Motivation

Spectroscopic methods represent pivotal tools in the determination of an unknown chemical material. Among various approaches, the virtual 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 $|f\rangle$. The energy of the final state is chosen such that $E_f - E_g = h\nu_0$, where $\nu_0$ 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 $\nu_0 = \lambda_0/2$. Among generated photons a time-delay $\tau$ between them is introduced. Information about virtual transitions is then obtained by monitoring the two-photon absorption rate $\sigma(\tau)$ as a function of delay. The studied experimental arrangement is shown in Figure below.

Detected Spectroscopic Signal

Detected TPA signal:

\[
S_{k-m} = \frac{1}{h} \int \int \int dt_2 \int dt_1 \int \int \int dt'_2 \int dt'_1 M^* (t_2, t_1) 
\times M (t'_2, t'_1) |\hat{E}^{(-)} (t_2) \hat{E}^{(-)} (t_1) \hat{E}^{(+)} (t'_2) \hat{E}^{(+)} (t'_1)|^2,
\]

Electric field operator:

\[
\hat{E}^{(+)} (t) = \hat{E}^{(+)} (t) + \hat{E}^{(-)} (t)
\]

Response function of matter:

\[
M(t_2, t_1) = \sum_k \mu_{k2} \mu_{k1} \langle \hat{E}(t_1 - \varepsilon_k) \hat{E}(t_1 + \varepsilon_k) \rangle \tau_2 \tau_1
\]

Numerical results

1) Intense entangled beams allow to increase the detection probability, provided that the intensity is below a critical value.
2) This paws a way to a practical experimental implementations.

Conclusion

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