Demonstration of quantum interference metrology with amplified ultrashort laser pulses

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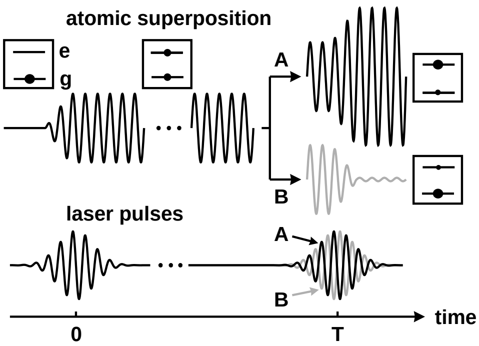
The measurement of the absolute frequency of light is a subject of great interest in many areas of science, as precise frequency control over light allows for detailed studies of the interaction between light and matter. The ongoing improvements in laser spectroscopy has made frequencies the most accurately measurable quantity in nature, and the invention of the femtosecond frequency comb has simplified such metrology experiments considerably. However, high-precision spectroscopy still requires ultrastable continuous-wave (CW) lasers, which do not exist in e.g. the extreme ultraviolet (XUV) and soft x-ray spectral regions.

We demonstrate that amplified frequency comb laser pulses can be used to measure optical transistion frequencies with high accuracy without the need for narrowband CW lasers. The method is similar to Ramsey spectroscopy, but uses pulses seperated in time rather than in space to probe the evolution of an atomic superposition (see figure). As this approach combines high peak power with high precision, it seems particularly promising for precision measurements that require vacuum-ultraviolet or shorter wavelengths. Interesting candidates are the precise determination of the ground state Lamb shift in helium and nuclear size effects of helium and hydrogen-like ions.



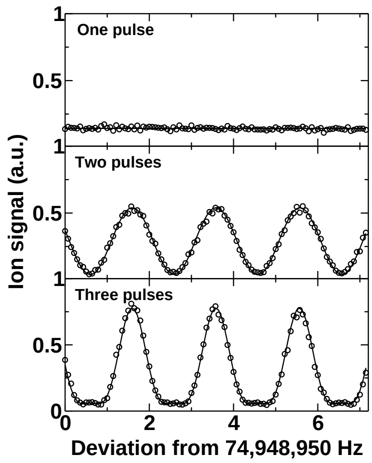


Figure 1: Left: the principle of quantum interference metrology. An atom is brought in a superposition of ground state and excited state by the first pulse, and depending on the phase difference between the second pulse and the atomic superposition, the excited state probability is either enhanced or suppressed when a second pulse interacts with the atom. Right: experimental krypton quantum interference fringes for one, two and three phase-locked pulses, as a function of the pulse repetition rate.

The experiment is performed on the $4p^6 \rightarrow 4p^5 5p[1/2]_0$ two-photon transition in krypton at a wavelength of $2 \times$ 212.55 nm. The required phase-locked pulses (13.3 ns apart) are obtained by amplification of pulses at 850 nm from a stabilized frequency comb laser in a multi-pass Ti:sapphire amplifier. Light at 212.55 nm is then generated by frequency-conversion in two consecutive BBO crystals. The UV pulses, together with a 532-nm ionization pulse, are then focused in a collimated atomic beam of krypton. Scanning the time delay between the pulses leads to interference fringes with a very high contrast (see figure). From the measurements and carefull investigation of all systematic effects, we have determined the isotope shifts with $\sim$ 200 kHz accuracy, while the absolute transition frequency has been measured with 3.5 MHz precision [1]. These results are an order of magnitude more accurate than previous work using nanosecond laser pulse excitation [2].

[1] S. Witte et al., accepted for publication in Science, DOI 10.1126/science.1106612
[2] F. Brandi et al., *J. Phys. B* **35**, 1071 (2002)