Atomic-Vapor-Based High Efficiency Optical Detectors with Photon Number Resolution

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The ability to detect very weak optical fields with high efficiency ($> 99\%$) and to distinguish the number of photons in a given time interval is a very challenging technical problem with enormous potential payoffs in quantum communications and information processing. We propose to employ an atomic vapor as the active medium, prepared in a specific quantum state using laser radiation. The absorption of a photon will be aided by a dressing laser, and the presence or absence of an excited atom will be detected using the "cycling transition" approach perfected for ion traps. By incorporating an appropriate up-conversion scheme, our method can be applied to a wide variety of optical wavelengths.

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Since its introduction more than 100 years ago [1], the notion of quanta has remained an elusive and misunderstood concept [2]. The ability to detect individual quanta of radiation is of central importance to fundamental physics. A direct example of the ramifications of photon detector efficiencies are the tests of violations of Bell's inequalities [3], which reveal the nonlocal nature of quantum mechanics. By rapidly and randomly measuring the correlations of spacelike separated particles, one can rule out all theories invoking a local realistic view of nature. In order for such a test to be indisputable, however, the detection efficiencies must be very high [4], and to date this has not been achieved in any optical experiment, though an experiment with rapid, random switching has been performed [5]. Recently, however, this "detector loophole" was closed in an elegant experiment using entangled ions [6] (though unfortunately the ions were so close together that the "locality loophole" ruled out any unambiguous test of nonlocality). The cold trapped ions were detected with near 100% efficiency by the technique of light scattering via a "cycling" atomic transition; in brief, each ion is made to emit a large number of photons, which are then easily detected.

The importance of high efficiency photon detectors is not confined to fundamental physics. In addition to the obvious relevance for metrology, recently there have been a number of proposals for realizing scalable quantum computing using only linear optics [7]. A key ingredient for these proposals is very high efficiency photon-counting detectors ($> 99\%$ [8]). In addition, it is crucial that the detectors be able to distinguish the *number* of incident photons (e.g., tell the difference between one and two photons, or more generally, between n and $n + 1$). It has also been noted that such detectors would enable the preparation of novel quantum states of light, e.g., many-photon entangled states [9], which could be of great utility in other quantum information schemes, such as quantum lithography [10]. Finally, there are other applications, such as telecom fiber-based quantum cryptography, where the low efficiencies and noise levels of current detectors significantly limit the achievable distances [11].

Most modern photon detectors rely to a greater or lesser extent on the photoelectric effect: incident photons are converted to individual photoelectrons, either ionized into vacuum or excited into the conduction band of some semiconductor. Either way, one is relying on the capability of amplifying single electrons up to detectable levels of current in order to produce a tangible signal. For example, the silicon avalanche photodiodes used in a many-photon counting experiment typically have efficiencies $\eta \approx 75\%$ [12]. A number of experiments have used a variation of this technology, in which the silicon is lightly doped. These "visible-light photon counters" have displayed $\eta \approx 88\%$ and predicted to be as high as 95% [12,13]. Moreover, they have demonstrated the ability to distinguish the number of initial photoelectrons produced (which for $\eta \approx 100\%$ is the same as the number of incident photons). Unfortunately, these devices require cooling to 6 K and display very high dark count rates (up to $50\,000\ \mathrm{s}^{-1}$), undesirable for quantum communication.

We propose a new approach to the problem: instead of converting each photon to a single photoelectron, we propose a compound process by which a single photon can be converted into many photons. The basis of our proposal is to combine the controlled absorption of light, in a manner inspired by the unprecedented ability to coherently slow and stop light [14] and the high efficiency scheme for projective quantum state measurements in ion traps mentioned above [6].

Our scheme consists of a cell containing the vapor of some atomic species, e.g., an alkali such as cesium (Fig. 1). This vapor will be used to absorb coherently the radiation from an incident beam in a controlled fashion. A number of auxiliary lasers prepare the initial quantum state of the

atoms in the vapor and control the interaction of the atoms with the radiation field. The radiation to be detected is directed into the cell along with an "escort" pulse, giving each photon some small probability to excite an atom to a metastable state — because there are many atoms, however, the chance that each photon is absorbed by one of them can be near unity. Next, a strong readout light is applied, which repeatedly excites any atom in the metastable state; the photons resulting from spontaneous decay may then be detected. And because there are many photons, the chance of not detecting any at all becomes vanishingly small for realistic detector efficiencies. In fact, if an imaging photon detection scheme is used, the number of excited atoms may even be counted, thereby allowing one to reliably distinguish input states of a different photon number.

Four atomic levels will concern us; $|1\rangle$ and $|2\rangle$ are assumed to be the two sublevels of a $^2S_{1/2}$ alkali ground state (for simplicity, we neglect hyperfine structure). The level $|3\rangle$ is the $^2P_{1/2}, m_J = -1/2$ sublevel, and $|4\rangle$ is the $^2P_{3/2}, m_J = 3/2$ sublevel. The atoms are then prepared in state $|1\rangle$ by optical pumping [Fig. 2(a)]. Collisions between atoms will degrade this state preparation by exciting population back into state $|2\rangle$. The time taken for such collisions to occur is $\tau_{\rm col} \approx \sqrt{M/3k_BT}/n\sigma$, with M the atomic mass, k_B Boltzmann's constant, T the temperature, n the number density of atoms, and σ the collisional cross section ($\sim 10^{-14}$ cm^2 for alkalis). Collisions can be mitigated by use of buffer gas, decreasing the atomic density, cooling the atoms, using heavier atoms, or by raising the energy difference between $|1\rangle$ and $|2\rangle$ (thereby making it less likely that a single collision will impart sufficient energy to induce the transition). The following two operations (i.e., photon absorption and readout) must be accomplished in a time considerably smaller than $\tau_{\rm col}$.

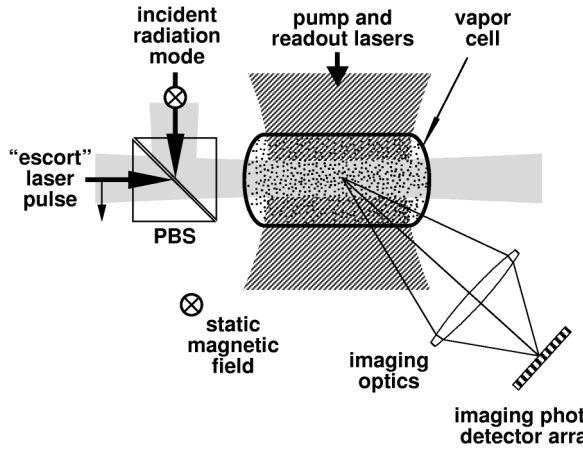

FIG. 1. Diagram illustrating schematically the proposed photodetection technique. The polarizing beam splitter (PBS) dictates the polarizations of the applied optical fields, as assumed in deriving the Hamiltonian Eq. (1).

Once the atoms are in state $|1\rangle$, the photon field is directed into the cell, accompanied by a strong escort laser pulse. The resulting two-photon Raman excitation to state $|2\rangle$ is described by the following interaction picture Hamiltonian:

$$\hat{H}_I(t) = \sum_{i=1}^{N} \frac{\hbar\Omega(\mathbf{r}_i,t)}{2}(|3\rangle\langle 2|)_i \exp[i(\omega_{32}-\omega_e)t] + i\hbar\sum_{i=1}^{N}\sum_{\lambda} g_{\lambda,i}\hat{a}_\lambda(|3\rangle\langle 1|)_i \exp[i(\omega_{31}-\omega_\lambda)t] + \text{H.a.} \quad (1)$$

Here $\Omega(\mathbf{r}_i,t) = \langle 3|\hat{\mathbf{d}}|2\rangle \cdot \mathcal{E}_e(\mathbf{r}_i,t)/\hbar$ is the Rabi frequency of the $|2\rangle \leftrightarrow |3\rangle$ transition of the ith atom due to the escort pulse [represented by the analytic signal $\mathcal{E}_e(\mathbf{r}_i,t)\exp(-i\omega_e t)$, with central frequency ω_e]. The coefficient $g_{\lambda,i} = \sqrt{\omega_\lambda/2\varepsilon_0\hbar}\Phi_\lambda(\mathbf{r}_i)\langle 3|\hat{d}_z|1\rangle$, where $\Phi_\lambda(\mathbf{r}_i)$ is the spatial mode function for the λth mode of the incident field (with annihilation operator $\hat{a}_\lambda$ and frequency ω_λ),

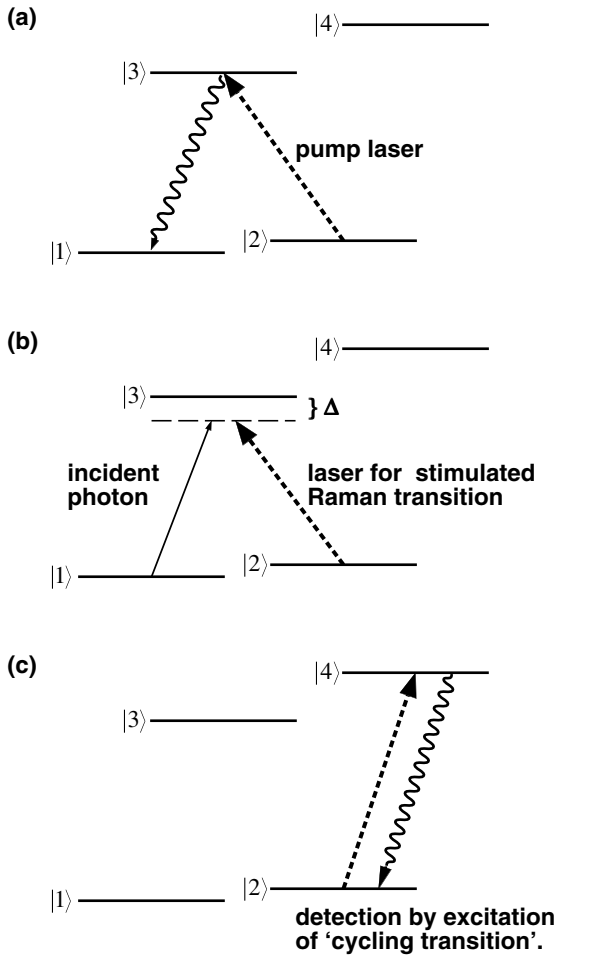

FIG. 2. Diagram illustrating the atomic transitions envisioned for the three-stage photodetection procedure. Stage (a) is optical pumping to level $|1\rangle$; stage (b) is the absorption of the photon with the assistance of a classical escort pulse; stage (c) is the detection of any atom in state $|2\rangle$.

ε_0 is the permittivity of the vacuum, and $\hat{\mathbf{d}}$ is the dipole moment operator. In writing (1), we assume that the escort and photon fields have negligible effect on the $|1\rangle \leftrightarrow |3\rangle$ transition; this is valid for a particular orientation of static magnetic field and specific polarizations for the escort field and the photon (see Fig. 1).

We assume that both the photon and the escort field are far detuned from resonance with the transitions $|1\rangle \leftrightarrow |3\rangle$ and $|2\rangle \leftrightarrow |3\rangle$, respectively. We then adiabatically eliminate the population of the upper level $|3\rangle$, and the dynamics of the system may be described to a good approximation by the following effective Hamiltonian:

$$\hat{H}_{\text{eff}}(t) = i\hbar \sum_{i=1}^{N} \sum_{\lambda} f_{\lambda,i}(t) \hat{a}_{\lambda}^{\dagger} (|1\rangle\langle 2|)_i \exp[i(\omega_{\lambda} - \omega_0)t] + \text{H.a.}, \tag{2}$$

where $f_{\lambda,i}(t) = \Omega(\mathbf{r}_i, t) g_{\lambda,i}^{*}/2\Delta$, $\omega_0 = \omega_{21} + \omega_e$, and $\Delta = \omega_{31} - \omega_0 \approx \omega_{31} - \omega_{\lambda}$ [15]. This is the Hamiltonian describing an effective two-level system interacting with a quantized field; the coupling constant $f_{\lambda,i}$ can be controlled by shaping the profile of the escort pulse.

An important property of this Hamiltonian is that it commutes with the "total excitation" operator $\sum_i (|2\rangle\langle 2|)_i + \sum_{\lambda} \hat{a}_{\lambda}^{\dagger} \hat{a}_{\lambda}$. As a consequence, the number of quanta of the radiation field plus the number of excited atoms must be a constant. Therefore, once the incident radiation field has been completely absorbed, the number of quanta it contained may be determined by measuring the number of atoms in the excited state. If we confine ourselves to single-photon incident fields, the wave function will thus have the form

$$|\psi(t)\rangle = \sum_{\lambda} \alpha_{\lambda}(t) |1_{\lambda}\rangle |g\rangle + \sum_{i} \beta_i(t) |\text{vac}\rangle |2_i\rangle, \tag{3}$$

where $|g\rangle$ represents the state in which *all* of the atoms are in state $|1\rangle$; $|2_i\rangle \equiv (|2\rangle)_i |g\rangle$ the state in which they are in state $|1\rangle$, except the ith atom which is in state $|2\rangle$; $|\text{vac}\rangle$ is the field vacuum state; and $|1_{\lambda}\rangle \equiv \hat{a}_{\lambda}^{\dagger} |\text{vac}\rangle$ is the state with one photon in mode λ. The probability amplitudes $\alpha_{\lambda}(t)$ and $\beta_i(t)$ obey the equations

$$\begin{aligned} \dot{\alpha}_{\lambda}(t) &= -\sum_{i} f_{\lambda,i}(t) \beta_i(t) \exp[i(\omega_{\lambda} - \omega_0)t], \\ \dot{\beta}_i(t) &= \sum_{\lambda} f_{\lambda,i}^{*}(t) \alpha_{\lambda}(t) \exp[-i(\omega_{\lambda} - \omega_0)t]. \end{aligned} \tag{4}$$

By formally solving for $\alpha_{\lambda}(t)$, we obtain an integro-differential equation for $\beta_i(t)$; under the Markov approximation and the assumption that we can neglect coherence between different atoms (so that superradiant effects are negligible), we obtain the following equation for $\beta_i(t)$:

$$\dot{\beta}_i(t) = \epsilon_i(t) \phi_i(t) - \frac{A_{31}}{2} |\epsilon_i(t)|^2 \beta_i(t), \tag{5}$$

where A_{31} is the spontaneous decay rate of the $|3\rangle \rightarrow |1\rangle$ transition, $\epsilon_i(t) = [i\Omega(\mathbf{r}_i, t) \exp(-i\omega_0 t)/2\Delta]^{*}$ characterizes the influence of the escort pulse, and $\phi_i(t) = \langle 3|\hat{d}_z|1\rangle \sum_{\lambda} \sqrt{\omega_{\lambda}/2\hbar\varepsilon_0} \alpha_{\lambda}(0) \Phi_{\lambda}(\mathbf{r}_i) \exp(-i\omega_{\lambda} t)$ characterizes the action of the photon we wish to detect. The first term on the right-hand side of Eq. (5) represents the absorption of the photon, the second term reemission. Equation (5) can be solved in closed form, yielding explicit expressions (dependent on the photon- and escort-pulse shapes) for the absorption and scattering probabilities. In what follows, we have employed expressions derived from simple square-shaped pulses for both the photon and the escort pulses (although model dependent factors of order unity have been suppressed).

Once our single photon is absorbed by the atoms, we can detect the excitation with very high probability by employing a third auxiliary laser. This "readout" laser is carefully tuned and polarized so that any population in each atom's excited state is pumped into some convenient upper state $|4\rangle$ [Fig. 2(c)], chosen so that it will rapidly decay spontaneously back to the excited state *only* [6]. If the readout light persists for some microseconds, the atom in the excited state $|2\rangle$ will scatter many photons. By detecting and imaging the scattered radiation, one can therefore determine which atom is excited. The time taken for one such photon to be registered is $t_{\text{ro}} = (2\Omega_r^2 + A_{24}^2)/A_{24}\Omega_r^2 \eta_{\text{det}}$, Ω_r being the readout laser Rabi frequency, A_{24} the decay rate of the $|4\rangle \rightarrow |2\rangle$ transition, and η_{det} the overall detection efficiency of the imaging system, including solid angle acceptance and photon detector efficiency.

The detector efficiency is limited by three effects: the possibility that the photon may be scattered rather than absorbed, the possibility that the photon may pass through the medium without being absorbed by any atom, and the possibility that the atom which has absorbed the photon may be collisionally deexcited before the readout can take place. Using results derived from Eq. (5), we find that the efficiency is given by

$$\eta \approx 1 - \left(\frac{T_p A_{31} \Omega_e^2}{16\Delta^2}\right)^2 - \exp\left[\frac{-q\ell_{\text{cell}}}{\ell_{\text{abs}}}\right] - \frac{t_{\text{ro}}}{2\tau_{\text{col}}}. \tag{6}$$

Here T_p is the duration of the photon and escort pulses, Ω_e is the (constant) value of the escort Rabi frequency, ℓ_{cell} is the length of the cell, q is the number of passes through the cell made by the photon and escort, and $\ell_{\text{abs}} = \Delta^2/(\lambda_{\text{ph}}^2 n \Omega_e^2 T_p A_{13})$ is the absorption length of the medium (λ_{ph} being the photon wavelength). Assuming a cesium density of 10^9 cm^{-3}, cooled to the Doppler limit of $\sim 10^{-3}$ K (thereby rendering collisional effects negligible [16]), a pulse duration T_p of 10 nsec, a detuning Δ of 0.5 GHz, a strong escort pulse with $\Omega_e = A_{13}$, a cell length $\ell_{\text{cell}} = 2$ mm, a beam area $A = 10^{-2}$ mm^2, $q = 100$ passes through the cell, and $\eta_{\text{det}} = 1/8$, then the theoretical detection efficiency will be $\eta \approx 99.8\%$.

Multiple photons can also be reliably measured by this technique. As long as the total number of incident photons is much less than the total number of atoms

participating in the measurement, each photon will be absorbed by a different atom, and the number of fluorescing atoms observed during readout will be equal to the number of photons. Being able to spatially resolve the atoms will assist in counting them (the image of the moving atom emitting photons is somewhat reminiscent of the tracks in a bubble chamber), though even this is not completely necessary, as demonstrated in [6]; as long as the number of emitting atoms in each optically resolvable volume is sufficiently small that the photon statistics of 1, 2, etc., atoms radiating are distinguishable, the total number of atoms in state $|2\rangle$ can be determined. For example, with $\eta \approx 99.8\%$, we could in principle reliably distinguish states with ~ 50 photons.

A serious potential problem in realizing this scheme will be "dark counts." Provided collisional excitation of state $|2\rangle$ can be kept negligible, the principle mechanism by which this can occur is excitation out of $|1\rangle$ by the readout laser. In the example discussed above, the readout laser polarized to address the $|2\rangle \leftrightarrow |4\rangle$ transition can excite population in $|1\rangle$ to the ${}^2P_{3/2}, m_J = 1/2$ sublevel. Because of the Zeeman splitting, this transition will be detuned by $\delta = 2\mu_B B/3\hbar$ (μ_B being the Bohr magneton and B the magnetic field strength), and due to the different dipole moment strengths, the Rabi frequency will be reduced by a factor of $\sqrt{3}$. The probability of an atom emitting a dark count photon during readout is then $P_{\mathrm{dc}} \approx t_{\mathrm{ro}} A_{42} \Omega_r^2/6(\delta^2 + \Omega_r^2/3)$. For a magnetic field $B = 1$ T and $\Omega_r = 0.01 A_{42}$, $P_{\mathrm{dc}} \sim 2 \times 10^{-5}$. The large number of atoms N present in the active medium may make dark counts a non-negligible effect (the above parameters yield $N = 20\,000$ and a net dark count probability of 0.4). Ways to mitigate this problem, e.g., allowing the photon and its escort to pass through the atoms multiple times (thereby allowing a reduction in the number of atoms without a detrimental effect on the detection efficiency), or reducing the area of the optical beams, are a subject of an ongoing investigation. Another potential source of dark counts is the inability to perfectly transfer all atoms to state $|1\rangle$ by optical pumping. However, this can be checked by performing a readout sequence *before* exposing the atoms to the incident and escort pulses.

It may be argued that a detector of this sort is of limited value because it operates only for a single frequency, and not, e.g., at the telecommunications wavelength of 1550 nm. If indeed we start out with a photon with a different wavelength (but still assumed to be of narrow bandwidth), we estimate that by first mixing the photon with a strong pulse of an appropriate frequency in a nonlinear optical crystal, we can have a near-unity probability of up-converting the photon to the frequency needed for our detection scheme. In this way the high efficiency detection method proposed above may be used over much of the optical spectrum.

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Note added.—While finishing our manuscript, a similar proposal came to our attention, suggested independently and essentially simultaneously [17].

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