Enhancement of Kerr nonlinearity via multi-photon coherence

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(November 10, 2018)

We propose a new method of resonant enhancement of optical Kerr nonlinearity using multi-level atomic coherence. The enhancement is accompanied by suppression of the other linear and nonlinear susceptibility terms of the medium. We show that the effect results in a modification of the nonlinear Faraday rotation of light propagating in an $^{87}$Rb vapor cell by changing the ellipticity of the light.

Efficient manipulation with optical quanta calls for materials possessing large and lossless optical nonlinearities. Recently it was shown that coherent atomic effects such as electromagnetically induced transparency (EIT) and coherent population trapping (CPT) are able to suppress the linear absorption of resonant multilevel media while keeping the nonlinear susceptibility at a very high level. Previous experimental work has shown that coherent media produce an effective interaction between two electromagnetic fields due to both refractive and absorptive Kerr nonlinearities.

One method of producing Kerr nonlinearity with vanishing absorption is based on the coherent properties of a three-level Λ configuration (see Fig. 1b), in which the effect of EIT can be observed. However, since an ideal CPT medium does not interact with the light, it also cannot produce any nonlinear effects. To get a nonlinear interaction in such a medium one needs to “disturb” the CPT regime by introducing, for example, the interaction with an additional off-resonant level (the Ν-type scheme, Fig. 1c). If the disturbance of CPT is small, we may distort this CPT and produce a strong nonlinear coupling among electromagnetic fields interacting with the atomic system.

An important advantage of the Ν-type level configuration is that this type of coherence is easily created on the Zeeman sublevels of alkali atoms. In this particular case large self-phase modulation of circularly polarized light is possible. We show here significant change in nonlinear magneto-optical polarization rotation due to Ν-type coherence in Rb vapor. Our experimental results confirm the theoretical predictions.

Our method of creation of a highly nonlinear medium with small absorption has prospects in fundamental as well as applied physics. An advantage of the Ν configuration is that by increasing the number of the levels it is possible to realize higher orders of nonlinearity. This can be used for construction of non-classical states of light as well as coherent processing of quantum information.

Let us first consider a medium (atomic, molecular, semiconductor, etc.) with an Ν-type energy level structure as in Fig. 1c. Here levels $|α_j⟩$ have natural decays $γ_j$ and we assume that the ground state levels $|b_j⟩$ have no decay. The coherence between levels $|b_i⟩$ and $|b_j⟩$ (i ≠ j) have slow homogeneous decay $γ_0$. The energy levels are coupled by weak probe electromagnetic fields of Rabi frequency $α_j$ and strong coupling fields of Rabi frequency $Ω_j ≫ |α_j|$. For the sake of simplicity we assume all the fields to be resonant with the corresponding atomic transitions except for the coupling field $Ω_2$ which has small detuning $δ ≪ γ_2$ from the $|b_3⟩ → |α_2⟩$ transition.

We first focus on the case when there is no coherence decay in the system. Then the interaction of the atoms and the fields can be described in the slowly varying amplitude and phase approximations by the Hamiltonian

$$H_M = -\hbar δ |b_3⟩⟨b_3| + \hbar \sum_{j=1}^{2} (α_j |a_j⟩⟨b_j| + |Ω_j⟩⟨a_j|b_{j+1}| + H.c.)$$

where $H.c.$ means Hermitian conjugation. If $δ = 0$, there exists the noninteracting eigenstate (“dark eigenstate” $|γ⟩$) of this Hamiltonian, corresponding to the zero eigenvalue $λ_D = 0$:

$$|D⟩ = \frac{α_1 α_2 |b_3⟩ - Ω_2 α_1 |b_2⟩ + Ω_3 b_1|}{\sqrt{|α_1|^2 |α_2|^2 + |Ω_2|^2 |α_1|^2 + |Ω_1|^2 |Ω_2|^2}}.$$  \hspace{1cm} (1)

Strictly speaking, there is no “dark state” for finite $δ$. However when the detuning is small enough ($δ ≪ γ_2$, $|Ω_2|^2/γ_2$), the disturbance of the “dark” state is small. The disturbed dark state $|D⟩$ and the corresponding eigenvalue of the Hamiltonian $λ_D$ in the limit of $|Ω_j| ≫ |α_j|$ are:

$$|D⟩ \approx \zeta \left[ |D⟩ - \frac{α_j^* |α_2|^2}{|Ω_1|^2 |Ω_2|^2} |a_1⟩ + \frac{δ}{|Ω_1||Ω_2|^2} |a_2⟩ \right],$$  \hspace{1cm} (2)

$$λ_D \approx -\hbar \frac{|α_j|^2 |α_2|^2}{|Ω_1|^2 |Ω_2|^2}.$$  \hspace{1cm} (3)

1

arXiv:quant-ph/0207141v1 24 Jul 2002
where $\zeta \simeq 1$ is a normalization parameter.

Because the system does not leave the dark state during static or adiabatic interaction with the probe fields we write the Hamiltonian as $H_M \simeq \lambda_1 \langle \bar{D} \rangle$. As $|\bar{D}| \simeq |b_1\rangle |b_1\rangle \simeq 1$, it is convenient to exclude atomic degrees of freedom from the interaction picture and to rewrite the Hamiltonian in Heisenberg picture with quantized probe fields. The relation between Rabi frequencies of the probe fields and quantum operators describing the corresponding field mode can be written as

$$\hat{a}_i = \sqrt{\frac{2\pi \nu_i^2 \alpha_i}{\hbar}} \hat{\alpha}_i = \xi_i \hat{a}_i, \quad (4)$$

where $\nu_i$ is the dipole moment of the transition $|a_i\rangle \rightarrow |b_i\rangle$, $\nu_i$ is the field frequency, $\nu_i$ is the quantization volume of the mode, $\hat{\alpha}_i$ and $\hat{\alpha}^\dagger_i$ are the annihilation and creation operators. Then the Hamiltonian takes the final form

$$H_M = -\hbar \delta \left[ \frac{\xi_1^2 \xi_2^2}{|\Omega_1|^2 |\Omega_2|^2} \right] \hat{a}_1^\dagger \hat{a}_1 \hat{a}_2^\dagger \hat{a}_2. \quad (5)$$

We see that the nonlinear coupling between the probe fields increases with an increase in the detuning $\delta$.

To understand the size of the nonlinear interaction we recall the interaction Hamiltonian for an $N$ scheme [6,13]:

$$H_N = \hbar \frac{\xi_1^2 \xi_2^2}{\Delta |\Omega_1|^2} \hat{a}_1^\dagger \hat{a}_1 \hat{a}_2^\dagger \hat{a}_2. \quad (6)$$

The ratio of coupling constants in (5) and (6)

$$\mathcal{R} = \frac{\delta \Delta |\Omega_2|^2}{|\Omega_1|^2} \quad (7)$$

determines the relative strength of the nonlinear interaction introduced by the schemes. If $|\mathcal{R}| > 1$, then the $M$ scheme is more effective than $N$.

However, this comparison is not consistent unless we take into account the absorption of the probe field introduced by spontaneous emission. Simple calculations demonstrate that the ratio of the spontaneous emission probabilities coincide with the ratio of nonlinear susceptibilities for the $M$ and $N$ schemes. Another source of probe absorption is due to the decay of coherence between ground state levels which leads to depopulation of the dark state created in the system and to the absorption of the probe fields independently of each other. It can be shown that the probability of this process is the same for both $M$ and $N$ configurations.

In the usual cells containing gases of alkali atoms the one-photon transitions are Doppler-broadened due to the motion of the atoms. If the condition for EIT is fulfilled $(\Omega_1 \gg W_d \sqrt{\gamma_0/\gamma_1})$, where $W_d$ is the Doppler linewidth $W_d \gg \gamma_1$, then in the $M$-scheme the population of level $|b_2\rangle$ is approximately equal to $|\alpha_1|^2/|\Omega_1|^2$. The nonlinear interaction results from the refraction and absorption of the second probe field $\alpha_2$, coupled to the second drive field $\Omega_2$. Fields $\alpha_2$ and $\Omega_2$ along with levels $|b_2\rangle$, $|b_3\rangle$, and $|a_2\rangle$ create $\Lambda$ system. Almost all population is in level $|b_2\rangle$. Therefore,

$$\chi_M = -i \frac{3}{8\pi^2 N \lambda_3^3} \frac{\gamma_2 (\gamma_0 + i \delta)}{W_d + i \Delta |\Omega_1|^2} \frac{|\alpha_1|^2}{|\Omega_1|^2}, \quad (8)$$

where $\mathcal{N}$ is the atomic density and $\lambda_{\alpha_2}$ is the wavelength of the field $\alpha_2$.

Similar calculations allow us to derive the susceptibility for the field $\alpha_2$ for the Doppler broadened $N$ scheme:

$$\chi_N = -i \frac{3}{8\pi^2 N \lambda_3^3} \frac{\gamma_2}{W_d + i \Delta |\Omega_1|^2}, \quad (9)$$

It is easy to see that Eqs. (8) and (9) are interchangeable if $\gamma_0 \rightarrow 0$, and $\Delta \leftrightarrow \delta/|\Omega_2|^2$. Therefore, ultimately $M$ and $N$ interaction schemes are equally efficient, even though quite different mechanisms are responsible for the nonlinear susceptibility enhancement.

To experimentally demonstrate the enhancement of nonlinearity in the $M$ type level scheme we study the rotation of elliptical polarized light resonant with the $F = 2 \rightarrow F' = 1$ transition of the $^{87}$Rb $D_1$ line (Fig. 3a). We consider the light as two independent circular components $E_+$ and $E_-$ which generate a coherent superposition of the Zeeman sublevels (a dark state). To disturb this dark state we apply a longitudinal magnetic field $B$ which leads to a splitting $\delta \propto B$ of the Zeeman sublevels. This atomic transition consists of $\Lambda$ and $M$ level configurations (Fig. 3a and 3b).

The nonlinear properties of the $M$ level scheme result in significant modification of the nonlinear polarization rotation as a function of the light ellipticity. We write the field amplitudes as $|E_\pm|^2 = (1 \pm q)|E_0|^2/2$, where $|q| < 1$ characterizes the light ellipticity and find an expression for the rotation angle $\phi$ of the polarization ellipse for small magnetic field. The details of the calculation will be given elsewhere. The enhancement of rotation with regards to the isolated $\Lambda$ system is given by

$$\frac{\phi(B)}{\phi_\Lambda(B)} \approx 1 + \frac{2 + q^2}{2 - q^2} \quad (10)$$

We have measured the polarization rotation in a cell containing Rb vapor using the technique described in detail in [14]. The results are shown in Fig. 3. The experimental dependence looks slightly different from the theoretical one (Fig. 3 inset) because of the influence of the Doppler broadening and AC-stark shifts due to light coupling to off-resonant atomic sublevels. However, numerical simulations based on steady state solution of exact density matrix equations give a good agreement with the experiment.
The experimental results pertain to a semiclassical description where the field is classical. An interesting application of the Kerr nonlinearity with quantized field as given in Eq. (3) is the possible implementation of a quantum phase gate. A quantum phase gate, together with a one-bit unitary gate, form the basic building block for quantum computation [2]. The transformation properties of a quantum phase gate leave the two qubits unchanged when one or both input qubits are in the logic state 0 and introduces a phase $\eta$ only when both the qubits in the input states are 1. For input photon states $|0\rangle$ or $|1\rangle$ for the two qubits, a unitary operator of the form $Q_\eta = \exp(i\eta a_1^\dagger a_2)$ can lead to such a phase gate, i.e., $Q_\eta|0, 0\rangle = |0, 0\rangle$, $Q_\eta|0, 1\rangle = |0, 1\rangle$, $Q_\eta|1, 0\rangle = |1, 0\rangle$, and $Q_\eta|1, 1\rangle = \exp(i\eta)|0, 0\rangle$.

It is clear that such a phase gate can be realized via Hamiltonian $H_M$ with the time-evolution unitary operator $\exp(-iH_M t)$ and the corresponding phase $\eta = h\hbar\xi \xi^2 t/|\Omega|^2 |\Omega|^2 |\Omega|^2 |\Omega|^2$.

The resonant enhancement of $\chi^{(3)}$ and higher orders of nonlinearity may be achieved using additional $\Lambda$ sections connected to $M$ scheme, similar to the generalized $N$ scheme [3]. Such $\chi^{(3)}$ nonlinearity may be so high that three-photon phase gates become feasible.

In conclusion, we have proposed a realization of media with resonantly enhanced Kerr nonlinearity where one-photon resonant absorption is suppressed due to coherence effects. Such media have certain advantages over already existing schemes of coherent resonant nonlinearity enhancement and hold promise for the use in the creation of non-classical states of light and in the implementation of quantum computing algorithms.

The authors gratefully acknowledge the support from Air Force Research Laboratory (Rome, NY), DARPA-QuIST, the Office of Naval Research, and the TAMU Telecommunication and Informatics Task Force (TITF) initiative.

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FIG. 3. The slope of nonlinear magneto-optic rotation as a function of the ellipticity of the incident light. The slope is normalized by the value for linearly polarized light (zero ellipticity). Experimental data are shown by squares (1 mW laser power) and (2 mW laser power), while the results of the numerical simulations for the case of 2 mW laser power are shown by the solid line. The laser beam has 2 mm diameter. Inset: the theoretical dependence for naturally broadened Rb vapor, from Eq. [10].