

If the symmetry breaking terms (χ_n, ω_n) are small relative to Ω_n, cooling will proceed similarly to the homogeneous case. The final state density matrix should then be of the type $\hat{\rho} = \sum_{n,\beta}\rho(n)|\mathcal{D}(n, \beta)\rangle\langle\mathcal{D}(n, \beta)|$. When χ_n and ω_n are small we can use Eq. (3) as an estimate for $\rho(n)$. After reaching saturation, cooling slows to a rate governed by the transfer of dark states into other states.

Following the above prescription for adiabatic transfer, symmetry breaking terms lead to additional errors. When $\chi_n \ll \Omega_n$, the rate of transfer is given by Ω_n, but the final state is an admixture of $|\mathcal{D}(n, \beta)\rangle$ and $|\mathcal{O}(n, \beta)\rangle$, leading to an error of order χ_n^2/Ω_n^2. The error from ω_n we estimate in the worst case by considering it as an incoherent loss mechanism. The effective decoherence rate becomes $\gamma_{\mathrm{eff},n} = \sqrt{\omega_n^2 + \gamma^2}$, the spin-decoherence rate optimization used for the homogeneous case holds, and the resulting probability of error for the full sequence goes as $3/2(\gamma_{\mathrm{eff},n}/\Omega_n)^{2/3}$. Combining these, the total probability of error goes as

$$p_{\mathrm{tot}} = 1 - \sum_n\left\{\left(\frac{\Omega_n^2}{\Omega_n^2 + \chi_n^2}\right)^4\left[1 - 3\left(\frac{\gamma_{\mathrm{eff},n}}{\Omega_n}\right)^{2/3}\right]\rho(n)\right\}.$$

Considering these errors numerically, the explicit form of the inhomogeneous dark states (8) allows us to express $\Omega_n, \chi_n, \omega_n$ as functions of the geometric coupling constants, α_k:

$$\Omega_n^2/a^2 = \sum_k \alpha_k^2 - 2\mathcal{N}_{2,1}(n)/\mathcal{N}_{0,0}(n), \tag{9}$$

$$\chi_n^4/a^4 = \frac{4\mathcal{N}_{2,2}(n)}{\mathcal{N}_{0,0}(n)} - \left(\frac{2\mathcal{N}_{2,1}(n)}{\mathcal{N}_{0,0}(n)}\right)^2, \tag{10}$$

$$\omega_n^2/a^2 = \frac{\mathcal{N}_{1,2}(n)}{\mathcal{N}_{0,0}(n)} - \left(\frac{\mathcal{N}_{1,1}(n)}{\mathcal{N}_{0,0}(n)}\right)^2, \tag{11}$$

with

$$\mathcal{N}_{\mu,\nu}(n) = \binom{N}{n}^{-1}\sum_{\{j\}_n}\left(\prod_{k\in\{j\}_n} 1/\alpha_k^2\right)\left[\sum_{k\in\{j\}_n}\alpha_k^\mu\right]^\nu.$$

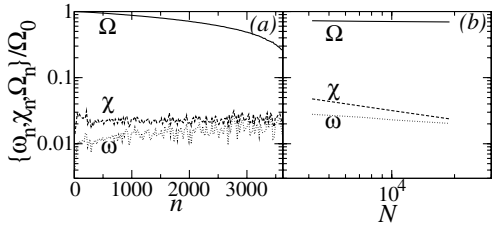

FIG. 2. (a) Ω_n (solid line), ω_n (dotted line), and χ_n (dashed line) versus number of flipped spins n with $N = 7280$. Values are averaged over every 20 n values and scaled by $\Omega_0 = A/\sqrt{N}$. (b) Average values of Ω_n/Ω_0, ω_n/Ω_n, and χ_n/Ω_n vs N.

To estimate the required $N_{\mu,\nu}$ we average over a statistically significant fraction of the allowed $\{j\}_n$ for each n sublevel. The α_k's are drawn from an oblate Gaussian electron wave function of ratio $(1, 1, 1/3)$, and we omit spins with $\alpha_k < 1/N$. We plot the three parameters Ω_n, χ_n, ω_n versus n in Fig. 2. The perturbative treatment used above is justified as $\Omega_n \gg \omega_n, \chi_n$ for all n.

In Fig. 3 we plot total probability of error for the saturated mixture as a function of the final saturated polarization P. We used the hyperfine constant for GaAs, $aN \simeq 2 \times 10^{10}\ \mathrm{s}^{-1}$ and $\gamma \sim 6 \times 10^{6}\ \mathrm{s}^{-1}$. Adiabatic transfer requires a small change of effective field ($\simeq$100 mT) over 10–100 ns, which could be implemented through g-factor engineering [19] or spin-dependent optical Stark shifts [7]. For 10^4 nuclei, transfer times of 100 ns and fidelities better than 0.8 are possible with realistic spin-decoherence rates even for vanishingly small polarizations. The error decreases further with increasing N.

In conclusion, we have demonstrated that electron-spin qubits can be used to effectively prepare and manipulate a local nuclear spin environment. Specifically, long coherence times and high fidelities for the storage of electron-spin states into nuclear spins can be achieved provided the same qubit is used for the cooling process. Such "coherent" cooling and storage is effective for nuclear spin preparation due to their long coherence times. Related techniques can be used for engineering quantum states of nuclear spins from a saturated bath state [4,20]. We further note that the techniques described in the present Letter may be applicable to other systems involving mesoscopic spin baths. For example, we anticipate that similar methods may be used to prepare the local environment of superconducting qubits.

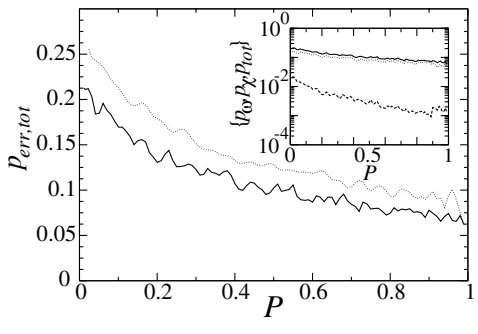

FIG. 3. Expected error of transfer and recovery for the inhomogeneous case versus final polarization P for $N = 4145$ (dotted line) and 18 924 (solid line). The inset shows the error due to ω_n (dotted line) and χ_n (dashed line), and total error (solid line) for $N = 18\,924$.

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[12] See, e.g., L. D. Landau and E. M. Lifshitz, *Quantum Mechanics: Nonrelativistic Theory* (Pergamon, New York, 1977), 3rd ed., pp. 342–351.
[13] The necessity of spin echo can also be understood by considering the spin-wave modes, $\hat{\boldsymbol{J}}^k_+ = \sum_j e^{i\vec{k}\cdot\vec{r}_j}\hat{\boldsymbol{I}}^j_+$. The cooling process cools only the completely symmetric mode ($\hat{\boldsymbol{J}}^{(k=0)}_+$, or in the inhomogeneous case, $\hat{\boldsymbol{A}}_+$), while $\hat{V}_{zz}$ couples the spin to *all* modes, dephasing the electron-spin state. Spin echo exactly cancels the $\hat{V}_{zz}$ evolution. We note that in the far-off resonant (large effective field) regime, $\hat{V}_{zz}$ is the only relevant system-bath coupling, and thus dephasing is reversible.
[14] Even without adiabatic transfer and at low field, a spin-echo pulse train applied faster than $\langle\Omega_J\rangle \sim 10^8\ \mathrm{s}^{-1}$ will entirely cancel phase and spin-exchange evolution, thereby decoupling electron-spin from nuclei and completely suppressing nuclear spin-related dephasing.
[15] The pulse is of the form $\Delta(t) = (\Delta_i + \Delta_f)/2 + (\Delta_i - \Delta_f)/\epsilon_1 \tan[\xi\pi(t - T/2 + \epsilon_2)/2 + \epsilon_3]$ with parameters $\epsilon_{1,2,3}$ set to give proper starting and ending detunings.
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