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Feasibility of single-photon cross-phase modulation using metastable xenon in a high finesse cavity

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ABSTRACT

Cross-phase modulation at the single-photon level has a wide variety of fundamental applications in quantum optics including the generation of macroscopic entangled states. Here we describe a practical method for producing a weak cross-phase modulation at the single-photon level using metastable xenon in a high finesse cavity. We estimate the achievable phase shift and give a brief update on the experimental progress towards its realization. A single-photon cross-phase modulation of approximately 20 milliradians is predicted by both a straightforward perturbation theory calculation and a numerical matrix diagonalization method.

1. Introduction

A wide variety of proposed experiments in quantum optics make use of cross-phase modulation at the single-photon level. It is particularly useful in the creation of Schrodinger cat states and entangled coherent states which have applications in quantum computing [1–3], teleportation [4–6], metrology [7], cryptography [8], and in nonlocal interferometry [9,10]. Nonclassical effects involving entangled coherent states are also useful for probing the boundary between classical and quantum behavior. Coherent states are the closest approximation to a classical state of light, making superpositions of sufficiently orthogonal coherent states a truly macroscopic quantum phenomenon.

Experiments to create cross-phase modulation at the single-photon level have been performed with many different technologies and nonlinear media. Single atoms in micro-cavities have been used [11], as well as atomic vapor in a hollow core fiber [12], transmons at microwave wavelengths [13], and a variety of systems using electromagnetically induced transparency [14–16]. Other efforts have used quantum dots in a cavity [17] and strongly interacting Rydberg atoms [18]. Large per photon phase shifts have been measured in many of these systems, but they require relatively complicated experimental setups, prompting a search for a simpler and more reliable source of low power cross-phase modulation.

Here we discuss the feasibility of a new cavity approach for single-photon level cross-phase modulation that uses metastable xenon atoms as the nonlinear medium. Meta-stable xenon is expected to be superior to alkali vapors such as rubidium and cesium since it is inert and does not adhere to optical surfaces [19]. Xenon also has a long metastable lifetime and relatively large dipole matrix elements with a convenient set of ladder transitions in the near infrared. The two level spacings are relatively close in wavelength, allowing approximately Doppler-free experiments with counterpropagating beams.

The use of a high finesse cavity should avoid the limitations in using freely propagating beams that have been pointed out by Shapiro and others based on a multi-mode analysis [20–22]. These difficulties do not occur as long as only a single cavity resonant frequency exists within the bandwidth of the medium.

We estimate that a single photon in the setup described here will be able to produce a nonlinear phase shift of approximately 20 milliradians, as described in Section 2. Two different methods for calculating the magnitude of the expected cross-phase modulation are found to be in good agreement. One of these consists of a straightforward analytical calculation based on perturbation theory. Those results are then verified using a numerical matrix diagonalization method which is more appropriate for large numbers of atoms and small detunings. In Section 3 we briefly describe the progress of an ongoing experimental effort towards the realization of the metastable xenon approach. Finally in Section 4 we provide a conclusion and summary of results.

2. Theoretical model

2.1. Three-level system

The xenon transitions of interest form a three-level ladder system as pictured in Fig. 1, where ω_1 and ω_2 represent the control

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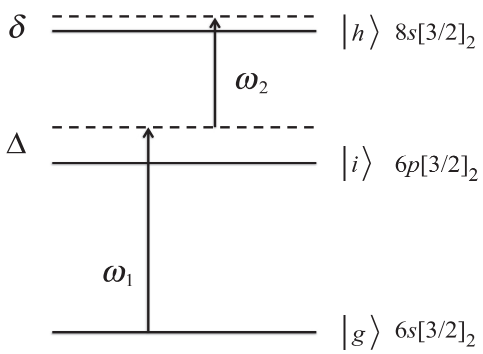

Fig. 1. Three-level system in metastable xenon used to generate cross-phase modulation. The detunings of the control and signal (ω_1 and ω_2) are given by Δ and δ respectively. The atomic levels are represented by $|g\rangle$, $|i\rangle$, and $|h\rangle$, where $|g\rangle$ is the metastable state with an intrinsic lifetime of approximately 43 s [23]. The transitions of interest correspond to wavelengths of 823 and 853 nm for the control and signal respectively [24,25].

and signal photon frequencies, respectively. Two-photon absorption can be minimized by detuning the signal and control photons from atomic resonance, so that the net effect is a conditional nonlinear phase shift. For applications involving the generation of phase-entangled coherent states [9,10], the control at ω_1 would be a single photon while the signal at ω_2 would be a weak coherent state.

Three-level systems of this kind have previously been analyzed using a density matrix approach [26]. Our goal here is to use a straightforward perturbation calculation to obtain an approximate estimate of the cross-phase modulation in metastable xenon, which we can use to demonstrate the feasibility of the approach.

We define a set of basis states for describing the interaction in Fig. 1 as

$$
\begin{aligned}
|1\rangle &= |0\rangle \otimes |h\rangle \\
|2\rangle &= \hat{a}^{\dagger}_{\omega_1}|0\rangle \otimes |i\rangle \\
|3\rangle &= \hat{a}^{\dagger}_{\omega_2}|0\rangle \otimes |i\rangle \\
|4\rangle &= \hat{a}^{\dagger}_{\omega_1}\hat{a}^{\dagger}_{\omega_2}|0\rangle \otimes |g\rangle.
\end{aligned} \tag{1}
$$

Here $\hat{a}^{\dagger}_{\omega_i}$ is the usual creation operator for angular frequency ω_i and $|0\rangle$ is the vacuum state of the field. In this basis the interaction Hamiltonian $\hat{V}$ can be defined in the usual way as [27]

$$
\begin{aligned}
\hat{V} = {} & m_1^*\hat{\sigma}^{\dagger}_{gi}\hat{a}_{\omega_1} + m_2^*\hat{\sigma}^{\dagger}_{ih}\hat{a}_{\omega_2} + m_1\hat{\sigma}_{gi}\hat{a}^{\dagger}_{\omega_1} + m_2\hat{\sigma}_{ih}\hat{a}^{\dagger}_{\omega_2} \\
& + m_3^*\hat{\sigma}^{\dagger}_{gi}\hat{a}_{\omega_2} + m_3\hat{\sigma}_{gi}\hat{a}^{\dagger}_{\omega_2},
\end{aligned} \tag{2}
$$

where $\hat{\sigma}_{gi}$ takes the atom from $|i\rangle$ to $|g\rangle$, $\hat{\sigma}_{ih}$ takes the atom from $|h\rangle$ to $|i\rangle$ and the m terms are the transition matrix elements. In general the matrix elements are given by $m = \langle -\vec{\mu} \cdot \vec{E} \rangle$ where μ is the dipole moment of the transition and $\vec{E}$ is the electric field, and the brackets indicate an average over orientations.

The basis states of Eq. (1) and the interaction Hamiltonian of Eq. (2) describe a system that can undergo several kinds of transitions. The system may initially transition from state $|4\rangle$ to either state $|3\rangle$ or state $|2\rangle$ by the absorption of the control (ω_1) or signal (ω_2) photons respectively. A second photon may then be absorbed to take the system from states $|2\rangle$ or $|3\rangle$ to state $|1\rangle$ [26,28]. Using Eqs. (1) and (2) the total Hamiltonian $\hat{H}$ of the system can be written as

$$
\hat{H} = \begin{pmatrix} \hbar(\omega_{hi}+\omega_{ig}) & 0 & m_2^* & 0 \\ 0 & \hbar(\omega_1+\omega_{ig}) & 0 & m_3^* \\ m_2 & 0 & \hbar(\omega_2+\omega_{ig}) & m_1^* \\ 0 & m_3 & m_1 & \hbar(\omega_1+\omega_2) \end{pmatrix} \tag{3}
$$

The finite lifetimes of the excited levels are not taken into account here in order to keep the presentation as transparent as possible. Inclusion of the lifetimes reduces the cross-phase modulation by an amount that is not significant for detunings much larger than the line width, as is expected to be the case in the planned experiments. The intrinsic lifetime of the metastable $6s[3/2]_2$ state (approximately 43 seconds [23]) has no significant effect on the results.

2.2. Perturbation theory

A straightforward perturbation theory approach can be used to estimate the cross-phase shift for sufficiently large detunings. In that limit, the nonlinear phase shift can be calculated for a single atom and then summed over the contributions from all of the atoms. This approach is valid as long as the depopulation of the initial state is sufficiently small, as will be verified below using a numerical diagonalization technique.

The level spacings and detunings are chosen in such a way that the control photon effectively interacts only with levels $|g\rangle$ and $|i\rangle$ while the signal photon only interacts with levels $|i\rangle$ and $|h\rangle$. To fourth order in perturbation theory, each photon is absorbed and re-emitted once, returning the atom back to the ground state. The assumption that only this 4th-order term is necessary to predict the phase shift is confirmed by the numerical approach of Section 2.3, which takes all orders into account.

The fourth order term of interest gives a change $E^{(4)}$ in the energy of the system given by [26]

$$E^{(4)} = \frac{|m_1|^2|m_2|^2}{\hbar^3\Delta^2\delta}. \tag{4}$$

The matrix elements are a function of position within the cavity, which is assumed to contain a uniform density ρ of metastable xenon atoms. The total phase shift from a single atom is determined by the fact that the time dependence of the state is proportional to $exp[-iE^{(4)}t/\hbar]$, which gives a total phase shift of

$$\phi = \rho \int \frac{E^{(4)}t}{\hbar}\, dV. \tag{5}$$

Here the integral is over the cavity volume and t is the interaction time inside the cavity.

The integral of Eq. (5) can be simplified by making two approximations regarding the electric field within the cavity. First we replace the sinusoidally varying field with a suitable average, since the field oscillates on a length scale much smaller than the size of the cavity. This average is found by normalizing the energy of the electric field in the cavity to that of a single photon of the proper wavelength. Secondly we model the cavity mode field distribution as a constant electric field over a cylinder with a diameter equal to the gaussian beam diameter and length equal to that of the cavity. These approximations are made only to simplify the presentation. Numerical integration of Eq. (5) using the exact field distribution calculated from the geometry of the cavity [29] is in good agreement and will be discussed in Section 2.4.

The average total phase shift can now be written as

$$\phi \approx \rho V_{cyl} \frac{|\overline{m}_1|^2|\overline{m}_2|^2}{\hbar^4\Delta^2\delta} t, \tag{6}$$

where V_{cyl} is the volume of the cylinder we have used to model the cavity mode field distribution, and $\overline{m}_i$ represents the matrix element defined in Section 2.1 where the electric field has been averaged over the cavity field distribution. In addition, the number of atoms involved in the interaction is effectively reduced to 1/3 of the number present in the ensemble due to averaging of the electric field and dipole moment orientations.