Two and three-photon double ionization of lithium

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Synopsis. Differential studies on two and three-photon double ionization (DI) of atomic lithium have been performed for photon energies of 50 eV and 59 eV at I ≈ 5 × 1012 W/cm2. At 50 eV DI proceeds via the simultaneous absorption of two photons resulting in the correlated emission of two electrons. In contrast for a photon energy of 59 eV, the resonant one photon transition (1s → 2p) is followed by DI through a second photon. Additionally a competing sequential three photon reaction is observed for both energies. The experimental data is compared with results from non-perturbative close-coupling calculations.

The advent of free electron lasers, as a source of intense and short VUV laser pulses, paved the way to investigate new regimes in light-matter interaction. In these, the most fundamental reactions are constituted by the interaction of few-photons with few-electrons, resulting in multiple ionization of the parent atom or molecule.

Pioneering differential experiments performed at the free electron laser in Hamburg (FLASH) on two-photon double ionization (TPDI) of the closed shell atoms helium and neon [1, 2], could already identify the direct (non-sequential), sequential and virtual sequential removal of the electrons. The results presented here aim to explore how two or three photons interact with electrons in different shells.

Therefore, a cold and dense lithium target, confined in a magneto-optical trap (MOT) combined with a many-particle momentum spectrometer (reaction microscope) was set up at FLASH. Recoil ion momentum distributions, giving insight into the electrons' sum momentum and mutual emission angle, have been recorded.

In case of the non-sequential reaction (at En ≈ 50 eV) no intermediate real state is reached and thus the equation for the two-photon process reads:

\[
\text{Li}(1s^22s) + 2 \cdot \gamma \rightarrow \text{Li}^{2+}(1s) + 2e^- . \quad (1)
\]

The resultant Li2+ momentum distribution shown Fig. 1(a), is diamond shaped and centered at zero momentum. The shape and intensity of the recoil ion cross section indicates the strongly correlated motion of the two outgoing electrons. If the photon energy is tuned to the (1s → 2p) transition in lithium, significant changes in the cross section pattern (Fig. 1(b)) are observed. Now DI proceeds via an intermediate state:

\[
\text{Li} + \gamma \rightarrow \text{Li}^+ (1s2e2p) + \gamma \rightarrow \text{Li}^{2+} (1s) + 2e^-. \quad (2)
\]

Here, the second step resembles photo double ionization (PDI) of the doubly excited state. The cross section exhibits a dumbbell like structure with two main peaks oriented along the laser polarization axis ε and a plateau region between them. For both experiments a competing three-photon reaction yields recoil ions with larger momenta.

The results are discussed and compared to two state-of-the-art calculations. These apply the convergent close coupling (CCC) [3] and time-dependent close coupling (TDCC) [4] approach.

![Figure 1. Li2+ momentum distributions as observed for two and three photon absorption at energies of 50 eV (a) and 59 eV (b).](image)

References


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