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LIGHT-MATTER COUPLING FOR NONLINEAR OPTICS: A MICROSCOPIC PERSPECTIVE

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ABSTRACT

In this article, we focus on the observation of single-level nonlinear interactions, the binding of light-substances for nonlinear optics, and the use of weak nonlinear responses relative to the state of the microscopic perspective.

Keywords:laser, nonlinear, optical, light, microscopic, quantum.

INTRODUCTION

Typically, high-intensity lasers are required for nonlinear optical processes because the nonlinear optical cross-section for electromagnetic field coupling in matter is quite small and requires large field strengths [10] to observe deviations away from linear behavior. In other words, the nonlinear polarization current induced by an incident optical field is quite small compared to the linear displacement field in naturally occurring media. In the regime where the wavelength of light is of the order 10^{-6} m, a single atomic system (typical size, $\lesssim 10^{-10}$ m) can be approximated as a dipole, with the intra-atomic distance being between the positive nucleus and an electron in the relevant orbital. In the dipole approximation, the interaction energy between the electron and the electromagnetic field can be written as, $\hat{H}_1 = -\hat{\mu} \cdot \hat{E}$, where $\hat{\mu} = -e\hat{r}$ is the electric dipole moment operator and $\hat{r}$ is the position operator. In the rest of this section, we consider a semi-classical theory of the nonlinearity, where only the medium is treated quantum mechanically and the field is treated classically, i.e. a c-number represents the field instead of an operator. In the Schrodinger picture, the quantum state of the medium can be represented by a density matrix, p, that obeys the Liouville-von Neumann equation [10],

$$\hat{p} = \frac{-i}{h}[\hat{p}, \hat{H}] \tag{1.1}$$

where $\hat{H} = \hat{H}_0 + \hat{H}_I$, where $\hat{H}_0$ is the Hamiltonian of the unperturbed atomic system. However, due to the interaction with the environment, i.e. with neighboring dipoles and vacuum fluctuations, the quantum state evolution needs to describe decohering processes as well, which can be modeled as damping terms. Thus, Eq. 1.1 is transformed into,

$$\hat{p}_{nm} = -\frac{-i}{h}\left[\hat{p}, \hat{H}\right]_{nm} - \gamma_{nm}(p_{nm} - p_{nm}^{(e)}) \tag{1.2}$$

where the subscripts *n, m* are the row and column indices for the density matrix, $\hat{\mathrm{p}} = \sum_{\mathrm{nm}}\langle \mathrm{n}|\mathrm{p}|\mathrm{m}\rangle\langle \mathrm{m}|$, which is decomposed in energy eigenfunction basis for the medium (in the

absence of the interaction energy $\hat{H}_I$, i.e. $\hat{H}_0|n\rangle \backslash n) = E_n|n\rangle)$, γ_{nm} describe the damping rates for the various energy levels due to decoherence of the quantum state, and $\mathrm{p}_{nm}^{\mathrm{e}}$ is the equilibrium state of the material. In this formulation, under perturbation theory, writing $p_{nm} = \mathrm{p}_{nm}^{(0)} + \lambda \mathrm{p}_{nm}^{(1)} + \lambda^2 \mathrm{p}_{nm}^{(2)} \ldots$, we can solve for the various higher-order contributions independently using a Volterra series expansion. Without reproducing the derivation verbatim from [10], we will explicitly write the form of the second-order nonlinear correction term for the electric susceptibility. In this case, with the positive-frequency components of the applied electric field taking the form of $E(t) = \sum_q E(\omega_q) e^{-i\omega_q t}$, the lowest-order nonlinear term contributing to the susceptibility can be expressed as,

$$p_{nm}^{(2)} = e^{-(i\omega_{\mathrm{nm}}\mathrm{t} + \gamma_{\mathrm{nm}}\mathrm{t})} \int_{\infty}^{\mathrm{t}} \mathrm{dt}' \frac{-i}{h} \left[\hat{p}^{(2)}, \hat{H}_I\right]_{nm} e^{(i\omega_{\mathrm{nm}}\mathrm{t}' + \gamma_{\mathrm{nm}}\mathrm{t}')} =$$

$$= e^{-(i\omega_{\mathrm{nm}}\mathrm{t} + \gamma_{\mathrm{nm}}\mathrm{t})} \int_{\infty}^{\mathrm{t}} \mathrm{dt}' \frac{-i}{h} \sum (\mu p_{km}^{(1)} - p_{km}^{(1)} \mu) \cdot E(t) e^{(i\omega_{\mathrm{nm}}\mathrm{t}' + \gamma_{\mathrm{nm}}\mathrm{t}')} \qquad (1.3)$$

which upon expansion can be related to the second-order polarization of the medium as,

$$\hat{P}^{(2)}(\omega_{\mathrm{p}} + \omega_{\mathrm{q}}) = \mathrm{N}\langle \mu\omega_{\mathrm{p}} + \omega_{\mathrm{q}} \rangle = \mathrm{N} \sum_{\mathrm{nm}} p_{nm}\mu_{nm} \qquad (1.4)$$

where N is the number density of atoms, and each cartesian component of the polarization current can be written as,

$$\hat{P}_i^{(2)}(\omega_{\mathrm{p}} + \omega_{\mathrm{q}}) = \epsilon_0 \sum_{\mathrm{jk}} \sum_{(\mathrm{pq})} \mathrm{X}_{\mathrm{ijk}}^{(2)}(\omega_{\mathrm{p}} + \omega_{\mathrm{q}}, \omega_{\mathrm{q}}, \omega_{\mathrm{p}}) \mathrm{E}_{\mathrm{j}}(\omega_{\mathrm{q}}) \mathrm{E}_{\mathrm{k}}(\omega_{\mathrm{p}}) \qquad (1.5)$$

where the (pq)-index notation implies that $\omega_{\mathrm{p}} + \omega_{\mathrm{q}}$ is held fixed while ω_{p} and ω_{q} could be correspondingly independently changed. $\mathrm{X}_{\mathrm{ijk}}^{(2)}(\omega_{\mathrm{p}} + \omega_{\mathrm{q}}, \omega_{\mathrm{q}}, \omega_{\mathrm{p}})$ then has terms proportional, where l_{mn} are the same dummy indices that are iterated over in the definition of p. In this expression, we see that the nonlinear response can be made very large by (i) increasing the number density of dipoles per unit volume (N) and (ii) resonant pumping such that the real part of the denominator terms is nearly zero. Let us consider the latter case first: we see that under resonant enhancement of the nonlinear response the linear decay terms characterized by γ play a big role, and for our physical system, translate to photon loss in the medium. Since such loss is deleterious for most, if not all, quantum optical technologies, some nonlinear strength must be sacrificed in order to retain low loss operation. Similarly for a given material, the number density of atomic dipoles is fixed by its own structure. However, we can compensate for the loss in nonlinearity by increasing the field strength that contributes to the nonlinear polarization current in Eq. 1.5 while simultaneously reducing the optical mode volume so that N is maximized. In this thesis, we choose to do this by using a high-Q, small-mode-volume (V) optical dielectric cavity.

We note that the objective of this discussion was to highlight the microscopic contribution to nonlinear responses in dielectric media, while also, simultaneously showing that experiments that can utilize ensembles of atoms can provide large nonlinear responses in the off-resonance regime by enhancing the applied electric field in a cavity-geometry. The description herein also shows the deviation for the off-resonant regime from the resonant light-matter coupling typically used to achieve large optical nonlinearities at the single photon level because we do

not consider atom-atom interactions, and importantly, in the density matrix picture used for the medium, there is no physical population transfer in any of the atoms, i.e. the medium was at thermodynamic equilibrium [10]. This has meant that we consider only the bulk macroscopic response of the nonlinearity (see Chap. 3).

Quantum nature of the electromagnetic field

In the previous section, we looked at the nature of the optical nonlinearities that are available in bulk media. In this thesis, we are focused on using such relatively weak nonlinear responses to observe single-photon-level nonlinear interactions. Because the electric and magnetic fields are physical observables, in quantum theory, they are to be represented by operators ($\hat{E}(r,t) \rightarrow \hat{E}(r,t), \hat{H}(r,t) \rightarrow \hat{H}(r,t)$), which are accompanied by their commutation relations. The resulting consequences of commutativity is outside of the purview of classical electromagnetic field theory. One such consequence is the presence of the vacuum state, i.e. the $|0\rangle$ state, and its non-zero variance in energy. Owing to experimental observations such as that of the Lamb shift [12], the interaction between the vacuum fluctuations of the electromagnetic field and a two-level system, which also results in spontaneous emission, was widely believed as the empirical proof for the quantum nature of light. However, it was found that spontaneous emission in vacuum from a two-level system could be explained [13] without invoking explicitly the quantum features of the electromagnetic field. However, the correlation functions for the fluorescence from such an interaction would directly invoke the quantum features, and therefore, are a better test for the "quantumness" of the field. It is in this vein that we interpret the quantum features of the field in this work. Given a source field $\hat{E}(r,t)$, the two-time normalized auto-correlation function,

$$g^{(2)}(r,r';t,t') = \frac{\langle :E^{(-)}(r,t)E^{(-)}(r',t')E^{(+)}(r,t)E^{(+)}(r',t'):\rangle}{\langle E^{(-)}(r,t)E^{(+)}(r,t)\rangle\langle E^{(-)}(r',t')E^{(+)}(r',t'):\rangle} \qquad (1.6)$$

where $\hat{E}^{(+)}(r,t)$ and $\hat{E}^{(-)}(r,t)$ are the annihilation and creation operators for the electromagnetic field, is used as the metric for determining the quantum behavior of a field and $: \cdots :$ denotes normal ordering [14]. In the case of a stationary field, the correlation function $g^{(2)}$ is only dependent on the time and spatial *differences* rather than absolute positions. Moreover, we can suppress the spatial arguments as the two field measurements will happen occur within a light-cone and will be made by detectors coupled to singlemode fibers. Thus, with this definition, $g^{(2)} = (\tau = t - t' = 0)$ has a few salient properties: (i) for coherent states, $g^{(2)} = 1$, and signifies Poissonian statistics for the field-correlations (ii) $g^{(2)} > 1$ denotes super-Poissonian statistics with the special case of $g^{(2)} = 2$ for thermal beams of light, and finally, (iii) $g^{(2)} < 0$ denotes sub-Poissonian statistics, and is a unique consequence of the quantization of the field [15]. It is in this final area that we are interested in. In the special case of a single photon Fock state in one mode, $g^{(2)} = 0$. For quantum information processing applications, it is important to consider the photonic quantum state transport through the active and passive devices that will be present in the optical circuitry. In this thesis, we study nonlinear interactions between single-photon-level quantum states to address the feasibility of large-scale QIP.

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