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## Manuscript submission to Nature Photonics

Dear Editor,

please find enclosed our manuscript on "Time-resolved Scattering of a Single Photon by a Single Atom", where we report on a measurement of the absorption dynamics of heralded optical photons by a single atom in a free-space geometry.

This, to our knowledge, is the first time the scattering dynamics is observed for single photons with different temporal envelopes. In particular, we tested the time symmetry of the model of spontaneous emission introduced by Weisskopf and Wigner in 1934. Our work complements the large number of experiments in this direction where the single photon was contained in a cavity with its discrete mode spectrum. In our case, we do not use an optical cavity, and therefore consider the full continuous mode spectrum of the electromagnetic field.

We are able to shape the temporal envelope of a single photon state of resonant light, send it onto a single atom, and measure the scattering dynamics with high temporal resolution. In agreement with a theoretical prediction, we do find that the excitation probability of an atom is higher for an exponentially rising single photon as compared to an exponentially decaying photon with the same spectral distribution. We reach a maximal excitation probability of about 3%, which is close to the theoretical maximum we expect for the spatial mode of the single photon impinging on the single atom.

We feel that this work could be interesting to the audience of Nature Photonics because it demonstrates, in an almost textbook-like manner, the elementary absorption process of photons by atoms in free space. It also contributes to the longstanding debate if heralded single photons have the same physical action as "true" single photons - which we can confirm with our experiment, involving a single atom as a measurement tool as compared to photodetectors with constraints on what they are able to observe.

With Best Regards on behalf of all authors,

Christian Kurtsiefer

Centre for Quantum Technologies