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Sequential double photodetachment of He⁻ in elliptically polarized laser fields

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Four-photon double detachment of the helium negative ion is investigated experimentally and theoretically for photon energies where the transient helium atom is in the 1s2s ${}^{3}S$ or 1s2p ${}^{3}P^{\circ}$ states, which subsequently ionize by absorption of 3 photons. Ionization is enhanced by intermediate resonances, giving rise to series of peaks in the He⁺ spectrum, which we study in detail. The He⁺ yield is measured in the wavelength ranges from 530 nm to 560 nm and from 685 nm to 730 nm and for various polarizations of the laser light. Double detachment is treated theoretically as a sequential process, within the framework of *R*-matrix theory for the first step and effective Hamiltonian theory for the second step. Experimental conditions are accurately modeled, and the measured and simulated yields are in good qualitative and, in some cases, quantitative agreement. Resonances in the double detachment spectra can be attributed to well defined Rydberg states of the transient atom. The double detachment yield exhibits a strong dependence on the laser polarization which can be related to the magnetic quantum number of the intermediate atomic state. We also investigate the possibility of non-sequential double detachment with a two-color experiment but observe no evidence for it.

I. INTRODUCTION

⁷ Double photodetachment, the process in which a neg-⁸ ative ion absorbs one or several photons and ejects ⁹ two electrons, has been much less studied than double ¹⁰ photoionization, its counterpart for atoms and positive ¹¹ ions [1]. While the important structural differences be-¹² tween anions and atoms are expected to alter the dynam-¹³ ics of double electron ejection, experimental investigation ¹⁴ however has been hampered by the difficulty of producing ¹⁵ anions in sufficiently high density.

Early work on double photodetachment involved mod-16 erately intense fields ($\sim 10^{10} \text{ W} \cdot \text{cm}^{-2}$) and aimed at ei-17 ther studying excess photon detachment, *i.e.* the absorp-18 tion by the system of more photons than is energetically 19 required, or at performing spectroscopy of autoionizing 20 states embedded in the continuum [2, 3]. The detection 21 of positive ions following photodetachment is also at the 22 23 basis of resonant ionization spectroscopy, although in this 24 case the atom is optically excited to a Rydberg state and ionized by a static electric field [4]. In these studies, the 25 production of positive ions is a means to study single 26 photodetachment of the negative ion, and the ionization 27 dynamics of the second electron from the neutral atom 28 are not considered in detail. More recently, a number 29 of studies have been devoted to double detachment in 30 an intense field (> 10^{13} W·cm⁻²), where the absence of 31 a long-range Coulomb potential and the lower binding 32 energy compared to atoms are expected to significantly 33 modify the dynamics of non-sequential processes [5–10]. 34 The work reported here concerns double detachment 35 ³⁶ of the 1s2s2p ⁴P^o metastable state of He⁻ in moderately

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³⁷ strong laser fields, with intensities comparable to those ³⁸ of earlier work [2, 3], where multiphoton processes domi-³⁹ nate. While no other experimental or theoretical data is ⁴⁰ available for this anion, the choice of such a few-electron ⁴¹ system allows the physics to be studied in detail both ex-⁴² perimentally and theoretically. For moderate intensities, $_{43}$ the process can be treated as sequential, *i.e.* as single ⁴⁴ photodetachment of the anion followed by ionization of ⁴⁵ the neutral atom. The first step, shown schematically ⁴⁶ on the left-hand side of Fig. 1, involves the one-photon ⁴⁷ detachment of He⁻: this has already been studied both ⁴⁸ experimentally and theoretically, with good overall agree-⁴⁹ ment [11–13]. Detachment in the wavelength ranges from $_{\rm 50}$ 530 nm to 560 nm and from 685 nm to 730 nm leaves he-⁵¹ lium in the 1s2s ³S and 1s2p ³P^o states, which can subse-⁵² quently ionize by absorption of three photons, as shown ⁵³ on the right-hand side of Fig. 1.

It has already been shown that multiphoton ioniza-54 ⁵⁵ tion of excited helium atoms is greatly enhanced by ⁵⁶ resonances with higher-lying states [14–17], a process 57 known as Resonance Enhanced MultiPhoton Ionization 58 (REMPI). In a previous paper [18], hereafter referred to ⁵⁹ as paper P1, we investigated three-photon ionization of ⁶⁰ He(1s2p ³P^o), observing two-photon resonances with Ry-61 dberg states as well as an extra resonant pathway via ⁶² the low-lying 1s3s ³S state which further complicates the ⁶³ ionization dynamics. Using the same experimental setup ⁶⁴ and effective Hamiltonian approach, we here extend the 65 study reported in P1 over a wider range of photon ener-⁶⁶ gies and investigate the effect of different laser polariza-⁶⁷ tions. The first, photodetachement step is also treated 68 in more detail.

⁶⁹ We have shown in paper P1 that the magnetic quan-⁷⁰ tum number M influences the double detachment dy-⁷¹ namics since for M = 0, extra ionization pathways are ⁷² allowed. The influence of M on double detachment in ⁷³ strong laser fields has also been discussed within the

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 He^{-} (1s2s2p ⁴P°)

FIG. 1. Schematic energy level diagram of He⁻ and He. The dotted arrows show the pathway responsible for double detachment of He⁻ via the 1s2s intermediate state and through (2+1) REMPI (Resonance Enhanced MultiPhoton Ionization) of the atom. The solid arrows show the pathway via the 1s2p state and through (1+1+1) and (2+1) REMPI of the atom.

74 framework of Ammosov-Delone-Krainov or Keldysh-like 107 by ground state helium atoms due to spontaneous de-⁷⁵ models [8, 9], and was shown to be of some importance in 108 tachment is very weak, of the order of one percent. ⁷⁶ the saturation of the second, ionization step. This influ-77 ence will also be investigated below, in particular how it 110 from a tunable dye laser pumped by the second or third ⁷⁸ depends on the wavelength and polarization of the laser 111 harmonics of a Q-switched Nd:YAG laser. Coumarin 500 79 light.

The paper is organized as follows: the experimental 80 setup is briefly described in Section II; the *R*-matrix cal-81 culation for the photodetachment of He⁻ together with 82 the effective Hamiltonian approach used to treat ioniza-83 tion of helium are presented in Section III. Section IV 84 presents and discusses the experimental and theoretical 85 results. 86

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II. EXPERIMENTAL SETUP

88 ⁸⁹ sentially the same as in paper P1. We therefore give ¹²⁵ is placed between the attenuator and the lens to change ⁹⁰ only a brief summary of its main features but describe in ¹²⁶ the polarization of the laser beam from linear to circular ⁹¹ more detail the few modifications required for the present ¹²⁷ or elliptic. To avoid spurious depolarisation, no mirrors study. 92

A beam of He⁺ ions is first extracted from a duoplas- ¹²⁹ 93 94 $_{95}$ a permanent magnet. Negative helium ions He⁻(1s2s2p $_{131}$ laser pulse itself (~ 5 ns FWHM). During their transit, 97 sium atoms in a vapor cell, with an efficiciency of about 133 neutral atoms can further be ionized by absorbing 3 more 98 1%. Double electrostatic deflection subsequently clears 134 photons. Beyond the interaction region, the He⁺ ions are ⁹⁹ the He⁻ beam of its positive (He⁺) and neutral (He⁰) ¹³⁵ analyzed in energy by a quadrupolar deflector combined 100 components before it enters the region of interaction with 136 with a 60° cylindrical deflector and an analyzing slit, and $_{101}$ the laser beam, pumped to high vacuum (~ 10^{-8} mbar). $_{137}$ detected by micro-channel plates (MCP). The anions are ¹⁰² Although He⁻(1s2s2p ⁴P^o) is metastable, the lifetimes ¹³⁸ collected on the other side of the quadrupole by a Fara- $_{103}$ of its J = 1/2, 3/2 and 5/2 fine structure components, $_{139}$ day cup. In order to collect only those He⁺ ions produced $_{104}$ 7.8 μ s, 12.3 μ s and 359 μ s respectively [19], are suffi- $_{140}$ by photodetachment and ionization within the same dye ¹⁰⁵ ciently long to perform the experiment using conventional ¹⁴¹ laser pulse, a bias of 100 V is applied to the interaction $_{106}$ beam transport techniques. Contamination of the beam $_{142}$ region with the dye laser. He⁺ ions produced by double



FIG. 2. Experimental setup. Cs: cesium vapor cell; PD: planar deflector; FC: Faraday cup; CD: cylindrical deflector; IR: biased interaction region; Q: quadrupolar deflector; MCP: multichannel plates; Ti:Sa: Ti:Sapphire laser beam; Dye: dye laser beam. Double arrows indicate convergent lenses. The laser beams propagate along the z direction and their polarizations, when linear, are along the y axis.

The ion beam is illuminated by nanosecond laser pulses ¹¹² and Pyridine 1-2 dyes were used to cover wavelength 113 ranges from 530 nm to 560 nm and from 685 nm to ¹¹⁴ 730 nm respectively. At the laser output, the pulse en-¹¹⁵ ergy is attenuated to the required value by the combina-116 tion of a $\lambda/2$ plate mounted on a high accuracy rotation ¹¹⁷ stage and a polarizing beam splitter. Laser light is then ¹¹⁸ focused onto the ion beam inside the vacuum chamber 119 by an f = 40 cm lens and collected, as it exits the cham-120 ber, by a pulse energy meter. The attenuation system 121 is servo-controlled in order to maintain a constant pulse 122 energy throughout the dye gain curve. The size of the ¹²³ waist at focus is about 54 μ m, resulting in a peak inten-The experimental setup, presented in Fig. 2, is es- 124 sity of 2.9 ×10¹⁰ W/cm² for 6 mJ pulses. A $\lambda/4$ -plate ¹²⁸ are used beyond the $\lambda/4$ -plate.

Ions cross the laser spot (twice the waist) in about matron source, accelerated to 4 keV and mass selected by 130 250 ps, an interval much shorter than the duration of the $^{4}P^{o}$) are produced by double charge transfer with ce- $_{132}$ some ions undergo photodetachment and the resulting

144 eV while those produced by ionization of incoming He 182 resonances are accessible and the cross section exhibits no 145 atoms gain only 100 eV. He⁺ ions produced by collisions 183 sharp variation. Overall, there is relatively good agree-146 $_{147}$ no energy. The subsequent energy analysis performed by $_{155}$ results in this region [11–13, 20–25]. Previous studies 148 the quadrupolar and cylindrical deflectors readily sepa- 186 however only dealt with *linear* polarization, whilst the 149 rates the various contributions. Moreover, the detection 187 present goal is to study double detachment in an el-¹⁵⁰ of laser-induced He⁺ ions is performed only in a narrow ¹⁸⁸ liptically polarized field. Moreover, while partial cross ¹⁵¹ time-window centered around their time-of-flight from ¹⁸⁹ sections for photodetachment into the He(1s2s ³S) and $_{152}$ the interaction region. A second, time-shifted window $_{190}$ He(1s2p 3 P^o) states have been considered by some au-154 155 free measurements, with less than 3 background counts 193 with magnetic quantum numbers $M_f = 0, \pm 1$. Such parper hundred laser shots. 156

157 ¹⁵⁸ ble detachment process, we have performed a two-color ¹⁹⁶ tion of He(1s2p ${}^{3}P^{o}$) strongly depends on M_{f} (see paper ¹⁵⁹ experiment where helium atoms are first prepared in the ¹⁹⁷ P1). We have thus derived a formula for partial cross ¹⁶⁰ 1s2p state outside the interaction region by photodetach-¹⁹⁸ sections to the magnetic sublevels of a given state and ¹⁶¹ ment by a CW Ti:Sapphire laser and ionized downstream ¹⁹⁹ calculated their values in the desired wavelength range 162 by pulses from the dye laser. By operating the tun- 200 based on reduced dipole matrix elements obtained from $_{163}$ able CW laser at a wavelength of 1005 nm, more than $_{201}$ an R-matrix calculation. $_{164}$ 99% of photodetachment occurred into He(1s2p $^{3}P^{o}$) as 202 165 the cross section of this process reaches a maximum of 203 only dipole-allowed transitions. Since the initial state $_{166}$ 3.6 × 10⁻¹⁹ m² due to a resonance with the He⁻(1s2p² 204 (1s2s2p ⁴P^o) has a total spin $S_i = 3/2$, the dipole selec-¹⁶⁷ ⁴P^e) autodetaching state [11, 12]. During transit between ²⁰⁵ tion rules impose that the residual atom is left in a triplet 168 the two laser foci, separated by less than 1 cm, 20% of the 206 state. For ease of notation, we therefore do not explicitly ¹⁶⁹ He(1s2p ³P^o) states decay spontaneously into He(1s2s ²⁰⁷ specify the spin quantum numbers in what follows. $_{170}$ ³S). By selecting downstream those ions that gained a 208 ¹⁷¹ kinetic energy of 100 eV, we could detect He⁺ ions pro-²⁰⁹ denoted by $|\alpha_i L_i M_i\rangle$, where L_i is the total orbital angu- $_{172}$ duced by sequential, two-color detachment and ionization $_{210}$ lar momentum of the state i, M_i its magnetic quantum 173 only.

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III. THEORY

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Single photodetachment of He⁻ Α.

176 177 some detail over the past few decades, with particular 221 orbitals. The length form of the partial cross section for 178 emphasis on resonances due to doubly excited states em- 222 photodetachment to a particular magnetic sublevel, aver-¹⁷⁹ bedded in the continuum (see the review article [1] and ²²³ aged over the initial states and summed over the possible ¹⁸⁰ references therein). In the wavelength ranges spanned by ²²⁴ spin projections of the final states is given by

143 detachment of He⁻ in this region gain an energy of 200 181 the present study (530-560 nm and 685-730 nm), no such with the residual gas outside the interaction region gain ¹⁸⁴ ment between the various theoretical and experimental is used to measure the background signal. The selection ¹⁹¹ thors, no data exist for partial cross sections to the variin energy and time-of-flight ensures quasi background- $_{192}$ ous magnetic sublevels of a final state, e.g., He(1s2p $^{3}P^{\circ})$ ¹⁹⁴ tial cross sections are not anecdotal since, for example, In order to assess the sequential nature of the dou- 195 the dynamics of resonance-enhanced multiphoton ioniza-

In what follows, we assume L-S coupling and consider

We consider an initial state of the unpolarized anion, ²¹¹ number and where α_i represents all other quantum num-²¹² bers required to specify the state. The residual atom is ²¹³ left in a state $|\alpha_f L_f M_f\rangle$ of total orbital angular momen- $_{214}$ tum L_f and magnetic quantum number M_f . The ejected ²¹⁵ electron is described by continuum orbitals $|\epsilon_f \ell_f m_f\rangle$ of ²¹⁶ energy ϵ_f , orbital angular momentum ℓ_f and magnetic 217 quantum number m_f . The final continuum state of to- $_{218}$ tal angular momentum L and magnetic quantum number ²¹⁹ M, denoted by $|\alpha_f L_f \ell_f L M\rangle$, is obtained by coupling to-One-photon detachment of He⁻ has been studied in 220 gether the residual atom wave function and continuum

$$\sigma_{\alpha_f L_f M_f}(\hat{\boldsymbol{\epsilon}}) = \frac{4\pi^2 \alpha a_0^2 \omega}{2L_i + 1} \sum_{M_i} \sum_{L,L'} \sqrt{(2L+1)(2L'+1)} \sum_{\ell_f,m_f} \begin{pmatrix} L_f & \ell_f & L \\ M_f & m_f & -M \end{pmatrix} \begin{pmatrix} L_f & \ell_f & L' \\ M_f & m_f & -M \end{pmatrix} \times \langle \alpha_f L_f \ell_f L' M | D(\hat{\boldsymbol{\epsilon}}) | \alpha_i L_i M_i \rangle^* \langle \alpha_f L_f \ell_f L M | D(\hat{\boldsymbol{\epsilon}}) | \alpha_i L_i M_i \rangle,$$
(1)

which is derived from the general expression of the dipole matrix elements given by Burke [26], and where α is the fine-structure constant, a_0 is the Bohr radius, ω is the photon angular frequency and $D(\hat{\boldsymbol{\epsilon}})$ is the dipole length

operator for a given polarization vector $\hat{\boldsymbol{\epsilon}}$. In the "natural" reference frame of Tumaikin and Yudin [27, 28], this is expressed as

$$\hat{\boldsymbol{\epsilon}} = \mathbf{e}_0 \sqrt{\cos 2\varepsilon} - \mathbf{e}_{\pm 1} \sqrt{2} \sin \varepsilon \tag{2}$$

²²⁵ where $\mathbf{e}_{0,\pm 1}$ are spherical unit vectors [29] and the ellip- $_{226}$ ticity angle ε can take the values $-\pi/4 \le \varepsilon \le \pi/4$. On $_{242}$ identical results for the partial photodetachment cross ²²⁷ the right-hand side, the helicity of the vector $\mathbf{e}_{\pm 1}$ corre-228 sponds to the sign of ε . The polarization is linear when 244 tions of the angular momentum along the quantization $_{229} \varepsilon = 0$, left circular when $\varepsilon = \pi/4$ and right circular when $_{245}$ axis, and an appropriate rotation must therefore be per- $_{230} \varepsilon = -\pi/4$. The choice of reference frame is arbitrary, and $_{246}$ formed in order to make meaningful comparisons. ²³¹ a commonly used convention is to choose the quantiza- $_{232}$ tion axis z along the electric field in the case of linear polarization, and along the direction of light propagation in 247 233 ²³⁴ the case of circular polarization. The natural frame pos-²⁴⁸ dipole matrix elements appearing in (1) can be obtained $_{235}$ sesses the advantage of bridging these two conventions by $_{249}$ from the reduced matrix elements $(\alpha_f L_f \ell_f L || D || \alpha_i L_i)$ ²³⁶ performing a continuous rotation of the reference frame ²⁵⁰ using the Wigner-Eckart theorem [29]:

237 as the ellipticity angle evolves from 0 to $\pm \pi/4$. The ori-238 entation of the reference frame in the laboratory frame ²³⁹ thus depends on the ellipticity angle, and it is identical 240 to standard conventions in the limiting cases of linear 241 and circular polarization. Any other frame would yield $_{243}$ sections, keeping in mind that the M values are projec-

With the definition (2) for the polarization vector, the

$$\langle \alpha_f L_f \ell_f LM | D(\hat{\boldsymbol{\epsilon}}) | \alpha_i L_i M_i \rangle = (-1)^{-L-M} \left[\begin{pmatrix} L & 1 & L_i \\ -M & 0 & M_i \end{pmatrix} \sqrt{\cos 2\varepsilon} - \begin{pmatrix} L & 1 & L_i \\ -M & \pm 1 & M_i \end{pmatrix} \sqrt{2} \sin \varepsilon \right] (\alpha_f L_f \ell_f L || \boldsymbol{D} || \alpha_i L_i),$$
(3)

²⁵¹ where **D** is the tensor operator corresponding to $D(\hat{\epsilon})$. ²⁷⁷ $_{252}$ Note that the reduced matrix element does *not* depend ²⁵³ on the polarization.

The cumulated photodetachment probability to a par-254 255 ticular final state $|\alpha_f L_f M_f\rangle$ is readily obtained from ²⁵⁶ the partial cross sections by solving the rate equation $_{257} dN(t)/dt = -\sigma \phi(t) N(t)$ with appropriate boundary con-258 ditions,

$$P_{L_f M_f}(t) = \frac{\sigma_{\alpha_f L_f M_f}}{\sigma} \times \left[1 - e^{-\int_{-\infty}^t dT \sigma \phi(T)}\right], \quad (4)$$

 $_{260}$ tal cross section σ is obtained by summing the partial $_{291}$ quasiresonant bound states, and its orthogonal comple- $_{261}$ cross sections over all quantum numbers. Note that the $_{292}$ ment \mathscr{Q} spans the rest of Hilbert space. The effec-262 intensity, or photon flux, required for double detachment 293 tive Hamiltonian is built from the exact Hamiltonian of $_{263}$ is very high and therefore, in the region where it occurs, $_{294}$ the \mathscr{P} -space, while the \mathscr{Q} -space is included through ad-264 the exponential term on the right hand side of the above 295 ditional, perturbative matrix elements coupling model-265 equation is essentially zero, *i.e.* photodetachment is sat- 296 space states. In this respect, EH theory can be considered 266 urated.

In order to compute the partial cross sections (1, 267 268 3) and hence the cumulated photodetachment proba-269 bility, we require the reduced dipole matrix elements $_{270}$ ($\alpha_f L_f \ell_f L || \boldsymbol{D} || \alpha_i L_i$), which are independent of the polar-²⁷¹ ization. In the work reported here, these were obtained $_{272}$ from a standard *R*-matrix calculation including the five ²⁷³ lowest triplet states of helium (1s2s ³S, 1s2p ³P^o, 1s3s ²⁷⁴ ³S, 1s3p ³P^o, 1s3d ³D), using the UK APAP (Atomic ²⁷⁵ Processes for Astrophysical Plasmas) suite of computer ²⁷⁶ codes [30]. Details are given in the appendix.

в. Three-photon ionization of He

After the first electron is ejected from He⁻, and as 279 the neutral atom moves forward through the laser pulse, the intensity rises. If the photon energy is appropriately 280 chosen, the intensity may become sufficiently high to fa-281 vor the (1+1+1) or (2+1) REMPI of the atom. In P1, 282 ²⁸³ we have shown that Effective Hamiltonian (EH) theory ²⁸⁴ can provide an accurate description of the phenomenon ²⁸⁵ and hence can be used to model the experiment in detail. EH theory has been described in some length elsewhere 286 [31–33], so we shall only briefly summarize it here, con-287 centrating on the details relevant to the present study. 288

In EH theory, Hilbert space is partitioned into two 289 259 where $\phi(T)$ is the instantaneous photon flux and the to- 290 different subspaces: the model space \mathscr{P} contains the ²⁹⁷ as a semiperturbative treatment of multiphoton ioniza-²⁹⁸ tion. Choice of the \mathscr{P} -space is critical since it must be 299 small enough so that the effective Hamiltonian matrix is 300 kept small and calculations are relatively simple, and yet 301 contain all states essential for the REMPI dynamics.

> The effective Hamiltonian $H_{\rm eff}$ satisfies the following 302 303 eigenvalue equation

$$H_{\text{eff}} \left| \psi_p \right\rangle = E \left| \psi_p \right\rangle,\tag{5}$$

 $_{\rm 304}$ where $|\psi_p\rangle$ is the model space wave function. Each eigen-

³⁰⁵ value can be written as $E = E_0 + \Delta - i\frac{\Gamma}{2}$, the real ³²⁹ or 1s2p initial states, N = 1 for the 1s3s and 1s3d states $_{300}$ tive Hamiltonian valid up to the second order of pertur- $_{332}$ where ϵ_f is the photoelectron energy. 309 bation theory is

$$H_{\text{eff}} = PH_0P + PVP + P\left(S + \Omega - i\frac{\Gamma}{2}\right)P \quad (6)$$

 $_{310}$ where V is the exact atom-field interaction operator $_{311}$ and H_0 is the exact "free" Hamiltonian, containing the $_{\rm 312}$ field-free atomic and light-field Hamiltonians. P is the 313 Feshbach projection operator, projecting the wave func-314 tion onto \mathscr{P} -space states: $\sum_{i\in\mathscr{P}}|i\rangle\langle i|$. The operator S $_{315}$ is a two-photon transition operator connecting the $\mathscr{P}\text{-}$ 316 space states *via* nonresonant bound states and the oper- $_{317}$ ators Ω and Γ represent two-photon couplings between ³¹⁸ model-space states through the ionization continua (see ³¹⁹ P1 for the detailed expression of these operators).

The eigenvalues of H_0 for bound states are E_i = 320 $_{321} \mathcal{E}_i - N\omega$, where \mathcal{E}_i is the field-free energy of the bound $_{349}$ ing these elements. $_{322}$ state and N is the number of photons absorbed. Field- $_{350}$ $_{323}$ free energies for low-lying bound states ($n \leq 3$) are taken $_{351}$ the REMPI of the 1s2s and 1s2p initial states respec-³²⁴ from the NIST atomic database [34]. The energies of Ry-³⁵² tively. For the 1s2s state, ionization proceeds via a 2-325 dberg states are computed from their principal quantum 353 photon resonance with the Rydberg 1sns and 1snd states. $_{326}$ number n and quantum defect $\delta_{n\ell}$. The latter is calcu- $_{354}$ The model space includes the 1s2s, 1sns and 1snd states $_{327}$ lated using Ritz's expansion with coefficients given by $_{355}$ with n in the range from 4 to 27, and the wave function ³²⁸ Drake [35]. By convention, N = 0 for either of the 1s2s ³⁵⁶ is expressed as

 $_{306}$ part being the Stark-shifted energy of the field-dressed $_{330}$ and N = 2 for the Rydberg states. The eigenvalues of ³⁰⁷ atomic state and Γ its total ionization width. The effec-³³¹ the continuum states of H_0 are, similarly, $e = \epsilon_f - 3\omega$,

> The one- and two-photon matrix elements (V_{ij}, S_{ij}) , 333 ³³⁴ Ω_{ij} and Γ_{ij}) coupling states of the model space are 335 expressed within the dipole and rotating-wave approx-³³⁶ imations [36], justified by the moderate laser intensities 337 and the explicit treatment of only quasiresonant bound 338 states. Their calculation for all polarization states $\hat{\epsilon}$ is ³³⁹ prohibitive, but can be greatly simplified, as for the pho-340 todetachment of He⁻, by virtue of the Wigner-Eckart ³⁴¹ theorem. Reduced matrix elements, independent of $\hat{\epsilon}$, 342 are calculated only once using: (i) two-electron DVR ba-³⁴³ sis functions for couplings between low-lying states; (ii) ³⁴⁴ Quantum Defect wave functions for couplings amongst ³⁴⁵ Rydberg states and between Rydberg and continuum ³⁴⁶ states (see paper P1). Dipole matrix elements for any, ³⁴⁷ arbitrary $\hat{\epsilon}$ are then readily calculated using Eq. (3) and 348 the effective Hamiltonian matrix is then constructed us-

We have built two effective Hamiltonians to describe

$$|\psi_p(t)\rangle = c_{2s0}(t) |2s0\rangle + \sum_{n=4}^{27} \left[c_{ns0}(t) |ns0\rangle + \sum_M c_{ndM}(t) |ndM\rangle \right].$$
 (7)

 $_{357}$ The 1s orbital has been omitted in the $|n\ell M\rangle$ basis vectors for brevity. The total orbital angular momentum L has $_{358}$ also been omitted and is equal to that of the outer electron ($L = \ell$). The summation over the total magnetic quantum ³⁵⁹ number M runs from $-\ell$ to $+\ell$ for each basis vector. For the 1s2p state, ionization proceeds through the (1+1+1) $_{360}$ and (2+1) REMPI schemes shown in Fig. 1. The model space is thus spanned by one of the magnetic sublevels of $_{361}$ the 1s2p state, defined by its magnetic quantum number M_f , and the 1s3s, 1s3d, 1snp and 1snf states. The wave 362 function is given by

$$|\psi_p(t)\rangle = c_{2pM_f}(t) |2pM_f\rangle + c_{3s0}(t) |3s0\rangle + \sum_M \left\{ c_{3dM}(t) |3dM\rangle + \sum_{n=6}^{45} \left[c_{npM}(t) |npM\rangle + c_{nfM}(t) |nfM\rangle \right] \right\}.$$
 (8)

 $_{364}$ finite size and includes only the bound and continuum $_{375}$ Hamiltonian, with a size up to 409×409 for the 1s2p $_{365}$ states involved in two-photon couplings between \mathscr{P} -space $_{376}$ state. ₃₆₆ states. It comprises bound states up to n = 70 and ₃₇₇ 367 continuum states up to energies where bound-free cou- 378 used to propagate the wave function along the atom's 368 $_{369}$ rules ($\Delta M = 0$) permit only those states with the same $_{380}$ ing the time-dependent Schrödinger equation [37], whose ³⁷⁰ magnetic quantum number as the initial state to be pop- ₃₈₁ formal solution may be written as ³⁷¹ ulated. This in turn keeps the size of the effective Hamil- $_{372}$ tonian small, e.g., 83×83 for the 1s2p initial state. In ³⁷³ the general case of elliptic polarization, looser selection

³⁶³ For both initial states, the 2-space is truncated to a $_{374}$ rules ($\Delta M = 0, \pm 1$) yield a significantly larger effective

Once the effective Hamiltonian is constructed, it is plings are negligible. For linear polarizaton, selection 379 trajectory through the laser pulse. This is done by solv-

$$\left|\psi_{p}(t+\Delta t)\right\rangle = \mathrm{e}^{-\mathrm{i}H_{\mathrm{eff}}(t)\Delta t} \left|\psi_{p}(t)\right\rangle. \tag{9}$$

382 The wave function is iteratively propagated by comput-383 ing numerically the matrix exponential $e^{-iH\Delta t}$ with the ³⁸⁴ EXPOKIT computer package [38]. The time dependence 385 of the effective Hamiltonian arises from the dependence ³⁸⁶ of the couplings between model-space states on the in-387 tensity of the laser field, itself evolving in time as the 388 atom travels through the laser focus. The wave func-389 tion is propagated up to about $t = 4 \times 10^7$ a.u. in steps $_{390}$ of $\Delta t \simeq 10^4$ a.u. The initial condition for propagation $_{391}$ starting from the He(1s2s 3 S) state is

$$|c_{2s0}(0)|^2 = P_{2s0}(t \to \infty),$$
 (10)

 $_{392}$ where $P_{2s,0}$ is the detachment probability, given by ³⁹³ Eq. (4). All other coefficients are zero. Similarly, the ³⁹⁴ initial condition for the He(1s2p ${}^{3}P^{o}$) state is given by

$$|c_{2pM_f}(0)|^2 = P_{2pM_f}(t \to \infty),$$
 (11)

³⁹⁵ and all other coefficients are zero. Such initial conditions ³⁹⁶ imply that the photodetachment and ionization processes ³⁹⁷ are sequential, and that the former occurs well before 398 the latter, *i.e.* at much lower intensities. Due to the ³⁹⁹ non-hermicity of the effective Hamiltonian, the norm of $_{429}$ in Fig. 3, for different light polarizations at $\lambda = 690$ nm $_{400}$ the wave function at the end of the propagation has de- $_{430}$ (1.797 eV). The population of the 1s2s state ($R_{2s} = 0.66$) $_{401}$ creased by an amount p. This quantity corresponds to $_{431}$ is nearly twice that of the 1s2p state ($R_{2p} = 0.34$), and $_{402}$ the probability of ejecting 2 electrons from the He⁻ ion $_{432}$ this sharing is independent of the ellipticity. The 1s2p 403 via either the He(1s2s ³S) state or one of the magnetic 433 state is slightly less populated at shorter wavelengths sublevels of the He(1s2p ${}^{3}\text{P}^{\text{o}}$) state. The total ejection ${}_{434}$ ($R_{2p} = 0.33$ for $\lambda = 685$ nm) and slightly more populated ⁴⁰⁵ probability p_{tot} is the sum over all channels. Note that ₄₃₅ at longer wavelengths ($R_{2\text{p}} = 0.39$ for $\lambda = 730$ nm). $_{\rm 406}$ for each photon energy, $p_{\rm tot}$ is computed for a range of $_{\rm 436}$ $_{407}$ peak intensities and appropriate averaging over the ion $_{437}$ isotropic sublevel $M_f = 0$ does not depend on the el-408 beam section and integration over the pulse duration is 438 lipticity. The 1s2p state has three magnetic sublevels ⁴⁰⁹ subsequently performed in order to simulate experimen- $_{410}$ tal conditions (see paper P1). Simulated results are also $_{440}$ $M_f = -1$ respectively increases and decreases slightly as $_{411}$ multiplied by 0.56 to account for the MCP detection ef- $_{441}$ the ellipticity angle goes from 0 to $\pi/4$. The branching 412 ficiency.

413

IV. **RESULTS AND DISCUSSION**

414

Single photodetachment Α.

415 $_{416}$ He⁻ under elliptical polarization and in the wavelength $_{450}$ 27% in $M_f = -1$. This may be expected since the dipole $_{417}$ range from 685 nm to 730 nm (1.698 eV to 1.810 eV). $_{451}$ transition selection rule is $\Delta M = +1$. For right circu-⁴¹⁸ Branching ratios to He(1s2s ³S) and He(1s2p ³P^o, $M_f = 452$ lar polarization σ^- ($\varepsilon = -\pi/4$), the opposite behavior is $_{419}$ 0, ±1) final states were calculated using Eq. (1) with $_{453}$ observed with the $M_f = -1$ state being more populated. $_{420}$ reduced dipole matrix elements obtained from the R_{-454} The branching ratios to the various magnetic sublevels ⁴²¹ matrix calculation described in the appendix. This ⁴⁵⁵ of the 1s2p state depend on the choice of the reference $_{422}$ calculation yielded an electron affinity of 75.5 meV, $_{456}$ frame, since M_f values represent the projection of the 423 which compares favorably with the experimental value 457 orbital angular momentum onto the quantization axis z. ⁴²⁴ of 77.516 meV [39]. The total and partial cross sections ⁴⁵⁸ In the natural reference frame, the polarization vector $_{425}$ are in good agreement with those of an earlier *R*-matrix $_{459}$ given by Eq. (2) rotates in the laboratory frame with ⁴²⁶ calculation [11].

427 428 dent on the wavelength. A representative set is shown 462 quantization axes with different orientations. Such de-



FIG. 3. Branching ratios $R_{2s,2p}$ for the photodetachment of He⁻ into the He(1s2s ³S) and He(1s2p ³P^o) states at $\lambda =$ 690 nm. Full thin line: 1s2s state, dashed line: 1s2p state. Branching ratios to the various magnetic sublevels of the 1s2p state are also shown, and corresponding M_f values are labeled on the right hand-side of the graph. Dotted line: $M_f = -1$, full thick line: $M_f = 0$, dash-dotted line: $M_f = +1$.

The branching ratio to the 1s2s state with a single, ⁴³⁹ with $M_f = 0, \pm 1$. The branching ratio for $M_f = 1$ and 442 ratio for $M_f = 0$ is essentially independent of ellipticity, ⁴⁴³ with only a slight (5%) increase between linear and circu-⁴⁴⁴ lar polarization. For linear polarization ($\varepsilon = 0$), the three $_{445}$ sublevels are almost equally populated, with 34% in each 446 of the $M_f = 1$ and -1 states and 32% in the $M_f = 0$ 447 state. For left circular polarization σ^+ ($\varepsilon = \pi/4$), the ⁴⁴⁸ $M_f = 1$ state is preferentially populated with 39% of the We have studied the one-photon, single detachment of $_{449}$ total 1s2p population, compared to 34% in $M_f = 0$ and ⁴⁶⁰ the ellipticity angle, so that magnetic quantum numbers The branching ratios are in fact only weakly depen- 461 for different polarizations correspond to projections onto ⁴⁶³ pendence however does not affect the final conclusions 464 since all calculations to determine measurable quanti-465 ties are performed within the same frame, and the final 466 results are summed over all M_f values, *i.e.* all orienta-467 tions, before being compared to experiment. As noted 468 in the previous section, for linear ($\varepsilon = 0$) and circular $_{469}$ ($\varepsilon = \pm \pi/4$) polarizations, the natural frame coincides 470 with the standard choice of reference frame and com-⁴⁷¹ parisons with other data are straightforward. Moreover, 472 calculations within other reference frames would yield re-473 sults equivalent to the present ones and which can be ⁴⁷⁴ compared to one another after appropriate rotation by $_{475}$ means of Wigner D matrices [29].

We note that in paper P1, a population distribution of 476 (0.25, 0.5, 0.25) was assumed for the (-1, 0, 1) magnetic 477 478 sublevels of the 1s2p state for linear polarization, as it ⁴⁷⁹ provided the best fit to some of the experimental results. The present calculation proves this assumption incorrect 480 481 and, while it does not modify the conclusions drawn in ⁴⁸² P1, it changes to some extent the relative height of the 1snp and 1snf peaks in the simulated He⁺ ion spectra. 483

The behavior of the branching ratios for magnetic sub-484 levels is established here for photodetachment of He⁻, 485 but the trends observed most certainly hold for other 486 anions. The magnitude of the photodetachment cross 487 section depends on the reduced dipole matrix elements, 488 but their dependence on magnetic number M_f and polar-489 490 ization $\hat{\epsilon}$ derives from angular momentum algebra. Rel-⁴⁹¹ atively small variations between different anions or final ⁴⁹² states are expected to arise since matrix elements and ⁴⁹³ geometrical factors are entangled in the various summa- $_{494}$ tions of Eq. (1).

495

Double photodetachment в.

496

Transient $He(1s2s \ ^{3}S)$ state 1.

We first consider double detachment in the photon en-497 ⁴⁹⁸ ergy range between 2.22 eV and 2.34 eV, corresponding to the wavelength region from 530 nm to 560 nm cov-499 ered by the laser when supplied with Coumarin 500 dye. 500 Inspection of the helium triplet spectrum indicates that 501 the three-photon ionization of $He(1s2s {}^{3}S)$ is strongly en-502 hanced by two-photon resonances with Rydberg 1sns and 503 1snd states, as shown in Fig. 1, while the two-photon 504 ⁵⁰⁵ ionization of He(1s2p ³P^o) is non-resonant. One may thus expect double detachment to proceed preferentially 506 ⁵⁰⁷ through the 1s2s intermediate state, even though the ⁵⁰⁸ number of photons required is higher than for the 1s2p ⁵¹⁷ photon. In the present wavelength range, double pho-⁵⁰⁹ state. This hypothesis is supported by the experimen-⁵¹⁸ todetachment therefore occurs by photodetachment and ⁵¹⁰ tal spectrum for linear polarization, shown in Fig.4(a). ⁵¹⁹ (2+1) REMPI of He(1s2s ³S). Ionization from higher Ry- $_{511}$ The spectrum represents the number of He⁺ ions, cre- $_{520}$ dberg levels, with n up to 115, has been studied by Wall ⁵¹² ated from an incoming He⁻ beam of 1 nA by a 6 mJ ⁵²¹ et al. [40] using a two-photon laser excitation and static ⁵¹³ laser pulse, as a function of the photon energy. The two ⁵²² field ionization scheme. In the low photon energy range, ⁵¹⁴ series of peaks correspond to two-photon excitation from ⁵²³ a continuous, slowly rising background is observed. It $_{515}$ 1s2s to respectively 1sns and 1snd states with n = 7 $_{524}$ can be attributed to the onset of the (1+1) REMPI of



FIG. 4. Experimental double detachment spectrum as a function of the photon energy. Top graph (a): linear polarization. Bottom graph (b): left circular polarization. Data shown is the number of $\mathrm{He^{+}}$ ions produced by a 6 mJ laser pulse and for a $\mathrm{He^-}$ beam of 1 nA .



FIG. 5. Details of the 1s9d peak in the double detachment spectrum. Full circles: experiment, full line: simulation. Data is for linear polarization and 6 mJ laser pulses. The vertical dotted line indicates the position of the field-free 1s2s-1s9d two-photon resonance. For larger photon energies, the 1s2s energy is Stark shifted and resonance condition is met for different laser intensities, as indicated by the intensity scale. A given intensity corresponds to a particular radius with respect to the center of the laser profile, as indicated by the radius scale. This radius defines an effective interaction cylinder, within which production of He⁺ can occur.

 $_{516}$ to n = 12, which are subsequently ionized by another $_{525}$ the 1s2p state through a one-photon resonance with the

⁵²⁶ 1s3d state, located at $\lambda = 587.7$ nm (2.110 eV). Although we are still far detuned from this resonance, the contribu-527 tion of this two-photon process to the double detachment 528 yield is non-negligible compared to the three-photon pro-529 cess examined here. 530

The detailed profile of the peaks in the double detach-531 532 ment spectrum, for example that corresponding to the 1s2s-1s9d resonance shown in Fig. 5, provides additional 533 information on the ionization dynamics. Note that in 534 this case the simulation was performed for a 68 μ m laser 535 waist in order to match the width of the experimental peak. No vertical scaling was applied to the simulated 537 data. The size of the waist has not been measured for 538 the present wavelength range and deviations from the 54 μm waist measured for the range from 685 nm to 730 nm 540 541 are considered possible. The width of the peak is much larger than the laser bandwidth of 0.05 cm⁻¹ (6.2 μeV), 543 and it is asymmetrical, with a pronounced spread towards higher photon energies. Processes leading to this 544 type of profile are threefold, and have been partly dis-545 cussed in paper P1 and by other authors [40]. We first 546 note that the 1s2s state is shifted down in energy for in-547 creasing laser intensities by the AC Stark shift, while the Rydberg series remain essentially unperturbed. There-549 fore, even if the laser is blue-detuned from the 1s2s-1s9d 550 resonance, the increasing intensity experienced by the helium atom moving through the laser focus will dynami-552 553 555 556 557 laser profile, the intensity at which resonance occurs cor-558 559 561 562 564 state occurs early on as the atom crosses the laser focus. 599 ments are identical, geometrical factors arising from the 566 567 Rydberg state and the laser field is long and the ioniza- 601 for circular polarization than for linear polarization. In 568 569 population transfer, reduced interaction time and there- 603 zero as the polarization becomes circular, a direct result fore reduced ionization probability, also resulting in a 604 of the dipole selection rules. 570 572 drop in the He⁺ yield.

Turning to circular polarization, Fig. 4(b) provides a 573 particularly illustrative example of dipole selection rules. 574 575 In the case of left circular polarization (σ^+), selection rules for a two-photon transition give $\Delta M = +2$. Ex-577 citation from the 1s2s state to a Rydberg 1sns state is therefore forbidden, and the 1sns peak in the He⁺ spec-578 trum disappears. The same argument applies in the case 579 of right circular polarization (σ^{-}), for which the selection 580 rule is $\Delta M = -2$, hence leading to the same spectrum. 581

582 ⁵⁸³ tion is tuned from linear to left circular is shown in Fig. 6. ⁶¹³ tachment leaves helium in its 1s2s ³S and 1s2p ³P^o states, 584 Measurements were performed both for the 1s2s-1s9d and 614 and three-photon ionization of the latter is enhanced due



FIG. 6. Experimental and simulated double detachment yield as a function of the ellipticity angle. Triangles are experimental data for $\lambda = 539.08$ nm and correspond to resonance with the Rydberg 1s9d state. Circles are experimental data for $\lambda = 540.43$ nm and correspond to resonance with the Rydberg 1s9s state. Both full lines are the result of simulations for the same wavelengths, and have been scaled by 0.8 and 1.02 for the 1s9d and 1s9s states respectively.

⁵⁸⁵ 1s2s-1s9s resonances, corresponding to photon energies of 586 2.2999 eV and 2.2942 eV respectively, by stepwise rotacally bring the 1s2s state into resonance with the 1s9d $_{587}$ tion of the $\lambda/4$ plate while recording the corresponding state, where population transfer occurs. As the laser 588 He⁺ signal. The effective Hamiltonian approach was used is further blue-detuned, the exact resonance condition 589 to simulate the experiment for the same photon energies, is met at increasing intensities, until the value required $_{590}$ and the final results are scaled by 0.8 and 1.02 respecexceeds the laser peak intensity. Assuming a Gaussian 591 tively in order to best fit the experimental values. We ⁵⁹² first note that the shape of the simulation curves follow responds to a specific radius with respect to the center of 593 the experimental data very well. The 1s9d peak amplithe profile, and thus defines a certain interaction cylinder, $_{594}$ tude increases as the polarization gets closer to circuwithin which production of He⁺ can occur. The volume 595 lar, a fact that can be attributed to increasing coupling of this cylinder shrinks as detuning becomes larger, con- 596 strength and ionization rate. For example, inspection of sequently decreasing the $\mathrm{He^{+}}$ signal. Finally, close to a $_{597}$ the value of the matrix element coupling the 1s2s state to zero-field resonance, population transfer to the Rydberg 598 the 1s9d state shows that, while the reduced matrix ele-Therefore, the duration of the interaction between the 600 polarization result in a coupling that is a factor 1.5 larger tion probability high. Blue-detuning results in delayed 602 sharp contrast, the amplitude of the 1s9s peak drops to

Transient $He(1s2p \ ^{3}P^{\circ})$ state 2.

605

606 The double detachment of He⁻ for photon energies $_{\rm 607}$ around 1.76 eV also exhibits resonance series, as can be ⁶⁰⁸ seen in the experimental spectra shown in Fig 7. The ⁶⁰⁹ spectrum for linear polarization has already been studied ⁶¹⁰ in paper P1, so we shall only recall the most important ⁶¹¹ features before investigating the influence of the laser po-The evolution of double detachment as the polariza- ⁶¹² larization. In the present photon energy region, photode-



FIG. 7. Double detachment spectrum as a function of the photon energy. Top graph (a): linear polarization. Bottom graph (b): left circular polarization. The vertical dashed lines indicate the position of the 1s2p-1s3s resonance. Resonances with Rydberg states are observed up to n = 35. The laser pulse energy is 6 mJ.

 $_{615}$ to two-photon resonances with Rydberg 1snp and 1snf616 states. The shape of the spectrum is however very dif-617 ferent from that presented in Fig. 4 due to the presence ⁶¹⁸ of additional, one-photon resonances between the 1s2p 619 and 1s3s states and the 1s3s and 1snp states. Ioniza- $_{620}$ tion proceeds either through (1+1+1) REMPI via the $_{621}$ 1s3s state and a Rydberg 1snp state, or through (2+1)622 REMPI preferentially via a 1snf Rydberg state. In linear 623 polarization, the magnetic sublevel of 1s2p with $M_f = 0$ $_{624}$ ionizes through the (1+1+1) scheme while those with $_{625}$ $M_f = \pm 1$, which cannot couple to the 1s3s state, preferentially do so through the (2+1) scheme. 626

627 628 rightmost peak, corresponding to a 1snf resonance, ex- 658 cordingly. Therefore, the attribution of the 1snp peaks ₆₂₉ hibits the same spread towards higher photon energies $_{659}$ to $M_f = 0$ and the 1snf peaks to $M_f = \pm 1$ for linear $_{630}$ observed for the He(1s2s ³S) transient state, which can $_{600}$ polarization changes to $M_f = -1$ and $M_f = 0, 1$ respec-631 $_{632}$ most peak, corresponding to 1snp resonances, spreads in- $_{662}M_f = -1, 0$ for right circular polarization. As a result, 633 stead towards lower photon energies, indicating that the 663 by carefully tuning the laser wavelength and polariza-634 changes direction towards higher photon energies below $_{665}$ of the initial state of the transient atom. ⁶³⁶ the 1s2p-1s3s resonance (see Fig. 7). This spreading and 637 its reversal can be explained by the strong AC Stark shift 638 ₆₃₉ signs on either side of the resonance [18].

640 ⁶⁴¹ double detachment dynamics, and in particular on the ⁶⁷¹ from two different effects and differ for the 1snp and 1snf 642 ⁶⁴³ of the 1s2p state. The detailed spectrum corresponding ⁶⁷³ changes in the geometrical factors, the non-resonant AC $_{644}$ to resonances with n = 13 states is shown in Fig. 8, along $_{674}$ Stark shift of the 1s2p $(M_f = -1)$ sublevel for circular 645 with the simulated contributions from the $M_f = 0, \pm 1$ 675 polarization is smaller than that of the 1s2p ($M_f = 0$) 646 magnetic sublevels. The most important difference be- 676 sublevel for linear polarization. Hence, since Rydberg 647 tween linear and circular polarization is the change of 677 series are unperturbed and the downward shift of the $_{648}$ shape of the $M_f = 0$ and $M_f = -1$ contributions to the $_{678}$ 1s2p energy is smaller, the peaks will spread less towards



FIG. 8. Details of the double detachment spectrum around the 1s2p-1s13p (left peak) and 1s2p-1s13f (right peak) resonances. Top graph (a): linear polarization; bottom graph (b): left circular polarization. Full circles: experimental data, dash-dot-dot line: simulated $M_f = -1$ contribution, dash-dot line: simulated $M_f = 0$ contribution, dotted line: simulated $M_f = +1$ contribution. For linear polarization, $M_f = +1$ and $M_f = -1$ contributions are identical, therefore only twice the $M_f = -1$ contribution is shown. All simulated contributions are scaled by 0.45.

⁶⁴⁹ He⁺ yield. For linear polarization, selection rules allow $_{650}$ the 1s2p ($M_f=0$) state to couple to the 1s3s state, which ⁶⁵¹ itself couples to the 1s13p state, thus yielding a peak that ⁶⁵² is both broad, because of the strong AC Stark shift of the ⁶⁵³ 1s2p state, and intense, due to the large enhancement of $_{654}$ ionization by the (1+1+1) channel. In the case of left ⁶⁵⁵ circular polarization, coupling to the 1s3s state becomes allowed for the $M_f = -1$ magnetic sublevel, and the con-In the detailed peak profiles presented in Fig. 8(a), the 657 tributions of the various magnetic sublevels change acbe attributed to the same dynamical processes. The left- $_{60}$ tively for left circular polarization and to $M_f = 1$ and 1s2p state is shifted up in energy. The spread however ⁶⁶⁴ tion, it is possible to address a specific magnetic sublevel

666 Apart from the changes in the different M_f contribu-⁶⁶⁷ tions, other differences in the shape of the peaks can be of the 1s2p state due to the 1s3s state, which has opposite observed between the double detachment spectra for cir-⁶⁶⁹ cular and linear polarization, in particular on the high We next investigate the effect of polarization on the 670 photon energy side (see Figs. 7 and 8). These arise respective contributions of the various magnetic sublevels $_{672}$ peaks. Let us first consider the 1snp peaks. Because of ⁶⁷⁹ higher photon energies. However, above the 1s2p-1s3s resonance, the 1s2p state will be significantly shifted upward in energy due to its interaction with the 1s3s state. 681 The spread of the 1snp peak is therefore the result of a 682 competition between the non-resonant and resonant AC 684 Stark shifts, so that a smaller non-resonant contribution 685 leads to an increased upward shift, and therefore an in-686 creased spread towards lower photon energies. In Fig. 8, $_{687}$ the intensity of the 1snp peak is essentially the same for 688 linear and circular polarization. Although the ionization rate of the 1snp states is 16% higher for circular polar-689 $_{690}$ ization, this increase is compensated by the lower (19%) population of the $M_f = -1$ sublevel for circular polariza-691 tion compared to that of the $M_f = 0$ sublevel for linear 692 polarization. 693

Let us now consider the 1snf peak. For linear polar-694 ization, contributions from $M_f = -1$ and $M_f = +1$ are 695 ⁶⁹⁶ strictly equivalent. This is no longer true in the case of circular polarization, and the $M_f = +1$ sublevel gives 697 the dominant contribution while that for $M_f = 0$ is 698 very similar to those for $M_f = \pm 1$ in the case of linear 699 700 polarization. Investigating changes in matrix elements due to modified geometrical factors shows that the AC 701 Stark shift of the 1s2p $(M_f = 0)$ sublevel for circular 702 polarization is identical to that of the 1s2p $(M_f = \pm 1)$ 703 $_{\rm 704}$ sublevels for linear polarization, while that of the $\rm 1s2p$ $_{705}$ ($M_f = 1$) sublevel is twice larger. Matrix elements cou- $_{\rm 706}$ pling the 1s2p state with $M_f=1$ to Rydberg 1snf states 707 are also significantly larger for circular polarization. Fi-⁷⁰⁸ nally, the $M_f = 1$ sublevel is slightly more populated by ⁷⁰⁹ photodetachment in circular polarization. Such increases $_{710}$ explain why the 1snf peak becomes larger and signifi-711 cantly broadens when switching from linear to circular 712 polarization.

713 714 the transmission of the He⁺ ions to the detector, slightly 715 imperfect circular polarization of the laser light, imper-716 fect modeling of the experimental conditions and inaccu-717 racies in the effective Hamiltonian matrix elements. 718

719 720 as a function of the light polarization is shown in Fig. 9. 750 we have performed a two-color experiment where helium 721 722 723 724 725 726 (1.766 eV). The double detachment signal comes mainly 758 tiphoton ionization by the second. 727 from the 1s2p-1s13p resonance, with the contributions 759 728 729 of the various magnetic sublevels evolving from predom-760 in Fig. 10 and compared with the one-laser spectrum, ⁷³⁰ inantly $M_f = 0$ to mostly $M_f = -1$. The onset of ⁷⁶¹ where detachment and ionization occur within the same ⁷³¹ the contribution from $M_f = +1$ as the polarization be-⁷⁶² laser pulse. Since the first laser in the two-color exper-732 comes more circular is reminiscent of the broadening and 763 iment is less efficient in producing helium atoms, the ⁷³³ increase in magnitude of the 1s2p-1snf peaks discussed ⁷⁶⁴ measured yield is smaller and was scaled in Fig. 10 in ⁷³⁴ above, with n = 12 here.



FIG. 9. Experimental and simulated double detachment yield as a function of the ellipticity angle, at the photon energy corresponding to the minimum between the 1s2p-1s12f and 1s2p-1s13p resonances. Full circles: experimental yield, full line: simulated yield, dash-dot-dot line: $M_f = -1$ contribution, dash-dot line: $M_f = 0$ contribution, dotted line: $M_f = +1$ contribution. The experimental yield is for a photon energy of 1.767 eV (701.72 nm) and the simulated one for 1.766 eV(702.05 nm). Simulated yields are scaled by 0.65 to match experimental values. The laser pulse energy is 6 mJ.

Sequential vs. non-sequential 3.

735

The above treatment considers photodetachment and 736 ⁷³⁷ REMPI as sequential events. Given the satisfactory 738 agreement between simulated and measured ion yields, ⁷³⁹ this appears reasonable. Detachment is indeed very effi-⁷⁴⁰ cient and occurs at low intensity, i.e., early in the pulse. 741 The onset of REMPI requires much higher intensities and Discrepancies between experimental and simulated 742 occurs later, when detachment is fully saturated. One yields, as observed in Fig. 8, may be due to variations in ⁷⁴³ possible exception is when the photon energy is tuned $_{744}$ close to the 1s2p-1s3s resonance at $707\,\mathrm{nm},$ since popu-⁷⁴⁵ lation transfer to the Rydberg states via the 1s3s state ⁷⁴⁶ can take place very early in the pulse, at lower intensities ⁷⁴⁷ where photodetachment is not yet saturated.

In order to establish the sequential nature of the dou-748 Finally, the evolution of the double detachment yield ⁷⁴⁹ ble detachment process close the 1s2p-1s3s resonance, The experimental yield was measured for a photon en- 751 atoms are first prepared in the 1s2p state by photodeergy of 1.767 eV, chosen to probe the minimum between $_{752}$ tachment by a CW laser tuned to $\lambda = 1005$ nm, and the 1s2p-1s12f and 1s2p-1s13p resonances, where the in- 753 subsequently ionized downstream by the second, pulsed fluence of the polarization is most prominent. Simula- 754 dye laser used previously. The two laser beams are spations were also performed at the minimum, which in the 755 tially separated, and we select those He⁺ ions that result calculations is located at a slightly lower photon energy 756 from photodetachment by the first laser followed by mul-

> The resulting double detachment spectrum is shown ⁷⁶⁵ order to directly compare with the one-color spectrum.



FIG. 10. Double detachment spectrum around the 1s2p-1s3s resonance. Dot-dashed line: two-color, sequential measurement, multiplied by 35; full line: one-color measurement. The vertical dotted line indicates the exact position of the 1s2p-1s3s resonance. The laser pulse energy is 6 mJ.

766 767 768 769 770 within experimental error bars. 771

772 773 experiments (of the order of $10^{10} \,\mathrm{W/cm^2}$). The domi-774 775 777 778 model [41, 42]. For wavelengths around 707 nm corre-⁸³⁴ and no evidence of non-sequential processes was found. sponding to the 1s2p-1s3s resonance, intensities of over 835 780 781 782 11p electron. Coulomb focusing, which can greatly en- 837 parison with lower intensities, where multiphoton pro-783 784 and multiply charged ions [43], does not occur here since 839 help disentangle sequential and non-sequential processes. 785 the recollision is with a neutral atom.

786

CONCLUSION v.

We have reported a joint experimental and theoretical 787 788 investigation of the double detachment of He⁻ in moderately strong laser fields, for wavelengths ranging from 789 530 nm to 560 nm and from 685 nm to 730 nm, for var-790 ious polarizations of the laser light. The experiment re-791 lies on counting He⁺ ions, produced from a He⁻ beam 792 by 6 mJ laser pulses, as a function of the wavelength 793 and ellipticity angle. The theoretical work treats double 849 794 795 ⁷⁹⁶ ment step is studied within an *R*-matrix approach by ⁸⁵¹ Computational resources have been provided by the 797 calculating branching ratios into the various final atomic s52 supercomputing facilities of the Université catholique

798 states and their magnetic sublevels for arbitrary ellip-799 tical polarization. Subsequent ionization of the neutral atom is studied using effective Hamiltonian models de-800 signed to account for all possible polarizations. They allow lightweight calculations which reproduce faithfully the experimental conditions. 803

The observed double detachment yield contains series 804 ⁸⁰⁵ of peaks arising from resonances between the initial state of the transient atom and Rydberg series. In the range 806 from 530 nm to 560 nm, the 1s2s state produced by pho-807 808 todetachment is coupled, via a two-photon transition, to Rydberg 1sns and 1snd states. In this case, we showed 809 that double detachment proceeds through single detach-810 ment and (2+1) REMPI of the neutral. In the range 811 ^{\$12} from 685 nm to 730 nm, the 1s2p state of the atom is resonantly coupled by one photon to the 1s3s state and by two photons to the 1snp and 1snf Rydberg series. ^{\$15} Double detachment proceeds in this case through single detachment and both (1+1+1) and (2+1) REMPI of the 816 atom. 817

The influence of the laser polarization is manifest in 818 ⁸¹⁹ the 1s2s case, where 1sns resonances disappear for circu-⁸²⁰ lar polarization as a result of the dipole selection rules. No significant differences can be seen, suggesting that no ⁸²¹ Changes in the double detachment spectrum induced by strong non-sequential processes occur. The experiment ⁸²² different polarizations are further explained in terms of was run for linear polarization and we have also observed ⁸²³ geometrical factors. In the 1s2p case, the magnetic quanthat, when the two laser polarizations are parallel or per- 324 tum number M_f strongly influences double detachment pendicular, essentially the same spectra are obtained, to 225 and determines the ionization pathway in the neutral ⁸²⁶ atom. Changing the polarization from linear to ellip-The absence of any non-sequential process is not sur- z_{27} tical and circular substantially modifies the various M_f prising considering the moderate laser intensities in our ⁸²⁸ contributions, which could be computed with the the-⁸²⁹ oretical model. Differences in the shape of the double nant mechanism for non-sequential double ionization is 830 detachment spectra are further explained by geometrical the recollision of the first photoelectron with the residual ⁸³¹ factors and the slight orientation of the atom produced by atom or ion. The laser intensity required for this pro- 832 photodetachment. Finally, the sequential nature of doucess can be estimated using the semi-classical recollision ⁸³³ ble detachment was assessed by a two-laser experiment

The study of double detachment of He⁻ in more in- $8 \times 10^{11} \,\mathrm{W/cm^2}$ would be necessary in order to eject the set tense laser fields is certainly of great interest since a comhance the non-sequential double ionization rate of atoms 333 cesses dominate, is now possible. The present work could ⁸⁴⁰ Moreover, the influence of the magnetic quantum number M in strong field double detachment is expected to 841 842 be important [8, 9], and the present work provides a detailed understanding of this influence in the relatively low 843 intensity region. Finally, photoelectron spectroscopy of 844 the present process could be a perspective for future work 845 since it would provide additional information in terms of 846 double detachment channels and angular distributions. 847

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848

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Appendix: Reduced dipole matrix elements for the 859 photodetachment of He⁻ 860

The reduced dipole matrix elements 861 $(\alpha_f L_f \ell L || \boldsymbol{D} || \alpha_i L_i)$ required when evaluating Eq. (3) 862 ⁸⁶³ for the partial cross sections were extracted from a $_{864}$ standard *R*-matrix calculation. Here, we give details of this calculation, together with some illustrative results 865 to assess the reliability of the reduced dipole matrix 866 elements thus obtained. 867

In the *R*-matrix approach, configuration space is di-868 vided into two regions by a sphere encompassing the 869 ⁸⁷⁰ charge density of all states of the residual atom included ⁸⁷¹ in the calculation. Within this sphere, the states of the (N + 1)-electron system are represented by a discrete 872 873 set of antisymmetrized basis functions built from linear 874 combinations of the residual atomic states coupled with a set of continuum orbitals representing the ejected elec-875 tron, supplemented by a number of bound or short-range 876 correlation configurations. The (N + 1)-electron Hamil-877 tonian is diagonalized in this basis, and the resulting 878 eigenvalues and eigenvectors are used to construct the 908 In CIV3, the radial part of each orbital is written as a 879 $_{880}$ inverse logarithmic derivative matrix or *R*-matrix on the $_{909}$ sum of Slater orbitals: boundary of the inner region. In the outer region, the 881 ⁸⁸² ejected electron moves far from the target while the other 883 electrons remain bound. Exchange between the ejected ⁸⁸⁴ and bound electrons can then be neglected, so that the wavefunctions for the full system can be represented by as a standard close-coupling expansion involving products $_{910}$ The coefficients $C_{jn\ell}$ are uniquely determined by or- $_{287}$ of the residual atomic states and a set of unknown func- $_{911}$ thonormality constraints if $k = n - \ell$, while the in-888 889 differential equations, with the appropriate asymptotic 914 states. 890 boundary conditions determining if the solutions repre-915 891 892 893 894 895 896 897 which are found by an iterative search algorithm. 898

899 photodetachment the residual helium atom can only be 924 states. 900 left in a triplet state. The present calculation includes the 925 901 902 903 ⁹⁰⁴ code [44]. This code is based on a configuration inter-⁹²⁸ ues recommended by the National Institute of Standards ⁹⁰⁵ action approach, in which the wavefunctions for a par- ⁹²⁹ and Technology (NIST). Oscillator strengths and transi- $_{906}$ ticular symmetry $|LS\pi\rangle$ are expressed as antisymmetric $_{930}$ tion probabilities are compared with the NIST values in 907 linear combinations of products of one-electron orbitals. 931 table III.

TABLE I. Slater orbital parameters used to build the helium triplet state wavefunctions.

| | $C_{jn\ell}$ | $I_{jn\ell}$ | $\zeta_{jn\ell}$ | | $C_{jn\ell}$ | $I_{jn\ell}$ | $\zeta_{jn\ell}$ |
|------------|--------------|--------------|------------------|------------|--------------|--------------|------------------|
| 1s | 5.65685 | 1 | 2.00000 | 2p | 0.25282 | 2 | 0.54467 |
| 2s | 1.02331 | 1 | 1.57920 | 3p | 0.14871 | 2 | 0.50810 |
| | -0.33960 | 2 | 0.59932 | | -0.00928 | 3 | 0.31496 |
| 3s | 0.51628 | 1 | 1.53349 | 4p | 5.69931 | 2 | 1.81486 |
| | -0.18699 | 2 | 0.46220 | r | -0.14792 | 3 | 0.88532 |
| | 0.02242 | 3 | 0.37501 | | 0.00010 | 4 | 0.31256 |
| | | | | | | | |
| $\bar{4}s$ | 3.72051 | 1 | 0.57067 | $\bar{5}p$ | 7.08316 | 2 | 2.03268 |
| | -23.34422 | 2 | 1.97582 | | -1.97318 | 3 | 0.90825 |
| | 7.95138 | 3 | 1.78719 | | 0.75867 | 4 | 1.01816 |
| | -1.16998 | 4 | 1.23540 | | -0.00003 | 5 | 0.35646 |
| _ | | | | | | | |
| 5s | 11.03473 | 1 | 1.17185 | 3d | 0.00904 | 3 | 0.33361 |
| | -24.98834 | 2 | 1.09440 | | | | |
| | 15.71030 | 3 | 1.09465 | $\bar{4}d$ | 1.20358 | 3 | 1.35014 |
| | -3.08663 | 4 | 0.98350 | | -0.00025 | 4 | 0.36349 |
| | 0.46320 | 5 | 1.03562 | | | | |
| | | | | $\bar{4}f$ | 0.57024 | 4 | 1.43379 |

$$P_{n\ell}(r) = \sum_{j=1}^{k} C_{jn\ell} r^{I_{jn\ell}} \exp(-\zeta_{jn\ell} r).$$
 (A.1)

tions representing the ejected electron. These unknown $_{912}$ dices $I_{jn\ell}$ and the exponents $\zeta_{jn\ell}$ are variational paramefunctions satisfy an infinite set of coupled second-order 913 ters chosen to minimize the energies of particular atomic

The values of the parameters used in the current study sent a bound state of the initial anion or a continuum ₉₁₆ are given in table I. The 1s orbital is simply that for state of the atom plus ejected electron. The initial bound $_{917}$ the hydrogenic He⁺ ion. The n = 2,3 orbitals were all state and final continuum states are then determined by $_{918}$ optimized on the associated $1sn\ell$ state of helium. The matching the solutions in the inner and outer regions ⁹¹⁹ pseudo-orbitals 4s and 5s were optimized on the 1s2s ³S at their common boundary. For the initial bound state, $\frac{1}{20}$ state, $\frac{1}{4}$ d and $\frac{1}{4}$ f on the 1s2p $^{3}P^{o}$ state. The $\frac{1}{4}$ p and this matching can only be performed at discrete energies, ⁹²¹ 5p pseudo-orbitals were optimized on a linear combina-⁹²² tion of the 1s2p ³P^o and 1s3p ³P^o states as it was found Since the initial He^- anion has a ${}^{4}P^{o}$ symmetry, after 923 that this gave the best energy separation between the two

The energies and excitation thresholds thus obtained five lowest triplet states of helium, whose wave functions 926 are presented in table II, where they are compared with were obtained using the CIV3 atomic structure computer 927 those of a more accurate calculation [35] and with the val-



FIG. 11. Total cross section for the photodetachment of He⁻ as a function of the photon wavelength. Full line: R-matrix calculation using the length form of the dipole matrix elements; dashed line: R-matrix calculation using the velocity form of the dipole matrix elements; broken line: reference [11]; circles: experimental results from reference [13].

In the R-matrix calculation, the inner region extends 932 out to $40 a_0$, and 30 continuum orbitals per angular mo-933 $_{934}$ mentum ℓ are used to represent the ejected electron. The (N+1)-electron Hamiltonian in the inner region is diag-935 ⁹³⁶ onalized for the initial ⁴P^o symmetry and the three final symmetries ⁴S ,⁴P, ⁴D allowed by the dipole selection 937 rules. In the outer region, imposing decaying boundary 938 ⁹³⁹ conditions on the solutions of the coupled second-order differential equations in the ⁴P^o symmetry yields an elec-940 tron affinity of about 75.5 meV for the initial He⁻ state. 941 This compares favourably with the value of 77.518 meV 942 obtained by a more extensive calculation [39], the ex-943 944 perimental value of 77.516 meV [39], and is slightly better than that of an earlier R-matrix calculation [11], in 945 which the computed electron affinity was then slightly 946 adjusted to agree with the accurate value. While such 947 ⁹⁴⁸ small differences may be important close to threshold, ⁹⁴⁹ they have little effect on the overall cross sections in the ⁹⁵⁰ range of photon wavelengths (500-800 nm) considered in 951 this study, and we do not perform such an adjustment ⁹⁵² here. As shown in figure 11, presenting the total cross ⁹⁵³ section for photodetachment of He⁻, the agreement with $_{954}$ the earlier work is very good [11].

TABLE II. Energies and excitation thresholds for the five lowest triplet states of helium. The theoretical values are compared with the accurate, non-relativistic energies taken from chapter 11 of [35] and the thresholds recommended by the National Institute of Standards and Technology (NIST) [34].

| | | Present (au) | Accurate (au) | Present (eV) | NIST (eV) |
|------|--------------|--------------|---------------|--------------|-----------|
| 1s2s | $2^{3}S$ | -2.17513 | -2.17523 | 0.0 | 0.0 |
| 1s2p | $2^{3}P^{o}$ | -2.13294 | -2.13316 | 1.14805 | 1.14449 |
| 1s3s | 3^3S | -2.06866 | -2.06869 | 2.89720 | 2.89885 |
| 1s3p | $3^{3}P^{o}$ | -2.05798 | -2.05808 | 3.18754 | 3.18746 |
| 1s3d | $3^{3}D$ | -2.05562 | -2.05564 | 3.25203 | 3.25404 |

TABLE III. Oscillator strengths f and rates A in length (L) and velocity (V) forms for dipole allowed transitions involving the five lowest triplet states of helium, compared with the values recommended by NIST [34]. The figures in parentheses are the powers of ten by which the preceeding number must be multiplied.

| Transitions | | f | | $A (\text{sec}^{-1})$ | |
|--|--------------|-------------|-------------|-----------------------|-----------|
| | | Present | NIST | Present | NIST |
| 1s2s ³ S - 1s2p ³ P ^o | \mathbf{L} | 0.5421 | 0.5394 | 0.1033(8) | 0.1022(8) |
| * | V | 0.5320 | | 0.1014 (8) | () |
| 1s2 s $^3{\rm S}$ - 1s3p $^3{\rm P^o}$ | \mathbf{L} | 0.5744 (-1) | 0.6448 (-1) | 0.8442(7) | 0.9475(7) |
| | V | 0.6378(-1) | | 0.9373(7) | |
| 1s2p $^3\mathrm{P^o}$ - 1s3 s $^3\mathrm{S}$ | \mathbf{L} | 0.7019 (-1) | 0.6951 (-1) | 0.2795(8) | 0.2785(8) |
| | V | 0.6786 (-1) | | 0.2703(8) | |
| 1s2 p $^3\mathrm{P^o}$ - 1s3 d $^3\mathrm{D}$ | \mathbf{L} | 0.6161 | 0.6102 | 0.7100(8) | 0.7070(8) |
| | V | 0.6085 | | 0.7012(8) | |
| 1s3 s $^3{\rm S}$ - 1s3 p $^3{\rm P^o}$ | \mathbf{L} | 0.9126 | 0.8914 | 0.1114(7) | 0.1074(7) |
| | V | 0.8693 | | 0.1061(7) | |
| 1s3 p $^3\mathrm{P^o}$ - 1s3 d $^3\mathrm{D}$ | \mathbf{L} | 0.1095 | 0.1120 | 0.1179(5) | 0.1292(5) |
| | V | 0.1055 | | 0.1136(5) | |

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