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Statistical sensitivity of phase measurements via laser-induced fluorescence with optical cycling detection

Zack Lasner* and D. DeMille
Yale University, New Haven, CT

In atomic and molecular phase measurements using laser-induced fluorescence detection, optical cycling can enhance the effective photon detection efficiency and hence improve sensitivity. We show that detecting many photons per atom or molecule, while necessary, is not a sufficient condition to approach the quantum projection limit for detection of the phase in a two-level system. In particular, detecting the maximum number of photons from an imperfectly closed optical cycle reduces the signal-to-noise ratio (SNR) by a factor of $\sqrt{2}$, compared to the ideal case in which leakage from the optical cycle is sufficiently small. We derive a general result for the SNR in a system in terms of the photon detection efficiency, probability for leakage out of the optical cycle per scattered photon, and the product of the average photon scattering rate and total scattering time per atom or molecule.

Atoms and molecules are powerful platforms to probe phenomena at quantum-projection-limited precision. In many atomic and molecular experiments, a quantum state is read out by laser-induced fluorescence (LIF), in which population is driven to a short-lived state and the resulting fluorescence photons are detected. Due to geometric constraints on optical collection and technological limitations of photodetectors, the majority of emitted photons are typically undetected, reducing the experimental signal. Optical cycling transitions can be exploited to overcome these limitations, by scattering many photons per particle. In the limit that many photons from each particle are detected, the signal-to-noise ratio (SNR) may be limited by the quantum projection (QP) noise (often referred to as atom or molecule shot noise). LIF detection with photon cycling is commonly used in ultra-precise atomic clock [1, 2] and atom interferometer [3] experiments to approach the QP limit.

Molecules possess additional features, beyond those in atoms, that make them favorable probes of fundamental symmetry violation [4–9] and fundamental constant variation [2, 10–14], as well as promising platforms for quantum information and simulation [15–19]. Many molecular experiments that have been proposed, or which are now being actively pursued, will rely on optical cycling to enhance measurement sensitivity while using LIF detection [4, 5, 7–9, 14]. Due to the absence of selection rules governing vibrational decays, fully closed molecular optical cycling transitions cannot be obtained: each photon emission is associated with a non-zero probability of decaying to a "dark" state that is no longer driven to an excited state by any lasers. However, for some molecules many photons can be scattered using a single excitation laser, and up to $\sim 10^6$ photons have been scattered using multiple repumping lasers to return population from vibrationally excited states into the optical cycle [20, 21]. This has enabled, for example, laser cooling and magneto-optical trapping of molecules [22–29]. Furthermore, some precision measurements rely on atoms in which no simply closed optical cycle exists [30, 31]; our discussion here will be equally applicable to such species.

These considerations motivate a careful study of LIF detection for precision measurement under the constraint of imperfectly closed optical cycling. Some consequences of loss during the cycling process have been considered in [32]. However, the effect of the statistical nature of the cycling process on the optimal noise performance has not been previously explored. In particular, the number of photons scattered before a particle (an atom or molecule) decays to an unaddressed dark state, and therefore ceases to fluoresce, is governed by a statistical distribution rather than a fixed finite number. We show that due to the width of this distribution, a naive cycling scheme reduces the SNR to below the QP limit. In particular, we find that in addition to the intuitive requirement that many photons from every particle are detected, to approach the QP limit it is also necessary that the probability of each particle exiting the cycling transition (via decay to a dark state outside the cycle) is negligible during detection. If this second condition is not satisfied, so that each particle scatters enough photons that it is very likely to have been optically pumped into a dark state, then the SNR is decreased by a factor of $\sqrt{2}$ below the QP limit.

Consider an ensemble of N particles in an effective two-level system, in a state of the form

$$|\psi\rangle = (e^{-i\phi}|\uparrow\rangle + e^{i\phi}|\downarrow\rangle)/\sqrt{2}. \tag{1}$$

The relative phase ϕ is the quantity of interest in this discussion. It can be measured, for example, by projecting the wavefunction onto an orthonormal basis $\{|X\rangle \propto |\uparrow\rangle + |\downarrow\rangle, |Y\rangle \propto |\uparrow\rangle - |\downarrow\rangle\}$ such that $|\langle X|\psi\rangle|^2 = \cos^2(\phi)$ and $|\langle Y|\psi\rangle|^2 = \sin^2(\phi)$. In the LIF technique, this can be achieved by driving state-selective transitions, each addressing either $|X\rangle$ or $|Y\rangle$, through an excited state that subsequently decays to a ground state and emits a fluorescence photon. This light is detected, and the resulting total signals, S_X and S_Y, are associated with each state. (This protocol is equivalent to the more standard Ramsey method, in which each spin is reoriented for detection by a spin-flip pulse and the population of spin-up

* zack.lasner@yale.edu

and spin-down particles is measured [33].) The measured value of the phase, $\tilde{\phi}$, is computed from the observed values of S_X and S_Y. In the absence of optical cycling, the statistical uncertainty of the phase measurement is $\sigma_{\tilde{\phi}} = \frac{1}{2\sqrt{N\epsilon}}$, where ϵ is the photon detection efficiency and $0 < \epsilon \leq 1$. Note that $N\epsilon$ is the average number of detected photons; hence, this result is often referred to as the "photon shot noise limit." In the ideal case of $\epsilon = 1$, the QP limit (a.k.a. the atom or molecule shot noise limit) limit $\sigma_{\tilde{\phi}} = \frac{1}{2\sqrt{N}}$ is obtained. This scaling is derived as a limiting case of our general treatment below, where the effects of optical cycling are also considered.

We suppose that the phase is projected onto the $\{|X\rangle, |Y\rangle\}$ basis independently for each particle. Repeated over the ensemble of particles, the total number of particles N_X projected along $|X\rangle$ is drawn from a binomial distribution, $N_X \sim B(N, \cos^2\phi)$, where $x \sim f(\alpha_1, \cdots, \alpha_k)$ denotes that the random variable x is drawn from the probability distribution f parametrized by $\alpha_1, \cdots, \alpha_k$, and $B(\nu, \rho)$ is the binomial distribution for the total number of successes in a sequence of ν independent trials that each have a probability ρ of success. Therefore, $\overline{N_X} = N\cos^2\phi$ and $\sigma^2_{N_X} = N\cos^2\phi\sin^2\phi$, where $\bar{x}$ is the expectation value of a random variable x and σ_x is its standard deviation over many repetitions of an experiment. We define the number of photons scattered from the i-th particle to be n_i, where a "photon scatter" denotes laser excitation followed by emission of one spontaneous decay photon, and define $\overline{n_i} = \bar{n}$ (the average number of photons scattered per particle) and $\sigma_{n_i} = \sigma_n$. Note that these quantities are assumed to be the same for all particles (i.e., independent of i). The probability of detecting any given photon (including both imperfect optical collection and detector quantum efficiency) is ϵ, such that each photon is randomly either detected or not detected. We define d_{ij} to be a binary variable indexing whether the j-th photon scattered from the i-th particle is detected. Therefore, $d_{ij} \sim B(1, \epsilon)$, and it follows that $\overline{d_{ij}} = \epsilon$ and $\sigma^2_{d_{ij}} = \epsilon(1-\epsilon)$.

We define the signal of the measurement of a particular quadrature $|X\rangle$ or $|Y\rangle$ from the ensemble, when projecting onto that quadrature, to be the total number of photons detected. For example, the signal S_X from particles projected along $|X\rangle$ is

$$S_X = \sum_{i=1}^{N_X}\sum_{j=1}^{n_i} d_{ij}. \tag{2}$$

Explicitly, among N total particles, N_X are projected by the excitation light onto the $|X\rangle$ state and the rest are projected onto $|Y\rangle$. The i-th particle projected onto $|X\rangle$ scatters a total of n_i photons, and we count each photon that is detected (in which case $d_{ij} = 1$). The right-hand side of Eq. 2 depends on ϕ implicitly through N_X, and we use this dependence to compute $\tilde{\phi}$, the measured value of ϕ. Because N_X, n_i, and d_{ij} are all statistical quantities, the extracted value $\tilde{\phi}$ has a statistical uncertainty. The QP limit is achieved when the only contribution to uncertainty arises from N_X due to projection onto the $\{|X\rangle, |Y\rangle\}$ basis.

We can compute $\overline{S_X}$ by repeated application of Wald's lemma ([34, 35]), $\overline{\sum_{i=1}^{m} x} = \bar{m}\bar{x}$. This results in

$$\overline{S_X} = N\cos^2\phi\,\bar{n}\epsilon. \tag{3}$$

That is, the expected signal from projecting onto the $|X\rangle$ state is (as could be anticipated) simply the product of the average number of particles in $|X\rangle$, $N\cos^2\phi$, the number of photons scattered per particle, $\bar{n}$, and the probability of detecting each photon, ϵ.

We compute the variance in S_X by repeated use of the law of total variance [36], $\sigma^2_a = \overline{\sigma^2_{a|b}} + \sigma^2_{\overline{a|b}}$, where $\overline{a|b}$ denotes the mean of a conditional on a fixed value of b and, analogously, $\sigma^2_{a|b}$ denotes the variance of a conditional on a fixed value of b. This gives

$$\sigma^2_{S_X} = N\cos^2\phi\,\bar{n}\epsilon^2\left(\frac{1}{\epsilon} + \frac{\sigma^2_n}{\bar{n}} - 1 + \bar{n}\sin^2\phi\right). \tag{4}$$

The results for S_Y are identical, with the substitution $\cos^2\phi \leftrightarrow \sin^2\phi$. Many atomic clocks [37–41] and some molecular precision measurement experiments [4, 7] measure both S_X and S_Y, while others detect only a single state [5, 6, 30, 31]. In what follows, we assume that both states are probed. The case of detecting only one state, with some means of normalizing for variations in $N\bar{n}\epsilon$, can be worked out using similar considerations.

In the regime $\phi = \pm\frac{\pi}{4} + \delta\phi$, where $\delta\phi \ll 1$, sensitivity to small changes in phase, $\delta\phi$, is maximized. In this case, we define the measured phase deviation $\delta\tilde{\phi}$ by $\tilde{\phi} = \pm\frac{\pi}{4} + \delta\tilde{\phi}$. This is related to measured quantities via the asymmetry $\mathcal{A} = \frac{S_X - S_Y}{S_X + S_Y} = \mp\sin(2\delta\tilde{\phi}) \approx \mp 2\delta\tilde{\phi}$. When $N \gg 1$, the average value of $\tilde{\phi}$ computed in this way is equal to the phase ϕ of the two-level system.

The uncertainty in the asymmetry, $\sigma_{\mathcal{A}} \approx \frac{1}{N}\sqrt{\sigma^2_{S_X} + \sigma^2_{S_Y} - 2\sigma^2_{S_X,S_Y}}$, can be computed to leading order in $\delta\phi$ from σ_{S_X}, σ_{S_Y}, and the covariance $\sigma^2_{S_X,S_Y} = \overline{S_X S_Y} - \overline{S_X}\,\overline{S_Y}$ using standard error propagation [42]. We relate $\sigma_{\mathcal{A}}$ to the uncertainty in the measured phase by $\sigma_{\mathcal{A}} = 2\sigma_{\tilde{\phi}}$. This relationship defines the statistical uncertainty in $\tilde{\phi}$, the measured value of ϕ, for the protocol described here. The covariance, $\sigma^2_{S_X,S_Y} = -\frac{N}{4}\bar{n}^2\epsilon^2$, can be calculated directly using the same methods already described. This result can be understood as follows: the photon scattering and detection processes for particles projected onto $|X\rangle$ and $|Y\rangle$ are independent, so the covariance between signals S_X and S_Y only arises from quantum projection. In the simplest case of perfectly efficient, noise-free detection and photon scattering, e.g., $\epsilon = 1$, $\bar{n} = 1$, and $\sigma_n = 0$, the quantum projection noise leads to signal variances $\sigma^2_{S_X} = \sigma^2_{S_Y} = \frac{N}{4}$. The covariance is negative because a larger number of particles projected onto $|X\rangle$ is associated with a smaller number of particles projected onto $|Y\rangle$. The additional factor of $\bar{n}^2\epsilon^2$ for the general case accounts for the fact that both signals S_X and S_Y are scaled by $\bar{n}\epsilon$ when $\bar{n}$ photons are scattered per particle and a proportion ϵ of those photons are detected on average.

The uncertainty in the measured phase, computed using the procedure just described, has the form $\sigma_{\tilde{\phi}} = \frac{1}{2\sqrt{N}}\sqrt{F}$, where we have defined the "excess noise factor" F given in this phase regime by

$$F = 1 + \frac{1}{\bar{n}}\left(\frac{1}{\epsilon} - 1\right) + \frac{\sigma_n^2}{\bar{n}^2}. \tag{5}$$

It is instructive to evaluate this expression in some simple limiting cases. For example, consider the case when exactly one photon is scattered per particle so that $\bar{n} = 1$ and $\sigma_n = 0$. (This is typical for experiments with molecules, where optical excitation essentially always leads to decay into a dark state.) In this case, $F = \frac{1}{\epsilon}$ and the uncertainty in the phase measurement is $\sigma_{\tilde{\phi}} = \frac{1}{2\sqrt{N\epsilon}}$, as stated previously. Alternatively, as $\bar{n} \to \infty$, $F \to 1 + \left(\frac{\sigma_n}{\bar{n}}\right)^2$. This is in exact analogy with the excess noise of a photodetector whose average gain is $\bar{n}$ and whose variance in gain is σ_n^2 [43]. By inspection, the ideal result of $F \to 1$ can be achieved only if $\frac{\sigma_n}{\bar{n}} \to 0$, and either $\epsilon \to 1$ or $\bar{n} \to \infty$.

We now compute $\bar{n}$ and σ_n^2 for a realistic optical cycling process. We define the branching fraction to dark states, which are lost from the optical cycle, to be b_ℓ. We assume that each particle interacts with the excitation laser light for a time T, during which the scattering rate of a particle in the optical cycle is r. Therefore, an average of rT photons would be scattered in the absence of decay to dark states, i.e. when $b_\ell = 0$. (All of our results hold for a time-dependent scattering rate $r(t)$, with the substitution $rT \to \int r(t)dt$.) Note that in the limit $rT \to \infty$, $1/b_\ell$ photons are scattered per particle on average. Recall that the number of photons scattered from the i-th particle, when projected to a given state, is n_i. We define the probability that a particle emits exactly n_i photons to be $P(n_i; rT, b_\ell)$. This probability distribution can be computed by first ignoring the decay to dark states. For the case where $b_\ell = 0$, the number of photons emitted in time T follows a Poisson distribution with average number of scattered photons rT. For the more general case where $b_\ell > 0$, we assign a binary label to each photon depending on whether it is associated with a decay to a dark state. Each decay is characterized by a Bernoulli process, and we use the conventional labels of "successful" (corresponding to decay to an optical cycling state) and "unsuccessful" (corresponding to decay to a dark state) for each outcome. Then $P(n_i; rT, b_\ell)$ is the probability that there are exactly n_i events in the Poisson process, all of which are successful, or there are at least n_i events such that the first $n_i - 1$ are successful and the n_i-th is unsuccessful. (For concreteness, we have assumed that "unsuccessful" decays, i.e., those that populate dark states, emit photons with the same detection probability as all successful decays. The opposite case, in which decays to dark states are always undetected, can be worked out with the same approach and leads to similar conclusions.) Direct calculation gives

$$\bar{n} = \frac{1 - e^{-b_\ell rT}}{b_\ell} \text{ and} \tag{6}$$

$$\sigma_n^2 = \frac{1 - b_\ell + e^{-b_\ell rT} b_\ell (2b_\ell rT - 2rT + 1) - e^{-2b_\ell rT}}{b_\ell^2}. \tag{7}$$

Therefore,

$$F = 1 + \frac{1}{1 - e^{-b_\ell rT}}\left(\frac{b_\ell}{\epsilon} + \frac{1 - 2b_\ell + 2b_\ell e^{-b_\ell rT}(1 - rT(1 - b_\ell)) - e^{-2b_\ell rT}}{1 - e^{-b_\ell rT}}\right). \tag{8}$$

The behavior of the SNR (proportional to $1/\sqrt{F}$) arising from Eq. 8 is illustrated in Fig. 1.

To understand the implications of this result, we consider several special cases, summarized in Table I. We first consider the simple case when cycling is allowed to proceed until all particles decay to dark states, i.e., $b_\ell rT \to \infty$. We refer to this as the case of "cycling to completion." In this case, for the generically applicable regime $\epsilon \leq \frac{1}{2}$ we find $F \geq 2$, even as the transition becomes perfectly closed ($b_\ell \to 0$). We can understand this result intuitively as follows. As the optical cycling proceeds, the number of particles that will still be in the optical cycle after each photon scatter is proportional to the number of particles that are currently in the optical cycle, $\frac{dP}{dn_i} \propto P$. Hence, we expect $P(n_i; rT \to \infty, b_\ell) \propto e^{-\alpha n_i}$ for some characteristic constant α. In fact, one can show that for $rT \to \infty$, this result holds with $\alpha \approx b_\ell$. The width σ_n of this exponential distribution is given by the mean $\bar{n}$; that is, $\sigma_n \approx \bar{n}$. Therefore, we should expect that cycling to completion reduces the SNR by a factor of $\sqrt{F} = \sqrt{1 + (\sigma_n/\bar{n})^2} \to \sqrt{2}$ compared to the ideal case of $F = 1$, which requires $\frac{\sigma_n}{\bar{n}} = 0$.

Surprisingly, this reduction in SNR can be partially recovered for an imperfectly closed optical cycle, by choos-

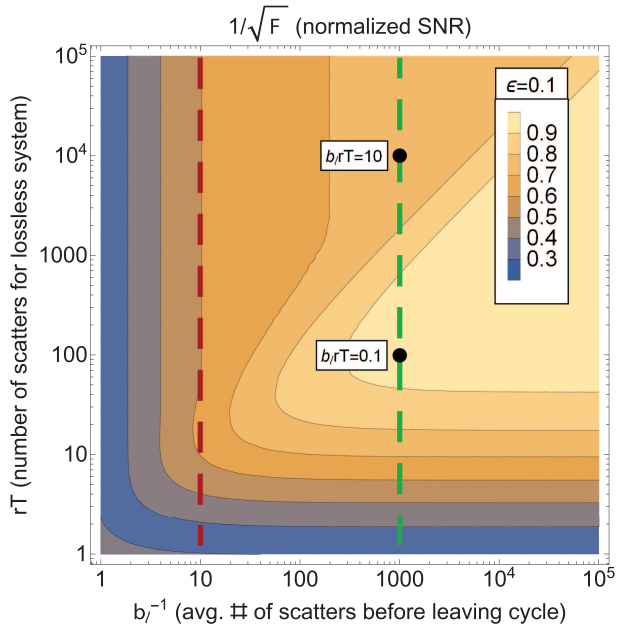

Figure 1. (Color online) $1/\sqrt{F}$, the SNR resulting from Eq. 8, normalized to the ideal case of the QP limit ($F = 1$). This plot assumes $\epsilon = 0.1$. When few photons per particle can be detected, i.e., when $\epsilon/b_\ell \ll 1$ (far left of plot), cycling to very deep completion ($b_\ell rT \gg 1$) does not significantly affect the SNR. Even when one photon per particle can be detected on average, i.e., when $\epsilon/b_\ell = 1$ (dashed red line), the SNR never exceeds roughly half its ideal value. By further closing the optical cycle, i.e. such that $\epsilon/b_\ell \gg 1$ (right of dashed red line), the SNR can be improved to near the optimal value given by the QP limit. However, to reach this optimal regime, the number of photons that would be scattered in the absence of dark states, rT, must be small compared to the average number that can be scattered before a particle exits the optical cycle, $1/b_\ell$. For example, with $1/b_\ell = 1{,}000$ (dashed green line) and $rT = 100$ so that $b_\ell rT = 0.1$ (lower circle), the SNR is more than 30% larger than in the case when $rT = 10{,}000$ and $b_\ell rT = 10$ (upper circle).

ing a finite cycling time, $rT < \infty$, to minimize $\sigma_{\tilde{\phi}}$. The best limiting case, as found from Eq. 8, preserves the condition that many photons are detected per particle, $rT\epsilon \gg 1$, but additionally requires that the probability of decaying to a dark state remains small, $rTb_\ell \ll 1$. In this case, photon emission is approximately a Poisson process for which $\left(\frac{\sigma_n}{\bar{n}}\right)^2 \approx \frac{1}{rT} \ll 1$, and the excess noise factor, F, does not have a significant contribution from the variation in scattered photon number. The optimal value of rT for a finite proportion of decays to dark states, b_ℓ, and detection efficiency, ϵ, lies in the intermediate regime and can be computed numerically.

A special case of "cycling to completion," which must be considered separately, occurs when every particle scatters exactly one photon, corresponding to parameter values $b_\ell = 1$ and $rT \gg 1$ so that $\bar{n} = 1$ and $\sigma_n = 0$. As we have already seen, in this case there is no contribution to the excess noise arising from variation in the scattered

	Condition	Sub-conditon	F
1a	$b_\ell rT \to \infty$		$2 + b_\ell(\frac{1}{\epsilon} - 2)$
1b	$b_\ell rT \to \infty$	$\epsilon \leq 0.5$	≥ 2
2a	$b_\ell rT \to 0$		$1 + \frac{1}{rT\epsilon} + \frac{1}{2}b_\ell(\frac{1}{\epsilon} - 2)$
2b	$b_\ell rT \to 0$	$\epsilon rT \to \infty$	1
3a	$b_\ell \to 1$		$\frac{1}{\epsilon}\frac{1}{1-e^{-rT}}$
3b	$b_\ell \to 1$	$rT \to \infty$	$\frac{1}{\epsilon}$

Table I. The excess noise factor F in some special cases. (1a) All particles are lost to dark states during cycling. (1b) With all particles lost and realistic detection efficiency, $\epsilon \leq 0.5$, $F \geq 2$. (2a) No particles are lost to dark states. (2b) No particles are lost, but many photons per particle are detected. The QP limit is reached. (3a) Up to one photon can be scattered per particle. (3b) Exactly one photon is scattered per particle and the photon shot noise limit is reached.

photon number, and hence the SNR is limited only by photon shot noise: $F = \frac{1}{\epsilon}$.

In atomic physics experiments with essentially completely closed optical cycles, $b_\ell \approx 0$, the limit $b_\ell rT \to \infty$ is not obtained even for very long cycling times where $rT \gg 1$. Instead, in this case $b_\ell rT \to 0$ and hence $F \to 1 + \frac{1}{rT\epsilon}$, which approaches unity as the probability to detect a photon from each particle becomes large, $rT\epsilon \gg 1$. Therefore, the reduction in the SNR associated with the distribution of scattered photons does not occur in this limit of a completely closed optical cycle.

We have also considered how the additional noise due to optical cycling combines with other noise sources in the detection process. For example, consider intrinsic noise in the photodetector itself. Commonly, a photodetector (such as a photomultiplier or avalanche photodiode) has average intrinsic gain $\bar{G}$ and variance in the gain σ_G^2, with resulting excess noise factor $f = 1 + \frac{\sigma_G^2}{\bar{G}^2}$. Including this imperfection in the model considered here leaves Eq. 8 unchanged up to the substitution $\epsilon \to \epsilon/f$. Similar derivations can be performed assuming a statistical distribution of N or ϕ to obtain qualitatively similar but more cumbersome results.

In conclusion, we have shown that a quantum phase measurement, with detection via laser-induced fluorescence using optical cycling on an open transition, when driven to completion, incurs a reduction in the SNR by a factor of $\sqrt{2}$ compared to the QP limit when the optical cycle is driven to completion. This effect has been understood as due to the distribution of the number of scattered photons for this particular case. This reduction of the SNR does not occur for typical atomic systems, where decay out of the optical cycle and into dark states is negligible over the timescale of the measurement. An expression for the SNR has been derived for the general case, in which the cycling time is finite and the probability of decay to dark states is non-zero. For a given decay rate to dark states, an optimal combination of cycling rate and time can be computed numerically to obtain a

SNR that most closely approaches the QP limit. This ideal limit can be obtained only when the photon cycling proceeds long enough for many photons from each atom or molecule to be detected, but not long enough for most atoms or molecules to exit the optical cycle by decaying to an unaddressed dark state.

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