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A MANY-BODY APPROACH TO TWO- AND THREE-PHOTON DOUBLE IONISATION AND EXCITATION OF XENON

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Résumé: Nous étudions les différents processus responsables de l'ionisation double du xénon par absorption de deux ou trois photons d'énergie comprise entre 17 et 21 eV. Nous calculons les sections efficaces d'ionisation simple et double en utilisant la théorie des perturbations à plusieurs corps et nous comparons l'efficacité des mécanismes direct et séquentiel conduisant à l'ionisation double.

Abstract: We study the different processes responsible for the double ionisation of xenon through the absorption of two or three photons with energy between 17 and 21 eV. We calculate the single and double ionisation cross-sections using many-body perturbation theory and we compare the efficiency of the direct and sequential double ionisation mechanisms.

A basic question raised by the multiphoton multiple ionisation experiments[1-3] is the mechanism responsible for the multiple electron ejection. Are the electrons emitted in a one-step direct process? or are they emitted one at a time, in a sequential (or stepwise) process? Another important question is whether ions can be produced into excited states.

The purpose of the present work[4] is to study the competition between direct and sequential double ionisation of xenon in a simple case: the photon energy is chosen so that the direct process requires the absorption of two photons and the sequential channels, three photons. Figure 1 presents the different double ionisation processes studied in this work: 2-photon direct double ionisation (Fig.1a); (1+2)-photon sequential double ionisation via Xe+ in its ground state (Fig.1b); finally, (2+1)-photon sequential double ionisation via Xe+ in an excited state (Fig.1c).
Figure 1: Schematic representation of different processes leading to double ionisation (see text).

We calculate the ionisation cross-sections involved using many-body perturbation theory and keeping to lowest non-vanishing order in the radiation field. The calculation of one-photon or two-photon single ionisation cross-sections has been described previously [4,5]. Polarisation effects are described within the random phase approximation with exchange (RPAE). We use screened dipole interactions which replace the usual dipole interactions in the one-electron expressions. We neglect the spin-orbit interaction and for $Xe^+$ open-shell effects.

The calculation of two-photon double ionisation and two-photon ionisation and excitation cross-sections is done in a similar way. We only consider the simple process where two electrons absorb independently a photon. We neglect correlation effects (e.g. ground-state correlation, shake-off) which lead to double ionisation in one-photon absorption. The amplitude is then given by

$$t = \frac{\langle \epsilon_2 \mid r(\epsilon, \omega) \mid \epsilon_1 \rangle \langle \epsilon_1 \mid r(\omega) \mid i \rangle}{\epsilon_1 - \epsilon_1 - \omega} + \frac{\langle \epsilon_1 \mid r(\epsilon, \omega) \mid \epsilon_2 \rangle \langle \epsilon_2 \mid r(\omega) \mid j \rangle}{\epsilon_2 - \epsilon_j - \omega}$$

$i, \epsilon_i, \epsilon_1, \epsilon_2$ denote one-electron Hartree-Fock (HF) energies for the initial and final states respectively. $\omega$ is the photon energy. $r(\omega)$ is the RPA-screened dipole operator [4,5]. For the second photon absorption, we introduce a screened interaction $r(\epsilon, \omega)$ depending on the excitation state of the system. Finally, final state correlation effects are approximately taken into account by using two basis sets, $vN-1$ and $vN-2$ HF. The electron energy distribution is represented in Fig.2 for different photon energies from 1.3 Ry up to 1.8 Ry. It remains finite as long as the photon energy is lower than the ionisation energy (1.57 Ry). Above this energy value, sequential double ionisation with two photons becomes possible [6,7] and the energy distribution is centered around two peaks at fixed energies.
Figure 2: Distributions of the photoelectrons emitted in a two-photon double ionisation process as a function of the energy difference (in Ry) for different photon energies: (-.--) 1.3 Ry; (.-.-) 1.4 Ry; (...) 1.5 Ry; (---) 1.6 Ry; (——) 1.8 Ry.

The direct double ionisation cross-section, i.e. the area delimited by the electron energy distribution (see Fig.2, ω < 1.57 Ry) is plotted in dot-dashed line in Fig.3. It first increases rather steeply at threshold, then reaches a nearly flat plateau (about $10^{-49}$ cm$^4$s) and finally, increases again before 1.57 Ry. In the threshold region, the cross-section varies about linearly with the total kinetic energy and the electron energy distribution is nearly uniform (see Fig.2, double-dot dashed line), in agreement with the predictions from Wannier theory [8]. As the photon energy increases, the electron energy distribution becomes more and more peaked. At high photon energy, direct double ionisation is nearly a sequential process, in the sense that one electron escapes very rapidly, with most of the energy; the second electron escapes much more slowly, with a nearly zero kinetic energy.

In Fig.3, we have also plotted the cross-sections for two-photon ionisation into the 6s, 5d and 6d excited ionic states. These processes are resonant when the photon energy is equal to the excitation energy of the ion (ionisation with excitation is then a two-step process: ionisation followed by a discrete excitation). At low photon energy, ionisation with excitation into d states dominates over double ionisation [9].
Figure 3: Direct double ionisation cross-section of Xe(DDI) (-.-); ionisation into an excited Xe\(^{+}\) state (-----) 6s, (----) 5d and (-----) 6d.

Finally, we compare the relative efficiencies of the double ionisation mechanisms represented in Fig.1. We develop a simple rate equation model describing the evolution of the atomic and ionic populations. We use the single and double ionisation cross-sections previously calculated and a realistic laser temporal distribution. Except for high laser intensities and very short pulse durations (in the femto-second range), double ionisation of Xe is essentially a sequential process via the ion in its ground state. The reason is that the probability for one-photon single ionisation remains much more important than the probability for any two-photon (single or double) ionisation process. The Xe population is then extremely rapidly depleted due to one-photon ionisation.

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