Identification of Intermediate Levels in Quantum Spectroscopy

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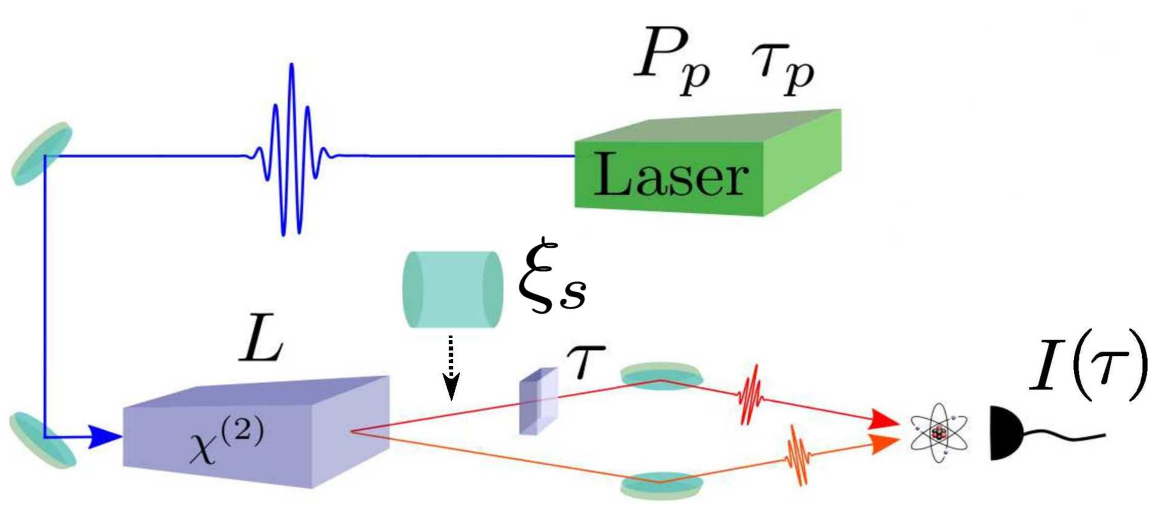
Motivation

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 $|k\rangle$, and the final state $|f\rangle$. The energy of the final state is chosen such that $E_f - E_g = hc/\lambda_{p0}$, where λ_{p0} is the central wavelength of the pump pulse with time duration τ_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 $\lambda_{s0} = \lambda_{i0} = 2\lambda_{p0}$. Among generated photons a time-delay τ 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:

$$S_{g\to f} = \frac{1}{\hbar^4}\int_{-\infty}^{\infty} dt_2 \int_{-\infty}^{t_2} dt_1 \int_{-\infty}^{\infty} dt_2' \int_{-\infty}^{t_2'} dt_1' M^*(t_2,t_1) \times M(t_2',t_1')\,\langle \hat{E}^{(-)}(t_2)\,\hat{E}^{(-)}(t_1)\,\hat{E}^{(+)}(t_2')\,\hat{E}^{(+)}(t_1')\rangle,$$

Electric field operator:

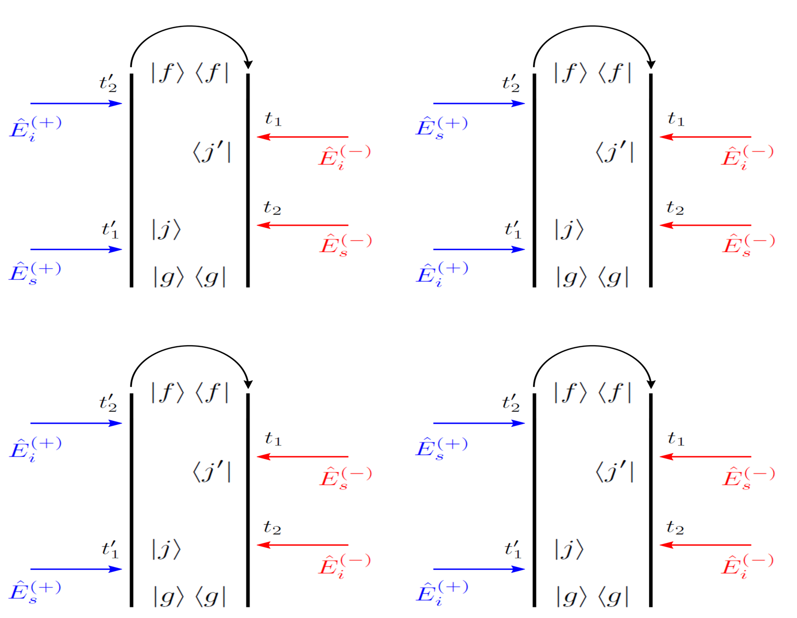
$$\hat{E}^{(+)}(t) = \hat{E}_s^{(+)}(t) + \hat{E}_i^{(+)}(t)$$

Response function of matter:

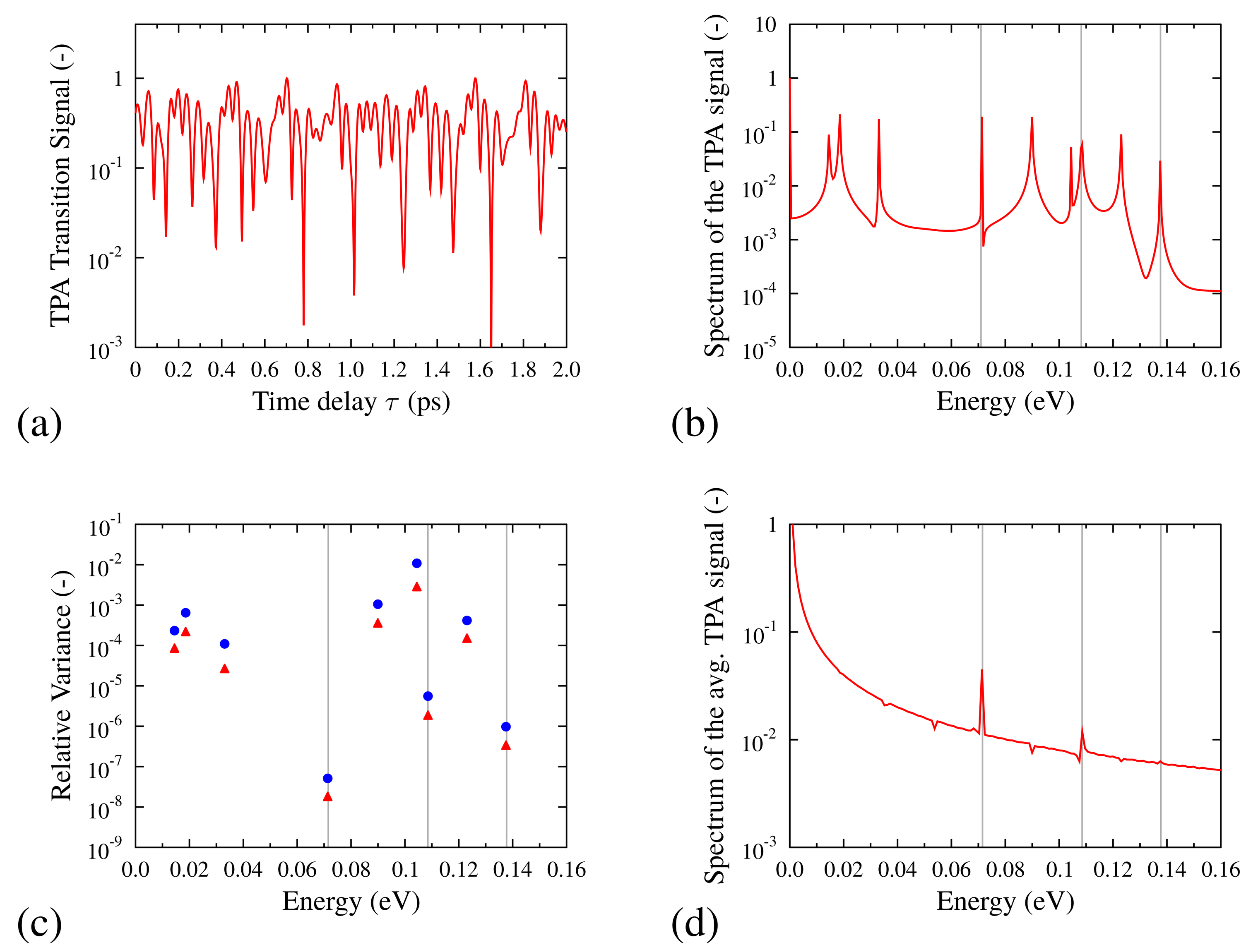
$$M(t_2,t_1) = \sum_k \mu_{fk}\mu_{kg} e^{[i(E_f-E_k)t_2 + i(E_k-E_g)t_1]}$$

Relative Variance

$$R_n = \frac{var_{\xi_s}[P_n]}{M_{\xi_s}[P_s]^2}$$

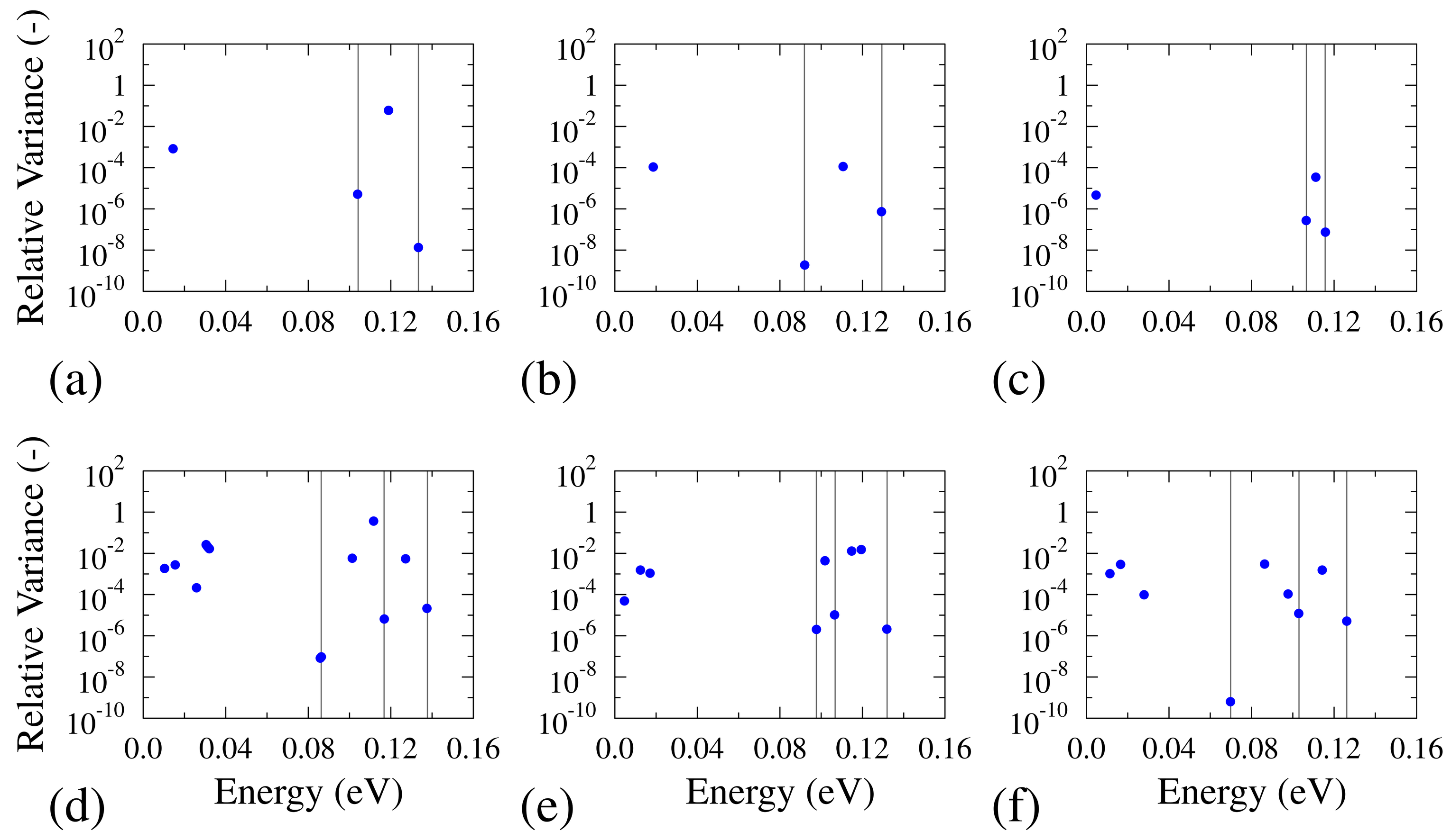


Numerical Results



(a) TPA transition signal as a function of the delay τ between fields carrying $N_j \approx 100$ 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 $\xi_s \in \langle 0.0, 9.5\rangle$ fs^2 (blue circles) and 40 values $\xi_s \in \langle 0.0, 3.9\rangle$ fs^2 (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 $|2\epsilon_j - \epsilon_f|$ 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 ξ_s.

References and Acknowledgments

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