Identification of Intermediate Levels in Quantum Spectroscopy

Motivation

Spectroscopic methods represent pivotal tools in the determination of an unknown chemical material. Among various approaches, the quantum state spectroscopy [1,2] offers the possibility of resolving intermediate levels in the process of two-photon absorption. Recent advances in both theoretical and experimental domains in the generation of intense entangled photon beams (twin beams) in the nonlinear process of spontaneous parametric down-conversion (SPDC) [3] enable to boost the atom-light interaction meanwhile quantum features of light persist.

Theoretical Introduction

We considered a simple atom with transitions between the ground state \( |g \rangle \), three intermediate states \( |i \rangle \), and the final state \( f \). The energy of the final state is chosen such that \( E_f - E_g = \hbar c/\lambda_{ph} \), where \( \lambda_{ph} \) is the central wavelength of the pump pulse with time duration \( \tau_p \), and power \( P_p \), that interacts with a nonlinear crystal of length \( L \). Photon pairs are generated in the SPDC process in a nonlinear crystal with the central wavelengths \( \lambda_{ph} = \lambda_0 = 2\lambda_{ph} \). Among generated photons a time-delay \( \tau \) between them is introduced. Information about inter. transitions is then obtained by monitoring the two-photon absorption rate \( \sigma(\tau) \) as a function of delay [4]. The studied experimental arrangement is shown in Figure below. The phase modulation is inserted in the signal photon path [5].

Detected Spectroscopic Signal

Detected TPA signal:

\[
S_{\text{TPE}}(\omega) = \frac{1}{h^2} \int_{-\infty}^{\infty} dt_{2} \int_{t_{2}}^{t_{1}} dt_{1} \int_{-\infty}^{t_{1}} dt'_{2} \int_{t'_{2}}^{t'_{1}} dt'_{1} \hat{M}^{*}(t_{2}, t_{1})
\times M(t'_{2}, t'_{1}) \left( \hat{E}_{\text{TPE}}(t_{2}) \hat{E}_{\text{TPE}}(t_{1}) \hat{E}_{\text{TPE}}(t'_{2}) \hat{E}_{\text{TPE}}(t'_{1}) \right)
\]

Electric field operator:

\[
\hat{E}_{\text{TPE}}(t) = \hat{E}_{\text{ph}}(t) + \hat{E}_{\text{TPE}}(t)
\]

Response function of matter:

\[
M(t_{2}, t_{1}) = \sum_{k} \mu_{ik} \mu_{kg} \epsilon_{\omega}[(\hat{E}_{\text{TPE}} - \hat{E}_{k})_{t_{2}}(\hat{E}_{\text{TPE}} - \hat{E}_{k})_{t_{1}}]
\]

Relative Variance

\[
R_{\text{\(\hat{E}\)}} = \frac{\variance(\hat{E})}{\mean(\hat{E})^2} = \frac{\sum_{\omega} \hat{E}^{*}(\omega) \hat{E}(\omega)}{\sum_{\omega} \hat{E}^{*}(\omega) \hat{E}(\omega)}
\]

Numerical Results

(a) TPA transition signal as a function of the delay \( \tau \) between fields carrying \( N_2 \approx 100 \) photons. (b) Fourier transform of the TPA transition probability shown in (a). (c) Relative variance of transition-probability fluctuations of the peaks resolved in the TPA spectra obtained for 20 different values of the signal-frequency chirp \( \xi \in (0.0, 9.5) f_{\text{ph}} \) (blue circles) and 40 values \( \xi \in (0.0, 3.9) f_{\text{ph}} \) (red triangles). (d) Spectrum of the TPA transition probability averaged over an ensemble of 100 crystals of different lengths. The vertical grey lines indicate the doubled relative \( \{2\epsilon_{ij} - \epsilon_{fj}\} \) energies of the intermediate levels.

Robustness Analysis

Relative variances originating from (a-c) 2-intermediate levels and (d-f) 3 intermediate levels considering 20 steps of the chirp parameter \( \xi \).

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