Protected state enhanced quantum metrology with interacting two-level ensembles

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Ramsey interferometry is routinely used in quantum metrology for the most sensitive measurements of optical clock frequencies. Spontaneous decay to the electromagnetic vacuum ultimately limits the interrogation time and thus sets a lower bound to the optimal frequency sensitivity. In dense ensembles of two-level systems the presence of collective effects such as superradiance and dipole-dipole interaction tends to decrease the sensitivity even further. We show that by a redesign of the Ramsey-pulse sequence to include different rotations of individual spins that effectively fold the collective state onto a state close to the center of the Bloch sphere, partial protection from collective decoherence and dephasing is possible. This allows a significant improvement in the sensitivity limit of a clock transition detection scheme over the conventional Ramsey method for interacting systems and even for non-interacting decaying atoms.

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Introduction The precise measurement of time using suitable atomic transitions is a major achievement of quantum metrology. The Ramsey interferometry procedure plays a crucial role in this context as it allows a quite accurate locking of the microwave or optical oscillator to the transition frequency in the atom. Typical early realizations were based on an atomic beam or a laser-cooled atomic fountain later on [1], where the atoms would interact with two consecutive Rabi pulses. With optical lattices (details see [2]) and proposals for optical lattice clocks, e.g. [3] time measurements were expected to become even more accurate due to longer interaction times and the elimination of collisions (see [4, 5] for recent reviews). To reduce quantum projection noise (scaling as $1/\sqrt{N}$, where $N$ is the number of atoms) and to speed up the measurement, experimental setups usually involve an as large as possible number of atoms. In a finite volume, of course, this brings collective effects like superradiance and dipole-dipole shifts to the table [6]. While some techniques rely on the engineering of particular geometries without the need to alter the internal atomic states [7], exploiting the uncertainty principle by employing squeezed states [8, 10] can be helpful as well to achieve less noise with lower atom numbers [11, 12]. These techniques heavily rely on entanglement [13, 14] among atoms and require very careful preparation and isolation of the ensemble.

When, finally, interrogation times reached the lifetime of the excited state, spontaneous emission became a critical factor for the contrast of the Ramsey fringes. Interestingly, despite the use of long lived clock states, for multiple atoms in close proximity to each other, collective spontaneous emission can still reach a detrimental magnitude. Here, the common atomic coupling to the same electromagnetic vacuum fluctuations enhances spontaneous emission by a factor proportional to the atom number [15, 16]. While this is usually limited to volumes of the order of a cubic wavelength, in regular arrays, such as an optical lattice, the effect can extend over tens of lattice sites [17]. In addition, excitonic energy level shifts in lattices can also induce significant dephasing of the Ramsey signal, which cannot be removed by simple echo techniques.

In this paper, we propose a strategy that works on the level of the Ramsey pulses and which we dub the ‘asymmetric Ramsey technique’, in contrast to the typical symmetric Ramsey technique that employs only identical $\pi/2$ pulses applied to all atoms. While the conventional Ramsey technique excites superposition states, which possess a maximum dipole moment and thus are most sensitive to superradiance, this new approach allows the selection of long-lived collective states (or ‘dark states’) to improve the sensitivity of the clock signal. The procedure requires two further manipulations of the atomic dipoles in addition to the usual sequence: after the initial $\pi/2$ pulse is applied, each atomic coherence is rotated by a distinct phase, resulting in a subradiant collective state (with a lifetime which can be even longer than that of the independent atoms). This results in a state of vanishing classical collective dipole which is typically well-protected from the environmentally-induced decoherence [18]. As a clarifying example, in the Dicke limit [19] of atoms positioned at the same spot, a state that exhibits infinite lifetime exists and is therefore perfectly suitable for state-protective spectroscopy.

Model for $N$ atoms We assume a collection of $N$ identical two-level emitters with levels $\ket{g}$ and $\ket{e}$ separated by $\omega_0$ in a general geometry defined by the positions $\mathbf{r}_i$ for $i = 1, ..., N$ and the angle $\theta$ drawn between the (identical) transition dipoles and their separations. We define individual Pauli ladder operators $\sigma_i^\pm$ and subsequently $\sigma_\phi^x = \sigma_i^+ + \sigma_i^-$, $\sigma_\phi^y = -i(\sigma_i^+ - \sigma_i^-)$ and $\sigma_\phi^z = \sigma_i^+ \sigma_i^- - \sigma_i^- \sigma_i^+$ as well as the corresponding unitary rotations $R_\phi^{(j)}[\varphi] = \exp(\varphi \sigma_\phi^j/2)$ where $\mu \in \{x, y, z\}$.
The independent coupling of each atom to the electromagnetic vacuum leads to a decay rate $\Gamma$; the cooperative nature of decay for atom pairs $i,j$ is reflected by mutual decay rates $\Gamma_{ij}$ (notice that in the following we will use the convention $\Gamma_{ii} = \Gamma$). A second effect of the collective coupling of the atoms to the vacuum is the coherent dipole-dipole interaction characterized by the frequency shifts $\Omega_{ij}$. Both functions depend on $r_{ij}$ and $\theta$ (as detailed for example in [6]). The dynamics of the system can be described by solving a master equation for $\rho$ (the density matrix of the whole system of $N$ emitters),

$$\frac{\partial \rho}{\partial t} = i[\rho, H] + \mathcal{L}[\rho],$$

(1)

where the Hamiltonian is given by

$$H = \frac{\omega}{2} \sum_i \sigma_i^z + \sum_{i \neq j} \Omega_{ij} \sigma_i^+ \sigma_j^-$$

(2)

with $\omega = \omega_0 - \omega_i$ ($\omega_i$ is the reference frequency) while the effect of dissipation is quantified by the Liouvillian

$$\mathcal{L}[\rho] = \frac{1}{2} \sum_{i,j} \Gamma_{ij} \left[ 2\sigma_i^-\sigma_j^+ - \sigma_i^+\sigma_j^- - \rho \sigma_i^z \sigma_j^z \right].$$

(3)

A typical procedure in spectroscopic experiments with two-level systems is the Ramsey method of separated oscillatory fields [20]. The sequence assumes the ensemble of spins initialized in the ground state at time $t_i$ such that $\langle S^z \rangle(t = 0) = -N/2$ where $S^z = \sum \sigma_i^z/2$. Three stages follow: (i) a first quick pulse between $t_i$ and $t = 0$ rotates the atoms into a collective state in the $xy$-plane that exhibits maximal dipole, (ii) the system evolves freely for the time $\tau$ and (iii) a second quick pulse flips the spins up. The detected signal is then a measure of population inversion and therefore proportional to $\langle S^z \rangle(t_f)$. Analysis of this signal gives the sensitivity as a figure of merit in metrology as

$$\delta \omega = \min \left[ \frac{\Delta S^z(\omega, \tau)}{\partial \omega \langle S^z(\omega, \tau) \rangle} \right],$$

(4)

where the minimization is performed with respect to $\omega$.

We follow the dynamics as described above in a density matrix formalism. We start with an initial density matrix $\rho_i = \langle G \rangle \langle G \rangle$, transform it into $\rho_0 = \mathcal{R}_1 \rho_i \mathcal{R}_1^\dagger$, evolve it into $\rho_\tau$ by solving Eq. (1) and finally transform it into $\rho_f = \mathcal{R}_2 \rho_\tau \mathcal{R}_2^\dagger$. The detected signal and its variance are computed as $\langle S^z \rangle$ and $\Delta S^z$ from $\rho_f$.

As a basis of comparison, let us consider the situation of independent systems ($\Gamma_{ij} = 0$ and $\Omega_{ij} = 0$ for $i \neq j$). The rotation pulses are $\mathcal{R}_1 = \mathcal{R}_2 = \otimes_j \mathcal{R}_y^{(j)}[\pi/2]$ and the resulting sensitivity is

$$[\delta \omega]_{\text{indep}} = \min \left[ \frac{\sqrt{\exp(\Gamma/\tau) - \cos^2(\omega\tau)} / \sqrt{N} \sin(\omega\tau)}}{\sqrt{N} \sin(\omega\tau)} \right] = \frac{\exp(\Gamma/\tau)}{\tau \sqrt{N}}. \tag{5}$$

Further optimization with respect to the interrogation time gives an optimal $\tau_{\text{opt}} = 2/\Gamma$ and optimal sensitivity $\Gamma e/2\sqrt{N}$, which shows that the main impediment of Ramsey interferometry is the limitation in the interrogation times owing to the decay of the transition dipoles.

As a principal advance of this paper, we propose a generalized Ramsey sequence (as illustrated in Fig. 1) that deviates from the typical one by a redesign of the two pulses at times $t = 0$ and $t = \tau$, intended to drive the spin system into states that are protected from the environmental decoherence. To accomplish this, one complements the normal $\pi/2$ pulse with a phase distribution pulse, which for a particular atom $j$ is represented by a rotation around the $z$-direction with the angle $\varphi_j^{(m)} = 2\pi m/N(j-1)$, where $m = 1, \ldots [N/2]$ and $[N/2]$ is...
the smallest integer before $N/2$. The first Ramsey pulse operator is then

$$\mathcal{R}_1 = \bigotimes_j \mathcal{R}_{z_j}^{(j)} \left[ \varphi_j^{(m)} \right] \cdot \mathcal{R}_{y_j}^{(j)} \left[ \frac{\pi}{2} \right].$$

The choice of the rotation angles is straightforward to motivate: at time $t = 0$, for any of the angle distributions defined above by the set of $\varphi_j^{(m)}$, the system is in a state of zero average collective spin. At an intuitive level this means that the system folds from a state of maximal classical dipole moment to a non-radiative dipole of zero average. Notice for example that for small atom-atom separations, collective states of higher symmetry are shorter lived than the rest of the states; in the Dicke picture of atoms identically coupled to the vacuum, this culminates in the maximally superradiant state that exhibits a decay rate $N\gamma$. One can eventually deduce the proper rotations that ensure the asymmetry of the chosen states. This can be derived from the orthogonality of the initial state $|\psi_\varphi\rangle = \bigotimes_{j=1}^N \left[ |g\rangle + (e^{i\varphi})^j |e\rangle \right] / \sqrt{2}$ to the multitude of symmetric states of the system. While generally this is an unsolvable problem, we can get some insight from the orthogonality to the symmetric state in the single excitation subspace, the so-called W-state $|W\rangle$.

The imposed orthogonality $\langle W | \psi_\varphi \rangle = \sum_{j=1}^N (e^{i\varphi})^j = 0$ leads to the solutions $\varphi = 2\pi m/N$ which we use to build the $\varphi_j^{(m)}$.

To prepare the system for population difference detection, at time $\tau$ the phase spread is reversed and a $\pi/2$ pulse follows

$$\mathcal{R}_2 = \bigotimes_j \mathcal{R}_{y_j}^{(j)} \left[ \frac{\pi}{2} \right] \cdot \mathcal{R}_{z_j}^{(j)} \left[ -\varphi_j^{(m)} \right].$$

**Analytical results for two atoms** Analytical results are easily derived for the case of two atoms and allow us to already elucidate the differences between typical Ramsey detection and the asymmetric Ramsey procedure. Let us consider atoms 1 and 2 separated by a distance $r$ with a mutual decay rate $\gamma = \Gamma_12(r)$ and dipole-dipole interaction quantified by $\Omega = \Omega_{12}(r)$ (their dependence on $r$ is shown in Fig. 2a). The diagonalization of the Hamiltonian is performed by a transformation from the bare basis $\{|g\rangle, |ge\rangle, |eg\rangle, |ee\rangle\}$ to the collective basis $\{|G\rangle, |S\rangle, |A\rangle, |E\rangle\}$ with $|G\rangle = |gg\rangle$, $|S\rangle = (|eg\rangle + |ge\rangle) / \sqrt{2}$, $|A\rangle = (|eg\rangle - |ge\rangle) / \sqrt{2}$ and $|E\rangle = |ee\rangle$. This change of basis diagonalizes the dissipative dynamics as well, and leads to two independent decay channels with damping rates $\gamma_S = \Gamma + \gamma$ and $\gamma_A = \Gamma - \gamma$ as illustrated in Fig. 2b).

We follow the evolution of the initially prepared $\rho_i = |G\rangle \langle G|$ in the collective basis and compute the detected signal and its variance from the density matrix at time $\tau$. For the symmetric Ramsey sequence one obtains $\langle S^z \rangle_S = 2\sqrt{2} \Re (\rho_{eg}^{SS} + \rho_{ee}^{EE})$ which can be calculated by solving the evolution between 0 and $\tau$ from the following set of coupled equations

$$\dot{\rho}^{ES} = \left[ -\frac{2\Gamma + \gamma_S}{2} - i(\omega - \Omega) \right] \rho^{ES}, \quad (6a)$$

$$\dot{\rho}^{SG} = \left[ -\frac{\gamma_S}{2} - i(\omega + \Omega) \right] \rho^{SG} + \gamma_S \rho^{ES}. \quad (6b)$$

The computational of the signal variance requires the derivative of $\langle (S^z)^2 \rangle_S = 2 \left[ 1 + \rho_{eg}^{SS} - \rho_{ee}^{AA} + 2\Re (\rho_{ee}^{EG}) \right]$ thus solving

$$\dot{\rho}^{EE} = -2\Gamma \rho^{EE}, \quad (7a)$$

$$\dot{\rho}^{SS} = -\gamma_S \left( \rho^{SS} - \rho^{EE} \right), \quad (7b)$$

$$\dot{\rho}^{AA} = -\gamma_A \left( \rho^{AA} - \rho^{EE} \right), \quad (7c)$$

$$\dot{\rho}^{EG} = -\left( \Gamma + 2i\omega \right) \rho^{EG}. \quad (7d)$$

In contrast, for the asymmetric Ramsey sequence we get $\langle S^z \rangle_A = 2\sqrt{2} \Re (\rho_{ee}^{AA} - \rho_{eg}^{AG})$ and $\langle (S^z)^2 \rangle_A = 2 \left[ 1 + \rho_{ee}^{AA} - \rho_{eg}^{AG} - 2\Re (\rho_{eg}^{EG}) \right]$, where the extra coherences can be derived from the solutions of

$$\dot{\rho}^{EA} = \left[ -\frac{2\Gamma + \gamma_A}{2} - i(\omega + \Omega) \right] \rho^{EA}, \quad (8a)$$

$$\dot{\rho}^{AG} = \left[ -\frac{\gamma_A}{2} - i(\omega - \Omega) \right] \rho^{AG} + \gamma_A \rho^{EA}. \quad (8b)$$

The minimum sensitivities depending on $\tau$ after opti-
mization with respect to $\omega$ can be expressed as

$$\delta \omega |_S = \sqrt{2} \frac{(1 + a_S e^{-2\delta \omega} + b_S e^{-2\delta \omega} - c_S e^{-2\delta \omega})}{\tau \cdot e^{-\gamma_S \tau/2} (e^{-\Gamma \tau} A_S^+ + A_S^0)}$$

(9a)

$$\delta \omega |_A = \sqrt{2} \frac{(1 + a_A e^{-2\delta \omega} + b_A e^{-2\delta \omega} - c_A e^{-2\delta \omega})}{\tau \cdot e^{-\gamma_A \tau/2} (e^{-\Gamma \tau} A_A^+ + A_A^0)}$$

(9b)

where $a$, $b$, $c$ and $A^\pm$ are given by the system’s geometry and are listed in the Appendix. While the above expressions are tedious, simplifications are possible in the limit of large $\tau$. Assuming a separation of timescales for example when $\gamma_A \ll \Gamma, \gamma_S$, the sensitivity $\delta \omega |_A$ scales similarly to the independent sensitivity of Eq. 5 with $\Gamma$ replaced by $\gamma_A$. This actually holds approximately even in the intermediate regime shown Fig. 2: where $\gamma_A \approx 0.59 \Gamma$, as transpiring from the scaling of the blue (squares) line. For closely spaced atoms, the result is easy to interpret and extremely encouraging since it allows for large interrogation times and direct improvement of the minimum sensitivity. In the general case, of varying the distance between atoms for example to the second region of Fig. 2, the symmetric state becomes subradiant instead and the symmetric procedure is the optimal one, however providing only a minimal gain over the independent atom case. This is relevant for the case of linear atom chains separated by a magic wavelength $21$, where the conventional Ramsey technique is optimal.

**Numerical results for several atoms** Let us now extend our model to more general configurations of a few two-level systems in various geometries. In principle, the configuration can be generalized to a 2D or 3D lattice but one ends up with large Hilbert spaces rather quickly that render simple numerical methods unfeasible. To illustrate the effectiveness of the asymmetric Ramsey method we particularize to the two situations depicted in Fig. 3 i.e., square and linear geometries. The results are presented in Fig. 3a,b for all possible phase-spread angle sets, i.e., varying the index $m$ of $\varphi_j^{(m)}$ from 1 to $[N/2]$ ($N = 4$ for square and 5 for the chain) and for a lattice constant $a/\lambda = 0.30$.

To provide a simplified general understanding of the results shown in Fig. 3 let us first present a numerical analysis of a simplified case of 5 atoms equally coupled to each other. To this end we shall point out that the method we present should be quite general and applicable to similar systems where the naturally occurring electromagnetic bath that provides mutual decay channels as well as dipole-dipole interactions for dense ensembles of quantum emitters is replaced, for example, by the common interaction of atoms with a decaying optical cavity field $22$. In such a case, by tailoring the atom-field interaction, one can simulate a reservoir that leads to equal mutual coupling between any pair of atoms and equal dipole-dipole couplings. Simultaneous diagonalization of the Hamiltonian and Liouvillian is then possible that leads to $2^N$ states $|\psi_j\rangle$ each with an associated decay channel $\Gamma_j$. In Fig. 3c, we show this exact diagonalization for $N = 5$ and associated $\Gamma_j/\Gamma$ arranged in increasing magnitude from left to right. The upper (red) histogram shows the population distribution for a Ramsey operation while the blue (lower) histogram provides the comparison with the asymmetric Ramsey excitation scheme. The conclusion is straightforward in that it shows that the conventional Ramsey technique Excites on average states decaying faster than $\Gamma$ and the asymmetric scheme populates subradiant states.

While the examples studied in Fig. 3 are a proof-of-principle for the phase-spread mechanism we propose, a general optimization for arbitrary distances and geometries is not straightforward and needs to be accompanied by more sophisticated numerical simulations. For example, in the linear chain case for a ratio $a/\lambda = 0.15$, the two nearest neighbors contribute positively while the outer ones feature a negative coupling (see Fig. 2a). The strategy to be employed is therefore not clear since the various phase shifts are distributed differently along the
chain. For example, as seen in Fig. 3, a simple \( \varphi_j^{(1)} \) phase distribution performs worse than the symmetric Ramsey sequence while great improvement is introduced by applying \( \varphi_j^{(2)} \) shifts.

Experimental investigations of the mechanism described above must mainly address the question of individual phase writing on distinguishable emitters. As one particular realization, a chain of atoms excited by a laser tilted by some angle \( \alpha \) opens up the possibility of imprinting a varying phase \( \varphi_j = k_0(j - 1)\alpha \cos(\alpha) \) for the \( j \)th atom. To realize an optimal phase-spread by angles \( \varphi_j^{(1)} \) for example, one has to fulfill \( \alpha = \arccos(Na/\lambda_0) \). Note, that interestingly for a strontium magic wavelength lattice, excitation at about 90° automatically excites long lived exciton states close to the optimum. In a 2D lattice this still is fulfilled quite well by excitation from the third direction perpendicular to the plane. For a cube the situation is more tricky and requires careful angle optimization for which preliminary calculations are promising and will be fully investigated in a future publication.

Dipole-dipole interactions and collective decay also play a major role in recent experiments of several superconducting q-bits coupled to CPW transmission lines and resonators [23–25]. Here, on the one hand the distance of the particles is much smaller than a wavelength so the effects are very large, but on the other hand the individual transition frequencies, Rabi amplitudes and phases can be controlled very well. The situation is similar to the above described engineered bath for atoms in an optical cavity where the cavity field dissipation induces non-local collective decay of atomic states. In both cases, the individual atoms can be separately addressed: for example a tunable magnetic field gradient can provide the necessary phase gradient across the ensemble allowing for the asymmetric Ramsey procedure to be tested.

Let us finally remark on the connection of our scheme to multipartite entanglement. The folding of the collective atomic state to a partially protected subspace that suffers less from the effects of decoherence hints towards the possibility of preparing entangled atomic states via dissipative techniques. More concretely, assuming the ideal case discussed above where the Liouvillian can be diagonalized and the dissipative evolution consists of independent channels of decay, and assuming the system in a state of minimal dissipation, the final state of the system after considerable evolution time \( \tau \) will be an eigenstate of the Hamiltonian with probable quantum correlations (as a basis of comparison consider the state \( |A \rangle \) and its entanglement for the 2 atom case). An optimization of our scheme by introducing a proper rotation of this final state before detection seems therefore feasible. Moreover, protection of collective states can as well be relevant to schemes where generated entanglement (such as spin squeezed state generation via one axis or two-axis twisting) is exploited and where decoherence has a extremely fast destructive effect. Application of a general principle that would allow multiparticle entanglement to be mapped from fast decaying state to subradiant states could be of great interest.

**Concluding remarks.** We have shown that quantum metrology applications such as frequency measurements via the Ramsey method can benefit from a state protective mechanism that can be directly connected to a transformation that folds initial collective states from the surface of the Bloch sphere to its center. While we have mainly analyzed the simplest collective bath where the vacuum mediates interactions among closely spaced quantum emitters, the procedure should be quite general as for example in the case of engineered baths (atoms in mode-structuring cavities, superconducting q-bits coupled to CPW transmission lines). The connection and application of the mechanism to multiparticle entangled systems indicates possible future directions as it hints towards investigation in i) dissipation-induced entanglement and ii) entanglement (spin squeezed states) protection mechanism.

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Appendix: The $a, b, c, A^\pm$ depend on the relative distance between atoms $r$ via the $\gamma_{S,A}$ decay rates

\[ a_S = \frac{1}{4} \left( \frac{\gamma_A - \gamma_S}{\gamma_S - \gamma_A} \right) \quad a_A = \frac{1}{4} \left( \frac{\gamma_S - \gamma_A}{\gamma_A - \gamma_S} \right) \]
\[ b_S = \frac{4\Gamma - \gamma_S}{4\gamma_A} \quad b_A = \frac{4\Gamma - \gamma_A}{4\gamma_S} \]
\[ c_S = \frac{\gamma_A}{4\gamma_S} \quad c_A = \frac{\gamma_S}{4\gamma_A} \]
\[ \alpha^\pm_S = 1 \pm \frac{\Gamma\gamma_S}{\Gamma^2 + 4\Omega^2} \quad \alpha^\pm_A = 1 \pm \frac{\Gamma\gamma_A}{\Gamma^2 + 4\Omega^2} \]
\[ B_S = \frac{2\Omega\gamma_S}{\Gamma^2 + 4\Omega^2} \quad B_A = \frac{2\Omega\gamma_A}{\Gamma^2 + 4\Omega^2} \]
\[ A^+_S = \sqrt{(\alpha^+_S)^2 + B^2_S} \quad A^+_A = \sqrt{(\alpha^+_A)^2 + B^2_A} \]