Witnessing Entangled Two-Photon Absorption via Quantum Interferometry

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Abstract: We demonstrate that a two-photon, N00N-state interferometer is insensitive to linear (single-photon) losses, and thus capable to unequivocally certify entangled two-photon absorption (eTPA) in arbitrary-sample transmission-based absorption measurements. © 2023 The Author(s)

1. Introduction

Recent investigations suggest that the use of non-classical states of light, such as entangled photon pairs, may open new and exciting avenues in experimental two-photon absorption spectroscopy. Despite several experimental studies of entangled two-photon absorption (eTPA), there is still a heated debate on whether eTPA has truly been observed. This interesting debate has arisen, mainly because it has been recently argued that single-photon-loss mechanisms, such as scattering [1] or hot-band absorption [2] may mimic the expected entangled-photon linear absorption behavior. Consequently, a large group of physicists, chemists and biologists has devoted considerable effort to developing novel experimental schemes and metrics for certifying true eTPA. Some authors have relied on the linear to quadratic transition in the two-photon absorption rate as a function of the incident photon flux [3]. Others have proposed new metrics based on single and two-photon coincidence measurements, which make use of Hong-Ou-Mandel (HOM)-like interferometers [4]. In this work we explore, in the context of eTPA, a number of two-photon quantum interferometry systems, leading to our demonstration that amongst them the so-called N00N-state configuration is the only one insensitive to single-photon losses. Our results show that, in transmission eTPA measurements, this N00N-state configuration is an ideal candidate for the true certification of entangled two-photon absorption. More importantly, and in contrast to other quantum-technology schemes in which N00N states are not typically robust [5], our findings show that N00N states could play an important role in quantum spectroscopy.

2. Results

Since its conception in the 1990s, eTPA has been described as a process in which correlated photon pairs satisfying the so-called two-photon resonance condition [6] are lost in order to drive a two-photon excitation of the absorbing medium [see Fig. 1(a)]. This means that the sample effectively acts as a filter that removes specific two-photon states. While the so-called two-photon resonance condition is lost in order to drive a two-photon excitation of the absorbing medium [see Fig. 1(a)], the so-called two-photon resonance condition [6] is lost in order to drive a two-photon excitation of the absorbing medium [see Fig. 1(a)]. This means that the sample effectively acts as a filter that removes specific two-photon states. While

\[
S(v_s, v_i) = |\Phi(v_s, v_i)|^2 = |f_{TP}(v_s, v_i)\Phi(v_s, v_i)|^2, \tag{1}
\]

where \(v_j = \omega_j - \omega_0 \) (\(j = s, i\)) are the frequency deviations from the photons’ degenerate central frequency, \(\omega_0\). \(\Phi(v_s, v_i)\) and \(\Phi(v_s, v_i)\) describe the joint amplitude of the photons before and after the interaction with the sample, respectively. The two-photon filter can readily be defined as

\[
f_{TP}(v_s, v_i) = 1 - \exp \left[-(v_s + v_i)^2 / (2\sigma_{TP}^2)\right],
\]

with \(\sigma_{TP}\) describing the filter bandwidth. As discussed above, in real experiments, during the light-matter interaction the two-photon beam might experience single-photon losses that remove, independently, signal or...
idler photons. These losses can be accounted for by writing a single-photon filter of the form 
\[ f_{s,i}(\nu_s, \nu_i) = 1 - \exp \left[ -\left( \nu_{s,j} - \nu_{s,i} \right)^2 / \left( 2\sigma_{s,i}^2 \right) \right] \]. Here \( \nu_{s,j} \) describe the central frequency deviations of the filter, whereas \( \sigma_{s,i} \) represent the single-photon filter bandwidth for the signal and idler modes, respectively.

In order to find a good candidate for eTPA certification, we study and compare the coincidence-rate measurement of three distinct two-photon quantum interferometers, namely single-port and two-port Hong-Ou-Mandel setups, as well as a NOO0N-state configuration. The three configurations are shown schematically in Figs. 1(b)-(d), respectively. Figures 1(e)-(g) show the coincidence rate as a function of the delay \( \tau \) for the three configurations, namely (e) single-port, (f) two-port, and (g) NOO0N interferometers, respectively. Note that the no-filter (blue solid line) and eTPA (dashed red line) curves are fully overlapped, with the linear losses curve (dash-dotted yellow line) nearly overlapped with the other two, implying that one would be unable to determine the presence or absence of an eTPA sample from a transmission-based measurement. In striking contrast, for the NOO0N-state configuration, while the no-filter and linear-filter curves are fully overlapped, the eTPA curve clearly deviates from the other two. This result suggests that not only the NOO0N-state configuration is insensitive to linear losses but it is also capable of resolving eTPA. These unique feature makes such a configuration a strong candidate for effectively certifying the absorption of correlated photon pairs in an arbitrary sample. Given the simplicity of the NOO0N-state configuration, and in contrast to other schemes for quantum technologies in which NOO0N states are not typically robust [5], we expect them to play an important role in transmission-based quantum spectroscopy.

References
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