Entanglement-enhanced probing of a delicate material system

Florian Wolfgramm1, Chiara Vitelli2, Federica A. Beduini1, Nicolas Godbout3 and Morgan W. Mitchell1,4*

Quantum metrology1 uses entanglement2–5 and other quantum effects6 to improve the sensitivity of demanding measurements7–10. Probing of delicate systems demands high sensitivity from limited probe energy and has motivated the field’s key benchmark—the standard quantum limit10. Here we report the first entanglement-enhanced measurement of a delicate material system. We non-destructively probe an atomic spin ensemble by means of near-resonant Faraday rotation, a material system. We non-destructively probe an atomic spin first entanglement-enhanced measurement of a delicate process). Examples are found in atomic17, molecular18, condensed systems that suffer significant damage as a result of the probing applications, such as the probing of delicate systems (material and analogous number-limited scenario with very broad potential damage analysis.

According to the Kramers–Kronig relations, interferometric phase shifts are necessarily accompanied by absorption, implying deposition of energy in the probed medium. Absorption also degrades any quantum advantage, as described by recent theory16,21,22. To further complicate matters, in real media the phase shift and absorption may depend on the same unknown quantity. To demonstrate the advantage of our procedure in a fully realistic scenario, we probe a precisely understood material system using a quantum state, permitting rigorous sensitivity and damage properties of the ensemble can be calculated from first principles (Supplementary Sections A–D).

We probe the ensemble with a polarization NOON state, a two-mode entangled state of the form \(|N\rangle_\sigma + \exp[i\varphi]|0\rangle_N|0\rangle_\pi\) (normalization omitted). This describes a photonic ‘Schrödinger cat’ state, a superposition of \(N\) photons that are either all \(\sigma\) - or all \(\pi\) -circularly polarized. NOON states have up to five photons have been produced1, larger NOON states have been made from smaller ones3, and heralded NOON states24,25 have been demonstrated. To this NOON technology we now add tunable, narrowband NOON states that are suitable for efficient interaction with atoms. The use of entangled photons in the single-photon regime allows a rigorous quantification (by quantum state tomography and Fisher Information (FI) theory) of the information gained. To minimize losses while maximizing rotation, we use a NOON state in a 7 MHz spectral window detuned four Doppler widths from the nearest \(^{85}\text{Rb}\) resonance. This requires matter-resonant indistinguishable photons, which we generate for the first time with an advanced downconversion source and ultranarrow atom-based filter.

We studied the ratio of FI to probe-induced damage, that is, to photons absorbed (equivalently scattered) by the ensemble. As both FI and damage scale linearly with the number of probe particles, the same advantage is gained with higher photon numbers, for example, at the projection-noise level. We found that the NOON state beats the SQL by 30 ± 5% for information gained per photon and by 23 ± 4% for damage to the ensemble.

The setup is shown schematically in Fig. 1b. Narrowband NOON states at \(\alpha_{\text{NOON}}\), the optical frequency of the \(5^2S_{1/2} F = 2 \rightarrow 5^2P_{1/2} F' = 1\) transition of the \(D_1\) line of \(^{87}\text{Rb}\), are generated by cavity-enhanced spontaneous parametric downconversion (CESPDC) and sent through the ensemble. The ensemble of \(^{87}\text{Rb}\) atoms is contained in an antireflection-coated vapour cell with internal length \(L = 75\) mm, in a temperature-controlled oven at 70 °C, together with a 0.5% residual \(^{85}\text{Rb}\) component. An applied axial magnetic field \(B\) of up to 60 mT produces resonantly enhanced Faraday rotation of the optical polarization. After leaving the vapour cell, the photons are separated in polarization, frequency-filtered and detected using single-photon counters. Two counters on each polarization output record all possible outcomes; that is, coincidences of HH, HV and VH polarizations are post-selected.

1ICFO – Instituts de Ciencies Fotoniques, Mediterranean Technology Park, 08860 Castelldefels (Barcelona), Spain, 2Center of Life Nanoscience at La Sapienza, Istituto Italiano di Tecnologia, Viale Regina Elena 255, 00181 Rome, Italy, 3COPL, Département de Génie Physique, École Polytechnique de Montréal, C.P. 6079, Succ. Centre-ville, Montréal (Québec) H3C 3A7, Canada, 4ICREA – Institució Catalana de Recerca i Estudis Avançats, 08015 Barcelona, Spain, 5Present address: innoFSPEC, Physical Chemistry, Institute of Chemistry, University of Potsdam, Am Mühlenberg 3, 14476 Potsdam, Germany.

*e-mail: morgan.mitchell@icfo.es
information per photon than possible without entanglement. In repeated probing, fewer photons are needed to achieve the same sensitivity, causing less damage to the sample.

Figure 1 | NOON state probing of an atomic ensemble by Faraday rotation. Polarized photons pass through a rubidium vapour cell (FR), where they experience an optical rotation dependent on the applied magnetic field $B$ (the Faraday effect) and are detected by a polarization analyser (PA) with photon counting detectors (D). 

**a.** With non-entangled photons, ‘singles detection’ can provide high visibility, while ‘coincidence detection’ can provide (low-visibility) super-resolution.

**b.** By means of Hong-Ou-Mandel interference, NOON states can provide both super-resolution and high visibility, providing more information per photon than possible without entanglement. In repeated probing, fewer photons are needed to achieve the same sensitivity, causing less damage to the sample.

![Figure 1](image1.png)

**Figure 2 | High-visibility super-resolving Faraday rotation probing using optical NOON states.** Top curve: for phase reference, singles detection rate (V polarization) versus field strength $B$ shows one oscillation in the range $B = 0–50$ mT. Other curves: coincidence detections $HH$, $HV$ and $VV$ show two oscillations in the same range (super-resolution) and high visibility. Symbols indicate measured data (no background subtracted) with $\pm 1\sigma$ statistical uncertainties. Curves represent models of equation (1) with state $\rho$ found by quantum state tomography.

As seen in Fig. 2, all coincidence outcomes oscillate as a function of $B$, with twofold super-resolution relative to the single-photon oscillation, visible in the singles counts due to a small imbalance between $H$ and $V$ in the input state. The interference visibilities are all $\geq 90\%$, well above the 33% classical limit for $HH$ and $VV$ visibility.$^{26}$ Also shown are predicted coincidence rates $R_i = R_0 P_i$, where $R_0$ is the input flux of pairs and

$$P_i(B) = \text{Tr}[\Pi_i T(B) \rho T^\dagger(B)]$$

(1)

are the outcome probabilities. Here $\rho$ is the two-photon state before the cell, $B$ is the magnetic field, $T(B)$ describes the transmission through the cell, and $\Pi_i$ is the positive-operator-valued measure (POVM) element for the $i$th outcome.

In the $\sigma_{\pm}$ basis, $T(B) = \text{diag}(t_1^2, t_+ t_-, t_- t_+, t_0^2)$, where $t_\pm = \exp[i n_{\pm} B_{\text{NOON}} L/c]$ are transmission coefficients and $n_{\pm}(B)$ are the complex refractive indices from a first-principles calculation (Supplementary Sections A–D). In the $H$, $V$ basis, $\Pi_{HH}$, $\Pi_{HV}$ and $\Pi_{VV}$ are $\text{diag}(1,0,0,0)$, $\text{diag}(0,1,1,0)$ and $\text{diag}(0,0,0,1)$, respectively. A completely analogous description is used for single-photon probabilities. Owing to atomic absorption and scattering, $|t_{\pm}| < 1$ so that $\sum_i P_i < 1$ in general.

The $\rho$ in equation (1) is found by quantum state tomography,$^{27}$ that is, a fit to the observed data using equation (1) and the known $T(B)$ and $\Pi_i$. We find a state, shown in Fig. 3a, with high purity $\text{Tr}[\rho^2] = 0.88$, low photon distinguishability$^{27}$ $\langle \psi |\rho|\psi \rangle = 0.02$, where $|\psi \rangle = (|1\downarrow \, 1\downarrow\rangle - |1\uparrow 1\uparrow\rangle)/\sqrt{2}$, and high fidelity $\langle N_{\psi} |\rho| N_{\psi} \rangle = 0.90$ with the NOON state $|N_{\psi}\rangle = (|2\downarrow 0\uparrow\rangle + \exp[2i\varphi]|0\downarrow 2\uparrow\rangle)/\sqrt{2}$, $\varphi = 0.22$. The phase $\varphi$ is twice the input polarization rotation, which can be chosen to optimize sensitivity at any given value of $B$. Note that both rotation and absorption of the NOON photons are evident in Fig. 2, and contribute to the state reconstruction. A simultaneously acquired time correlation

![Figure 2](image2.png)
I per input photon, the NOON state achieves an advantage in the practical scenario of 30\%. To our knowledge, this is the first demonstration of NOON state characterization. a, Density matrix \(\rho\) (magnitudes only) from quantum state tomography, showing large coherence between |LL\> and |RR\> components. b, Measured correlation of the filtered CESPDC pairs (no background subtracted). The absence of modulation at the 2 ns cavity roundtrip time indicates the presence of a single cavity mode.

Figure 4 | Quantum enhancement in probing of a delicate system, quantified by FI per mean scattering (S) from the \(^{85}\text{Rb}\) atomic ensemble. Thin black curve: FI/S for a single photon of an arbitrarily chosen linear input polarization. Thick orange curve: SQL, the largest FI

The NOON state surpasses the SQL by 40 ± 6\% (23 ± 4\% with \(^{85}\text{Rb}\) contaminant), giving a metrological advantage in the presence of fully realistic and parameter-dependent losses. Low points in the FI are associated with extrema of the coincidence probability curves, that is, regions of small signal sensitivity. For both single-photon and NOON-state probing, the input polarization determines the position of the maxima, allowing high-sensitivity measurement for any value of \(B\).

We rigorously quantify the metrological advantage using the FI,

\[
I(B) = \sum_i P_i |\delta_i \ln P_i|^2
\]

where \(P_i\) are the probabilities derived from the first-principles model in equation (1). The magnetic uncertainty is \(\delta B \rightarrow (IM)^{-1/2}\) in the practical scenario of \(M \gg 1\) uses of the state\(^{29}\). On the basis and the mean number of scattering events is \(s_{\text{scat}} = 2 - |t_{\text{scat}}|^2 - |t_{\text{scat}}|^2\), where \(t_{\text{scat}}\) is the \(^{85}\text{Rb}\) contribution to \(t_s\). A completely analogous calculation is made for single-photon scattering. The NOON state gives an advantage \((I/S)_{\text{NOON}}/(I/S)_{\text{SQL}} = 1.23 \pm 0.04\) over the SQL with our ensemble including the \(^{85}\text{Rb}\) contaminant. If this were

measurement, shown in Fig. 3b, confirms the spectral purity of the NOON state\(^{28}\). To our knowledge, this is the first demonstration of an atom-resonant state with multiphoton coherence.

Figure 5 | Spectroscopic characterization of the rubidium atomic ensemble. Circles show measured values and curves show predictions of first-principles model (see text). a, Saturated absorption spectra acquired with a natural-abundance cell at room temperature as a frequency reference. Horizontal axis shows detuning from the centre of the D1 spectral line. b–d, Transmission spectra for the cell containing \(^{85}\text{Rb}\) plus 0.5\% \(^{87}\text{Rb}\) at temperatures of 22 \(\pm\) 1, 53 \(\pm\) 1 and 83 \(\pm\) 1 \(\pm\) °C, respectively. For each temperature, spectra with measured field strengths of 0, 12, 24, 37, 49 and 58 m\(T\) are shown, in order of increasing line broadening. Grey vertical line shows \(\delta_{\text{NOON}}\), the probe detuning. This operating point gives strong Faraday rotation with low absorption over the range 0–49 m\(T\). Absorption from the small residual \(^{87}\text{Rb}\) component can be seen in d. For clarity, a–c have been vertically offset by 1, 0.75 and 0.5, respectively.
removed, the advantage would be 1.40 ± 0.06 (shown graphically in Fig. 4).

These post-selected results also imply an advantage without post-selection: with available detector efficiency ηdet = 0.95 and source-to-detector path efficiency ηpath = 0.984, the quantum enhancement is 1.21 ± 0.05 per sent photon and 1.15 ± 0.04 per photon scattered from the 85Rb ensemble (1.31 ± 0.06 without 85Rb). Practical applications such as measurement of time-varying fields will require brighter sources of entangled states; narrowband downconversion strategies promise orders-of-magnitude improvement31. Narrowband squeezed states are another route to quantum-enhanced probing, which may allow larger enhancements with bright beams and higher detection efficiencies, although squeezing-induced damage is predicted in some scenarios32 (Supplementary Sections E and F).

We note a previously unreported source of metrological advantage. In the ensemble, as in any material system, the loss depends on the measured quantity (here B). This dependence makes a positive contribution to the FI when it increases ∂1/2 ln P( )/∂B, offsetting the well-known15 FI reduction from ∑P < 1.

We have demonstrated quantum-enhanced probing of an intrinsically delicate system, an atomic spin ensemble. Using narrowband, cavity-enhanced downconversion, we generate a high-fidelity photonic NOON state tuned to an optical resonance of atomic rubidium. When used to probe a rubidium atomic ensemble, the NOON state surpasses the best possible non-entangled probe. The result shows that quantum entanglement can produce a more gentle measurement without sacrificing sensitivity. This is also, to our knowledge, the first use of multiphoton coherence in an atomic physics context. Quantum-enhanced probing of atomic ensembles paves the way for ultra-gentle probing of other delicate systems including exotic phases of cold quantum gases19, single molecules18 and living cells20.

Methods

We use type II CESPDC25,34 to produce N = 2 NOON states. A 795 nm laser, stabilized to νNOON, the frequency of the 5S2−1/2 → 5P1/2, F′ = 1 transition of the D1 line of 87Rb, is used to stabilize the cavity length and is frequency-doubled to pump the SPDC process. In the downconversion process, the cavity resonantly enhances degenerate downconversion into a TEM00 mode with bandwidth 7 MHz around νNOON. Non-degenerate emission, separated by at least the cavity’s 490 MHz free spectral range, is efficiently removed by filtering. The apparatus is described in detail in refs 33,34. By using a low pump power, we ensure that the emitted photons are in a two-photon state ρ, ideally the NOON state (1/2)(1) ρ(12) ρ(12) = (1/2)(1) ρ(12) ρ(12), with negligible four-photon components. H, V, I and R indicate horizontal, vertical, left circular (or −r) and right circular (or r) polarizations, respectively.

Transmission spectra, acquired with a low-power laser passing through the probe beam path, measure the scattering over a broad range of probe frequencies, field strengths and atomic densities, and find very good agreement with the first-principles model, as shown in Fig. 5. The frequency scale of the spectra is determined from simultaneous saturated absorption spectroscopy. Spectra taken at temperatures of 22 °C, 53 °C and 83 °C, and fields in the range 0–58 mT are used to determine the isotope fraction and to calibrate the field and temperature indicators, that is, the current in the coils and the resistance of a thermistor near the cell, respectively.

The filter, described in refs 35,36, has an 80 MHz full-width at half-maximum (FWMH) passband centred on νNOON and >35 dB out-of-band rejection, so that only atom-tuned photons are detected. The single-mode character is evident in the double-exponential arrival-time distribution of the filtered NOON state, shown in Fig. 3b. The combination of CESPDC and narrowband filtering produces a state with ideal characteristics for atomic probing: single-spatial mode, near-perfect indistinguishability and extremely high temporal coherence. In a previous experiment38, we showed that at least 94% of photon pairs are rubidium resonant.

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**Author contributions**

F.W. and M.W.M. conceived and designed the project. F.W., C.V. and F.A.B. designed,
constructed and tested the apparatus. F.W. acquired the data and performed the analysis.
M.W.M. and N.G. performed the modelling and simulation. All authors contributed to the
preparation of the manuscript.

**Additional information**

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and requests for materials should be addressed to M.W.M.

**Competing financial interests**

The authors declare no competing financial interests.