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# Radiative cascades from doubly excited He states

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Resonance profiles of spectroscopically resolved radiative transitions from $(2pnd)^1P^o$, $(sp,2n+)^1P^o$, and $(sp,2n-)^1P^o$ doubly excited He states with $n=3,\ldots,7$ were investigated. The $(2p3d)^1P^o$–$(1s3d)^1D^e$–$(1s2p)^1P^o$–$1s^2\,{}^1S^e$ cascade transitions were observed using photon induced fluorescence spectroscopy in the vacuum ultraviolet and the visible spectral range. The $(2pnd)$ levels populate predominantly $(1snd)^1D$ levels, while for the $(sp,2n+\text{or}-)$ levels, $n^1S$ levels are populated as well. The experiments demonstrate that further quantitave branching ratio measurements are needed in order to test matrix elements for radiative transitions from He**.

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The He atom is of general interest, as it is the fundamental neutral three-particle Coulomb system for the study of electron correlations. Photons are known to populate doubly excited states on resonances according to the dipole selection rules. Madden and Codling in their pioneering absorption experiments [1] observed two series of Beutler-Fano profiles. Synchrotron radiation facilities of the third generation allowed at last to resolve very high $n$ members and also to detect the $(2pnd)$ series [2]. The developement of the theoretical treatment of doubly excited He from the Sommerfeld orbits to the nowadays hyperspherical coordinate description has recently been reviewed by Tanner *et al.* [3]. These descriptions lead to classification schemes with quantum numbers for the angular $(K,T)$ and for the radial $(A)$ correlation. For the three resonance series leading in their limits to ionized He in the $N=2$ state, the traditional classification $(sp,2n+)$ for $A=+1$, $(sp,2n-)$ for $A=-1$, and $(2pnd)$ for $A=0$, introduced by Cooper *et al.* [4], is often used. Alternatively, the notations $(n,+1)$, $(n,-1)$, and $(n,0)$, respectively, are used here for brevity.

The Beutler-Fano profiles describe the coupling between autoionization and direct ionization. Radiative transitions from doubly excited states has been neglected until recently [5], when fluorescence radiation was detected undispersed by a channel-plate detector and by a channeltron in [5] and in [6], respectively. Fluorescence radiation as well as the formation of metastable He atoms were shown to be essential [6], with increasing resolution ($\geqslant 20\,000$) of the exciting radiation even series of $LS$—forbidden triplet doubly excited states were detected [7]. Detailed highly resolved spectra in the extreme ultraviolet (EUV) range around 30 nm from doubly excited He have only been measured by using a microwave discharge for excitation [8]. Only a few transitions from $(sp,2n-)$ states of $^1P^o$ symmetry and no population of the $(sp,2n+)$ states have been observed.

A spectroscopic analysis of the so far undispersed fluorescence is very important for a sensitve test of the description of physical processes governing the production and decay of doubly excited He. Transition elements that determine the fluorescence intensities are given by the overlap of the wave functions in the upper and the lower states and thus test different parts of the wave functions with respect to transition energies. Using linear polarized radiation for excitation the states of $^1P^o$ symmetry with $M_L=0$ are populated exclusively, yielding in principle additional detailed information about their radiative decay via the polarization of the fluorescence radiation.

In the following, we present transitions and relative intensities observed in a spectroscopic investigation. Further experiments will aim at the relative probabilities of single branches and at cross sections for a determination of fluorescence yields. These studies are important steps towards the complete description of the fundamental He atom.

A cascade of transitions can be expected, with first the inner $2p$ electron populating $(1snl)$ states of the helium atom. Excited states with electrons in $ns$ and $nd$ orbits will decay through $(ns,nd\text{-}2p)$ transitions that finally lead to a $(2p\text{-}1s)$ transition (Fig. 1). $np$ electrons undergo dominantly $(np\text{-}1s)$ transitions. Their intensity is very difficult to measure due to the strong resonance imprisonment [9]. The spectra of singly excited He and also the branching ratios for transitions from one upper level to lower levels are well-known [10]. The energies of the transitions between the doubly and singly excited He states can be calculated quite accurately from the resonance energies and the binding energies of the He electrons. Alternatively, doubly excited states can populate radiatively $2p^2$ states that decay through successive $(2p\text{-}1s)$ transitions.

In the present experiments, we apply the photon-induced fluorescence spectroscopy developed by us in a series of investigations of the photon-induced two-electron excitations of the rare gas valence shells [11,12], and references therein. Linearly polarized photons with energies around 64 eV from the undulator U125/1-PGM beamline of BESSY II are fo-

 

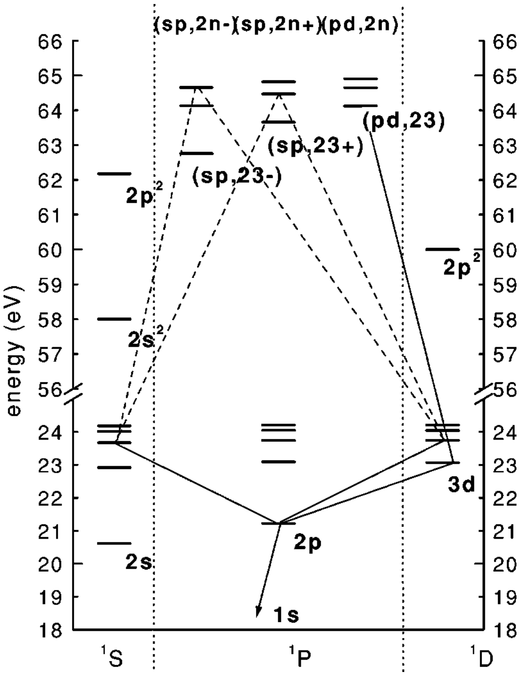


FIG. 1. He** and He* term diagram. Observed transitions indicated exemplarily by full lines.

cused into a differentially pumped gas target containing He at a pressure of 100 $\mu$bar. Two 1-m-normal-incidence spectrometers are oriented with their optical axis under 90° with respect to the photon beam direction. The photon beam with cross section dimensions of 50 $\mu$m×100 $\mu$m replaces the entrance slit of the spectrometers. One of them, observing the emitted fluorescence perpendicular to the electric-field vector of the undulator radiation, is equipped with a 600-lines/mm grating blazed at 300 nm and a position sensitive detector for the visible range. The second spectrometer uses a 1200-lines/mm grating blazed at 80 nm and an open microchannel-plate as a position sensitive detector for the vacuum ultraviolet (vuv) range. A small voltage of 10 V is applied to the entrance diaphragm of the target chamber in order to attract photoions. Their current intensity, scanned in parallel to the fluorescence signals as function of the primary-photon energy, displays the well-known resonance profiles.

In a first short test run, we measured vuv spectra emitted when the exciting photon energy was fixed either to the (3,0) resonance at 64.118 eV or to 66 eV. In this experiment, the spectrometer that had its optical axis perpendicular to the electric-field vector was equipped with the detector for the vuv. A line at 30.21 nm was observed in second order that was identified tentatively in [8] as belonging to the $(2p3d)^1P$–$(1s3d)^1D$ transition [Fig. 2(a)]. Our result agrees with this identification. Weaker features in Fig. 2(a) indicate a population of the $(1s4d)^1D$ level, too. Off resonance this line was absent, as well at 66 eV [Fig. 2(b)]. At this energy the He$^+(2p\text{-}1s)$ transition becomes energetically allowed and was observed. So is the He$(2p\text{-}1s)$ transition, the final member of the radiative cascade from the

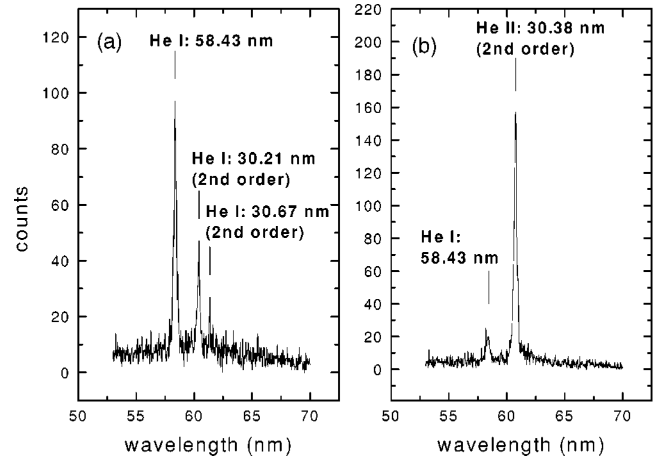


FIG. 2. VUV-fluorescence spectra for fixed (a) 64.118 eV [on the $(2p3d)$ resonance] and (b) 66 eV exciting-photon energy.

$(2p3d)^1P$ level [Fig. 2(a)]. Its observation at 66 eV results from impact excitation by photoelectrons.

In a second dedicated beam time fluorescence spectra scanned as a function of the exciting-photon energy across the He** resonances were registered. Transitions in the spectral range of the visible were studied in first place, since due to the low EUV sensitivity of the spectroscopic normal incidence mode no lines around 30 nm could be scanned. In Fig. 3 the intensity of the $(1s3d)^1D$–$(1s2p)^1P$ transition at a wavelength of 667.8 nm as function of the energy across the (3,0) and $(4,-1)$ resonance is shown. The $(1s3s)^1S$–$(1s2p)^1P$ transition at 728.1 nm was out of the spectral range of the spectrometer. Moreover, Fig. 3 contains the total ion yield and the intensity of the $(1s2p)^1P$–$(1s^2)^1S$ resonance transition. Both resonances are clearly separated. The intrinsicly very narrow (3,0) resonance allows a determination of the spectral width of the exciting radiation that amounts to 4 meV. Quantitative results for the He$(np\text{-}1s)$ resonance transitions are hampered by the falsification of the relative intensity through the strong resonance imprisonment.

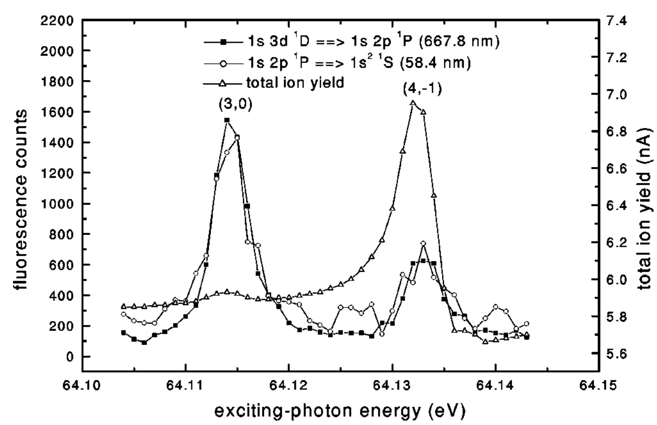


FIG. 3. Intensity of the $(1s3d)^1D$–$(1s2p)^1P$ transition at 667.8 nm, of the He I $(1s2p)^1P$–$1s^2\ {}^1S$ transition at 58.4 nm and of the ion yield as function of the photon energy across the (3,0) and $(4,-1)$ resonance.

Both transitions, the one in the VUV as well as the one in visible, are observed on both resonances with nearly the same intensity ratio as the undispersed intensities [5]. It is evident that also the $(4,-1)$ resonance populates the $3d$ level with an expected EUV line at 30.18 nm. This line was not observed in [8], but the presence of the $(1s3d)^1D$–$(1s2p)^1P$ line on the $(4,-1)$ resonance supports a forthcoming identification. No further fluorescence channels were investigated spetroscopically for the (3,0) and $(4,-1)$ resonances. As in [5], one notes the reversed order of magnitude of fluorescence intensity and ion yield .

In Figs. 4(a)–4(c) we have summarized the fluorescence intensities and the ion yield measured across the (4,0) and $(5,-1)$, the (5,0) and $(6,-1)$, and the (6,0) and $(7,-1)$ resonances. For all three resonances, the close lying $(1snd)^1D$–$(1s2p)^1P$ and $(1sns)^1S$–$(1s2p)^1P$ transitions with $n=4$, 5, and 6, respectively, were centered in the detection range. On the $(pd,2n)$ resonances the $(nd)^1D$ levels are predominantly populated, while on the $(sp,2(n+1)-)$ resonances the $(ns)^1S$ levels are also populated.Their relative contribution increases with $n$. Because of the limited beam time, the population of the $(ns)^1S$ levels on the corresponding $(sp,2n-)$ resonances was not investigated spectroscopically in the present study.

In a first estimation, the relative intensities of the $(1snd)^1D$–$(1s2p)^1P$ and of the $(1sns)^1S$–$(1s2p)^1P$ transitions as function of $n$ and $n+1$, respectively, were evaluated. Table I contains the relative peak values of the resonance profiles displaced in Figs. 4(a)–4(c), normalized to the exciting-photon flux. The different branching ratios in He* are taken into account, but a sensitivity correction for the detector photo cathode was neglected due to the limited investigated spectral range. We note an $n$-dependence close to $n^{-3}$ for the population of the He $(nd)^1D$ levels on the $(pd,2n)$ resonances. The $(ns)^1S$ levels show no such strong variation on the $(sp,2(n+1)-)$ resonances, while on the contrary the $(nd)^1D$ levels are again populated with a strong n-dependence.

These oberservations are discussed on the basis of the simple two-step model that relates the fluorescence intensity $I_n$ to the excitation cross section $\sigma_n$ and to the fluorescence yield according to

$$I_n=\sigma_n\times\Gamma_{fl,n}/(\Gamma_{fl,n}+\Gamma_{a,n}). \qquad (1)$$

The energy width $\Gamma_{fl,n}$ measures the radiative, and $\Gamma_{a,n}$ the autoionization decay rate. The relative peak values in Table I result from the product of $\sigma_n$ and the convolution of the natural width of the double excited states with the experimental width. A derivation of $\sigma_n$ even on a relative basis is not within the scope of the presented spectroscopic study. Nevertheless, a relation $\sigma_n\sim n^{-3}$ for the $(2p,nd)$ resonances can be concluded from Eq. (1), based on two approximations: $\Gamma_{fl}=\Gamma_{2p}$, with $\Gamma_{2p}\approx6.6$ $\mu$eV denoting the natural width of the $2p$ level in $He^+$. Secondly, $\Gamma_a\ll\Gamma_{fl}$ follows from calculations [13]. In consequence, $I_n\sim\sigma_n$ and $I_n$ is proportional to the measured fluorescence intensity. The situation is more complex for the $(sp,2n-)$ resonance, since $\Gamma_{2p}$ and $\Gamma_a$ are comparable for the considered $n$ values. More-

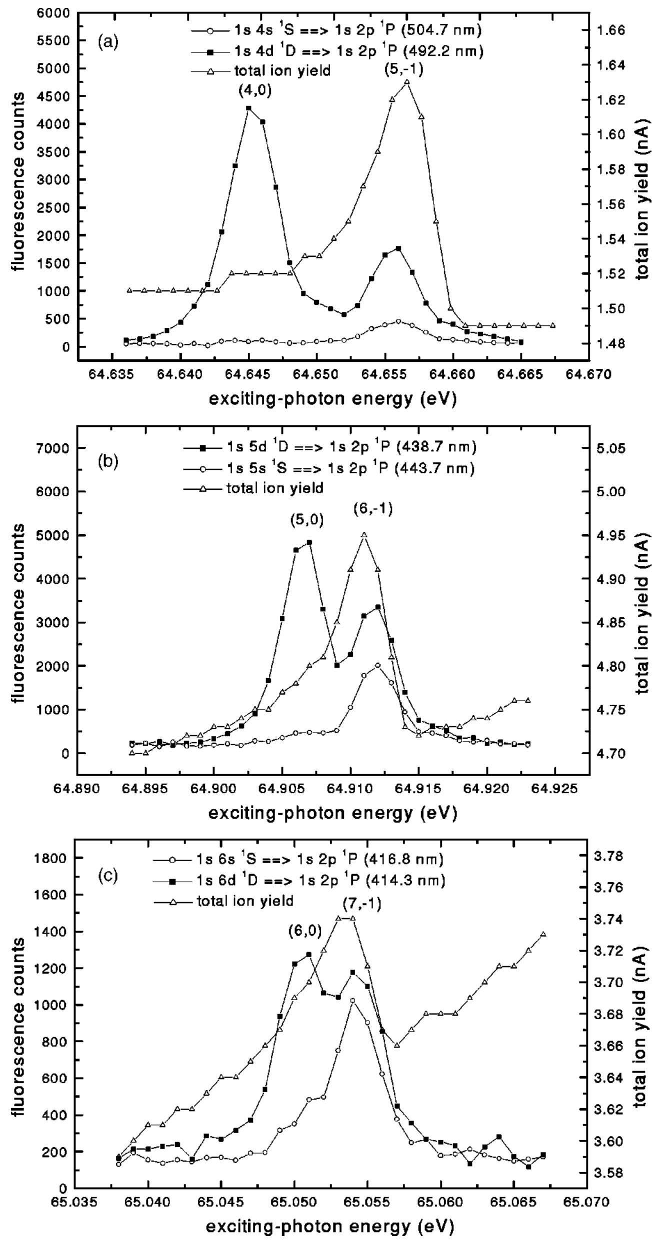


FIG. 4. Fluorescence intensities (and ion yield) of the (a) $(1s4d)^1D$– and $(1s4s)^1S$–$(1s2p)^1P$ transitions on the (4,0) and $(5,-1)$ resonances, (b) $(1s5d)^1D$– and $(1s5s)^1S$–$(1s2p)^1P$ transitions on the (5,0) and $(6,-1)$ resonances, and (c) $(1s6d)^1D$– and $(1s6s)^1S$–$(1s3p)^1P$ transitions on the (6,0) and $(7,-1)$ resonances, as function of the photon energy.

over, branchings of the radiative decay mode of the $(sp,2n-)$ levels have to be considered. Their importance is demonstrated by the present study, while for a quantitative interpretation of observable line intensities a wider range of transitions has to be studied.

Finally, we report the peak ratio of the $(1sns)^1S$–$(1s2p)^1P$ and $(1snd)^1D$–$(1s2p)^1P$ transitions observed on the $(sp,2n+)$ resonances that we studied for $n=4$ and $n=6$: The He I $ns$ levels are populated by a factor of 5 stronger than the $nd$ levels. The signal of the $(1s,4d)^1D$–$(1s2p)^1P$ transition on the $(sp,24+)$ resonance

TABLE I. Relative fluorescence intensities for $(1snd)^1D$–$(1s2p)^1P$ and $(1sns)^1S$–$(1s2p)^1P$ transitions ($n=4,5,6$).

| | $4s$ | $5s$ | $6s$ | $4d$ | $5d$ | $6d$ |
|---|---|---|---|---|---|---|
| (4,0) | | | | 100 | | |
| $(5,-1)$ | 14 | | | 43 | | |
| (5,0) | | | | | 40 | |
| $(6,-1)$ | | 22 | | | 22 | |
| (6,0) | | | | | | 13 |
| $(7,-1)$ | | | 14 | | | 12 |

is much lower than the corresponding signal on the (4,0) resonance. A comparison with the signals related to the $(sp,24-)$ resonance has to be postponed due to the lacking knowledge about the $(1s4s)^1S$ level population on the $(sp,24-)$ resonance.

In summary, the first spectroscopic analysis of the radiative decay of doubly excited He states resulting from single-photon absorption has been carried out. Emphasis was given to the spectral range of the visible. Transitions from $(sp,2n+)$, the $(sp,2n-)$, and the $(2pnd)$ resonances with $n=3,\ldots,7$ were observed. The signal rates on the $(2pnd)$ resonances dominate in agreement with the undispersed observations described in [5] and [6]. These doubly excited states populate in the first transition of their decay cascade $(1snd)^1D$ states of He I, while population of the $(1sns)^1S$ states is rather weak. Accordingly, the $(2pnd)$ resonances still have an appreciable amount of independent-electron character. To the contrary, $(sp,2n+)$ states populate preferentially $(1sns)^1S$ states. For the $(sp,2n-)$ states, a dependence of the branching ratio on $n$ was observed. This investigation has to be continued by efforts to measure accurate branching ratios for a wider range of transitions, to study the spectral range of the EUV around 40 eV, and to make use of the ability to measure the angular distribution of the fluorescence radiation.

*Note added in proof*. Recently, a theoretical study of the radiative decay of doubly excited $^1P^0$ states of helium was published [14].

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