

Quantum electronics enabled by high-field physics

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Abstract: Observation of lasing signals at multiple wavelengths in argon atoms pumped by a 3-photon resonance with an intense 260 nm laser were used to observe the dynamics of electronic excited states. © 2023 The Author(s)

1. Introduction

Strong field ionization of noble gases, by definition, involves creation of free electrons by multi-photon, tunnelling or barrier suppression ionization when ultra-short, high-intensity laser pulses are used. It is a highly nonlinear interaction where, in the multi-photon regime, the resulting plasma density increases as I^n , where I is the laser intensity and n is the minimum number of photons required to overcome the ionization potential. Quantum electronics on the other hand typically involves the physics of transitions or coupling between two or more bound states. And yet when one considers lasing in a minority constituent of air such as Argon (Ar partial pressure 7 torr or 1%) there is a strong connection between these two effects if the ionizing pump laser has one or band of multi-photon resonance(s) close to the ionization potential that, despite relatively short lifetime(s), can act as a reservoir to initiate a lasing cascade to the lower lying states. Our previous experimental work [1] on measuring strong-field ionization of noble gases has led to the observation of cascaded lasing, which can be used to investigate the physics of transitions and coupling between excited atomic states.

2. Experimental results

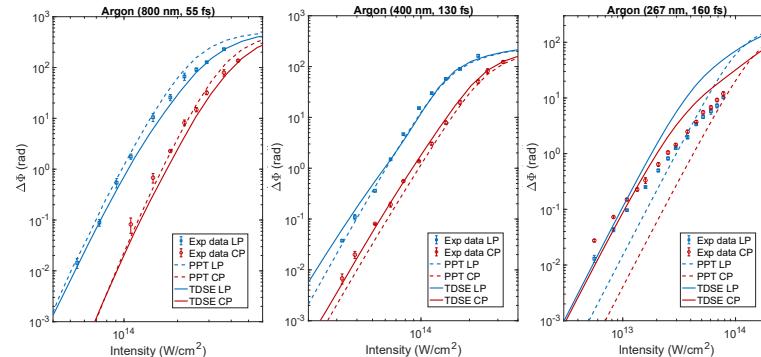


Figure 1: Ionization yield of Ar gas by laser pulses of different polarizations and wavelengths. Dashed curves are PPT theory [3], [4], solid curves are TDSE simulations [5], and experimental data were measured using the interferometry technique from Ref. [2]. Measured phase change is proportional to fraction of ionized electrons. Theoretical phase change was calculated from theoretical ionization rates.

Interferometry measurements [2] were used to measure strong field ionization rates as a function of laser intensity, polarization, and wavelength, as shown in Figure 1. While the experimental results showed good agreement with theory for 800 nm and 400 nm pump wavelengths, the measured ionization yields using a 267 nm pump deviated significantly from the analytical PPT theory [3], [4], as well as from the *ab initio* simulations using

the time-dependent Schrödinger equation (TDSE) [5]. The enhanced yield predicted by TDSE simulations compared to PPT theory indicates the presence of a resonance, which is not included in the analytical theory. However, even including the effects of the excited states on the refractive index did not lead to a good agreement with theory. This suggested the presence of a more complicated process involving many atoms coupled, e.g., via the dipole-dipole interaction, which is not captured by our single-atom TDSE simulation.

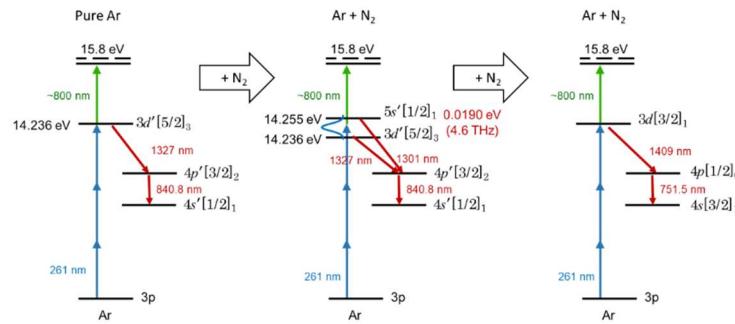


Figure 2: Cascaded electronic transitions observed in argon enabled by 3-photon resonance with 261 nm pump laser. As more N₂ is added to argon, different lasing wavelengths are observed, which all correspond to known transitions in the argon atom.

One such process is lasing on multiple transitions between argon excited states, as shown in Figure 2. Bidirectional lasing can be observed both as cascade superfluorescence or as amplification of an injected signal pulse. When the injected pulse follows the pump pulse closely (within a few ps), the injected pulse can further excite the electrons from the three-photon resonance band to higher-lying Rydberg states (resulting in lasing from these Rydberg states in proper conditions) thereby affecting the gain of the two lasing cascades; when the injected pulse delays longer time (10s of ps), a small frequency component of this pulse can be amplified by the second (cascaded) lasing step. We find that the quantum beating between two-levels involved in the initial three photon resonance is reflected in the temporal modulation of the superfluorescence output and that the frequencies of the amplified radiation can be controlled by varying the pressure of the most abundant gas nitrogen found in air but not by adding He which simply acts as a buffer gas. Last but not least, using atmospheric air we can reproduce these results showing that other air constituents, oxygen and CO₂ do not play a role in this Ar lasing in atmospheric air.

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