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Effect of electric fields on the decay branching ratio of $^1P^e$ doubly excited states in helium measured by time-resolved fluorescence

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We have measured the lifetimes of $^1P^e$ ($n=9-12$) doubly excited states in static electric fields (1–6 kV/cm) by observing the decay of the fluorescence signal as a function of time. The effects of the field on these helium states below the second ionization threshold are twofold: their excitation becomes possible due to the Stark mixing with the optically allowed $^1P^o$ series, and their lifetime is strongly modified by the opening of the autoionization channel, not accessible in zero field. Although the electric field represents only a tiny perturbation of the atomic potential, a substantial shortening of the lifetimes below 100 ps is observed. This is the simplest quantum system where the ratio of autoionization to fluorescence decay probability can be effectively controlled by an electric field of moderate strength.

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Easy to manipulate but still nontrivial to understand, the helium atom has been a subject of many experimental and theoretical studies from the early days of quantum physics ([1] and references therein). The fluorescence from low-lying singly excited states enabled the characterization of the Stark effect [2], and provided a definitive test of the quantum-mechanical treatment given by Schrödinger [3]. The fluorescence signal was also the first signature of helium doubly excited states [4]. Later on, the first photoabsorption spectra in the vuv region were recorded by using a tunable synchrotron source [5]. It immediately became clear that electrons in doubly excited states are strongly correlated; a single-configuration description fails to explain the data. Very narrow and weak resonances were detected in photoion yield [6] and confirmed the existence of three series of optically allowed $^1P^o$ states converging to the $N=2$ threshold: the (principal) + series, the − series, and the $2pnd$ series, usually denoted by $(0,1)^+$, $(1,0)^-$, and $(-1,0)^0$ correlation quantum numbers [7]. Over the last few years there has been renewed interest in the study of fluorescence from doubly excited states. Contrary to expectations, it was shown that the photon yield is far from negligible [8,9], and that it can provide a wealth of new information about the nature of doubly excited states that would be otherwise very difficult to obtain [10–13]. For example, the presence of $^3P^o$ and $^3D^o$ triplet states in photoinization spectra is masked by the strong Fano-like broadened signal of singlets and, unlike the ion signal, just below the threshold the resonant fluorescence signal is strong.

The properties of doubly excited states with even parity were studied by electron scattering, but only for the low-lying states of $^1S^e$ and $^1D^e$ symmetry [14,15]. Until recently, the states with $(-1)^{L+1}$ parity were seen only in fluorescence measurements of microwave discharges [16]; this is obvious below $N=2$ where the autoionization of these states is forbidden because of parity conservation. To excite these states simple photoabsorption from the helium ground state is not effective because again, the $2pnp$ $^1P^e$ $(0,1)^-$ series has the "wrong" parity and also the angular momentum exchange required for the other states is rather improbable in single-photon absorption. Exceptionally, the first term of the $^1D^e$ series has been detected in angle-resolved photoelectron spectra on account of the small quadrupole coupling with the photon field [17]. The solution to controlling the excitation is to set a homogeneous electric field **F** in the target and align it perpendicularly to the polarization **P** of the incoming light. This mixes some of the dipole-allowed $^1P^o$ character into the states and makes them visible in the ion as well as in the photon yield spectra. Exactly this approach was employed recently by Prince *et al.* [18] to observe the $n=7-10$ states of the $^1P^e$ series. Their intensity increased with the principal quantum number and also with the electric field strength. Deviations of the photon yield from the expected quadratic dependence on the field strength were reported and it was proposed that the missing intensity goes into the autoionization channel.

The only available measurements of photoions at very high electric field strengths up to 84.4 kV/cm cover the appropriate energy region [19], but they were done in the $\mathbf{F}\|\mathbf{P}$ geometry where the $^1P^e$ excitation does not occur. Another possibility to test the above proposition is to directly measure lifetimes of these states in electric fields by time resolved fluorescence. The total fluorescence widths of doubly excited states below $N=2$ are of the order of 1 μeV and are relatively stable in the moderate electric field [20]; almost always it is an (inner) $2p/2s$ electron which makes the transition into a singly excited state. The first directly measured lifetime was that of the $2p3d$ $^1P^o$ state, for which the fluorescence is a dominant decay channel. Lambourne *et al.* reported the value of 190±30 ps [21] and higher series members were also examined [22,23]. The zero-field lifetime of

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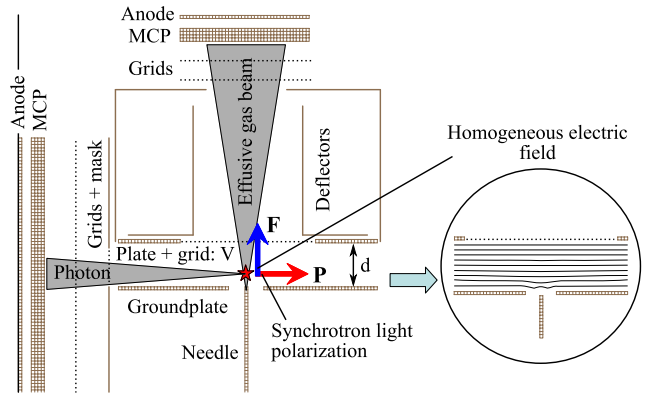

FIG. 1. (Color online) The schematic experimental setup showing calculated equipotentials in the target region.

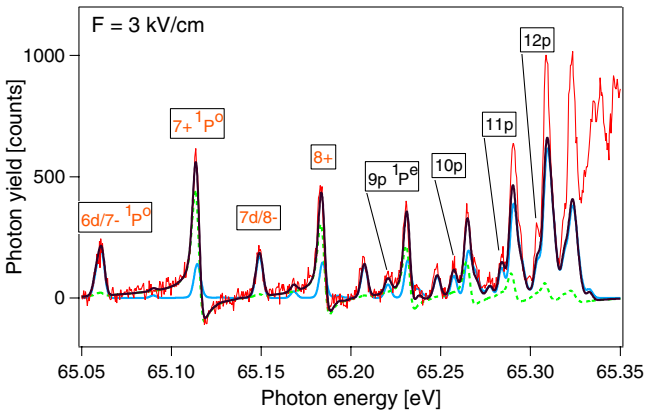

FIG. 2. (Color online) Measured vuv photon yield compared to the weighted sum (black) of the calculated primary fluorescence (gray, blue online) and the photoionization cross section (dashed line) convoluted with a 4 meV FWHM Gaussian.

$^1P^e$ states is expected to be close to 100 ps, the lifetime of $He^+(2p)$ ion, because the s orbitals cannot enter the configuration-interaction (CI) expansion of the wave function. On the other hand, the autoionization rate of the $^1P^e$ states is expected to increase sharply with the field strength because the largest admixture comes from the optically allowed and strongly autoionizing states with similar correlation pattern [24]—the principal $^1P^o$ series. So one should observe a shortening of the $^1P^e$ lifetime when the field strength is increased. A well-known effect of electric field is to modify lifetimes by redistributing oscillator strength among states decaying into the same channel, the $He^+(2s)$ quenching, for example. Below we report the ability of electric field to control the lifetime (and the excitation rate) of the upper state by drastically changing its branching ratio for decay into two different channels: fluorescence and autoionization.

We have measured the lifetimes of $n=9-12$ $^1P^e$ states in moderate electric field ranging from 1 to 6 kV/cm. The experiment was done on the secondary branch of the Gasphase beamline [25] at the Elettra synchrotron light source in the normal multibunch mode. In our case, the photons are most likely emitted in pairs, a quick-slow sequence where the primary photon is emitted with about three orders of magnitude shorter lifetime then the secondary one:

$$\mathrm{He}^{**}(n^1P^e) \xrightarrow{\tau\sim 100\ \mathrm{ps}} \mathrm{He}^{*}(1snp\,^1P^o) + \gamma_1(\approx 40\ \mathrm{eV}) \xrightarrow{100\ \mathrm{ns}} \mathrm{He}(^1S^e) + \gamma_2(\approx 20\ \mathrm{eV}).$$

To generate a homogeneous electric field in the interaction region, we have slightly modified the previous experimental setup [21,26]. The metastable signal is avoided by positioning photon detector with its axis perpendicular to the effusive gas beam (Fig. 1). The other detector is mounted for optimal detection of $He^*(^1S^e)$ atoms, but this signal will not be considered here. The interaction region has a lateral size of 0.3 mm and is at the crossing point of the gas beam and the focused synchrotron light beam. The gas needle can be moved vertically and is mounted perpendicular to the polarization **P** in the horizontal plane. The electric field is in between the grounded lower plate with a 5-mm-diameter hole and a planar mesh covering a 15×15 mm^2 aperture in the upper plate parallel to the first one. The plate dimensions are 40×40 mm^2 and they are separated for $d=6.00\pm 0.01$ mm so that fringe effects of the outer edges are negligible in the collision center. The tip of the needle (also grounded) is coplanar with the lower plate. A voltage V is applied to the upper plate. Although the field distortion due to the hole in the plate is strongly damped in the interaction region by the presence of the needle, the SIMION simulation predicts an effective electric field 10% lower than the nominal value V/d. The photon beam enters 2 mm above the lower plate and parallel to it. The background gas pressure of helium in the chamber was 7×10^{-6} mbar.

The main contamination of the fluorescence spectrum comes from the photon signal induced by photoions and photoelectrons. Accelerated by the electric field they hit the surface of nearby electrodes and generate secondary photons. The fluorescence lines of principal resonances therefore start to assume the asymmetric Fano line profiles when the field strength is increased. This unwanted effect is clearly visible in the fluorescence spectra taken by other groups [18,27] and is also present in ours (Fig. 2) although several precautions were taken to avoid it (colloidal graphite coating of the plates, grid for the upper electrode, detector front mask). Under such conditions it becomes difficult to estimate relative cross sections of different states from the photon yield. However, on the 2 ns time scale (bunch to bunch separation), this relatively slow parasitic signal contributes only to a constant background in time-resolved spectra.

Setting the incoming photon energy to the resonance position, the time difference is measured between the detection of a vuv photon ($h\nu \geqslant 10$ eV) and the synchrotron light pulse. The measurement relies on a time to amplitude converter (TAC) with the output digitized by a 2048-channel analog to digital converter. The start signal from the microchannel plate (MCP) detector is amplified by a 1 GHz bandwidth preamplifier and shaped by a constant fraction discriminator. The TAC stop signal is given by a multiple of the ring clock and is provided by a digital electronic unit synchronized to the radio frequency of the storage ring. Calibra-

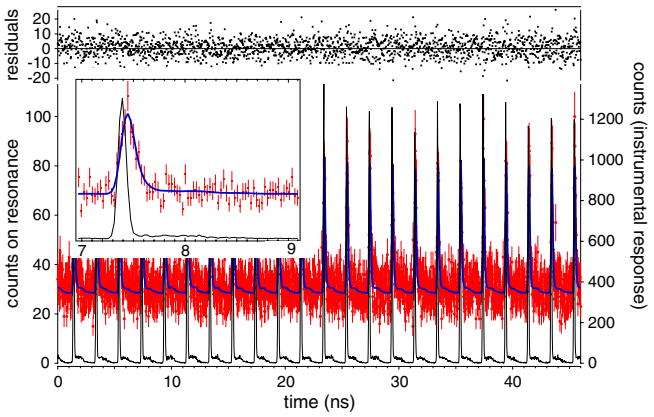

FIG. 3. (Color online) Time-resolved spectrum of $n=10\ ^1P^e$ state measured in a 3 kV/cm electric field perpendicular to the incoming polarization. The black (thin) line is instrumental response and the blue (thick) line is a fit giving the lifetime of 60 ± 10 ps. Black dots above are the fit residuals, and the inset shows the signal of one bunch out of 23.

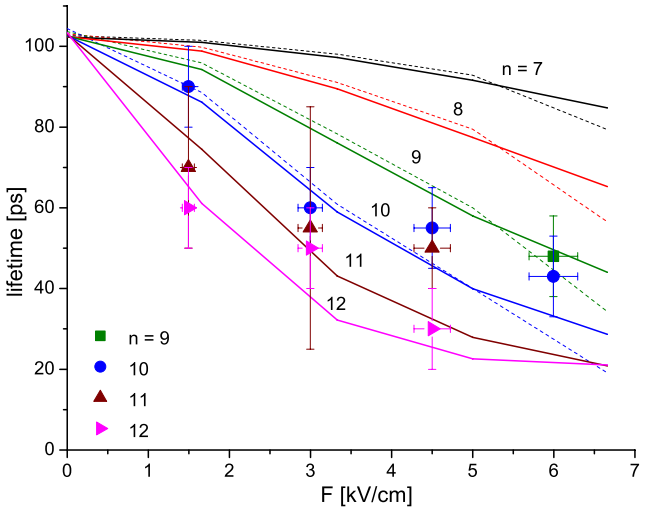

FIG. 4. (Color online) Comparison of measured lifetimes with the first-order perturbation (dashed) and the complex rotation (full line) predictions. Error bars include 5% uncertainty of the electric field strength.

tion with a bunch of exactly 2.001 ns period resulted in a 26.50 ps channel width averaged over the full TAC window. This was 50 ns long and contained the signal from 23 successive bunches. The instrumental response of the detection system corresponds to the prompt photon signal from 23 successive bunches with the same relative position in the storage ring. The experimental lifetime τ is obtained from the exponential decay function $e^{-t/\tau}$ convoluted with the instrumental response so that it best fits the signal of the time resolved spectrum. The cyclic convolution involved all the 23 bunches, and no summation of the signal from individual bunches was done (Fig. 3). Any nonlinear effects of the time scale were thus avoided and the efficiency of the measurement was substantially increased. Due to the repetition of 23 bunches the fluorescence signal is effectively sampled with about 1 ps time step since the bunch period is not an exact multiple of the channel width. This is very important when measuring the lifetimes which are predicted to be shorter than 100 ps.

For the procedure to work it is vital to obtain a reliable measure of the instrumental time response. A suitable signal was provided by photons scattered directly to the MCP when the tip of the needle was just slightly intercepting the incoming photon beam about 1 mm from its center [21]. The count rate was then not more than a few kilohertz and the dead time of the system was negligible. The full width at half maximum (FWHM) of the measured response is about 100 ps (Fig. 3) and is due to the 60 ps electron bunch width, the time response of electronics, and also the time spread of the signal on the MCP with 20 mm radius. A small structure delayed by about 0.5 ns from the moment of the peak intensity appears in the instrumental response, as well as in the signal-time spectrum. This may be due to photons hitting the MCP between the two channels. They generate electrons that return to MCP after being reflected by the grid potential.

TABLE I. Measured lifetimes (in picoseconds) of $n\ ^1P^e$ states in electric field. Asterisk denotes cases with smaller number of accumulated counts.

	n			
F (kV/cm)	9	10	11	12
1.5		90 ± 10	$70\pm20^*$	60 ± 10
3.0		60 ± 10	$55\pm30^*$	50 ± 10
4.5		55 ± 10	50 ± 10	30 ± 10
6.0	48 ± 10	43 ± 10		

Comparing instrumental response to the resonance signal, a time shift of 10 ps is typically observed. The shift depends on the exact vertical position of the needle and is due to the time delay of photons taking marginal trajectories through the 40 m long optical beamline system. A further consistency check was determination of $He^+(2p)$ lifetime whose zero field value is known to be 99.717 ± 0.075 ps [28]. From several fits we obtained the lifetimes in the range 97–103 ps with an error of ±5 ps only after allowing a Gaussian broadening of the instrumental response. This additional time broadening for approximately one channel originates from the instrumental instabilities which have larger effect during several hours long measurement of the ion (or resonance) fluorescence signal than in 5 minutes needed to measure the instrumental response. Using the improved fitting procedure (time shift + Gaussian time broadening) an excellent agreement was found also between the instrumental response and the time resolved signal of the principal $n=7\ ^1P^o$ state. Its lifetime is expected to be shorter than 1 ps, and the signal should therefore not differ from the exact instrumental response.

The measured lifetimes are displayed in Table I and in Fig. 4 where they are compared to the calculated values. According to first-order perturbation theory the lifetime of $^1P^e$ state varies as $1/(\Gamma_r+AF^2)$ where the fluorescence decay rate Γ_r is calculated in the zero field and A depends on the admixture and autoionization rate of the neighboring princi-

pal $^1P^o$ state [20]. Being aware of limitations of the first-order model we have also performed calculations based on exact consideration of bound-continuum coupling in the frame of the complex scaling method retaining the second order coupling of atom with the photon field ([29] and references therein). We have calculated the photoionization and total inelastic photon scattering cross sections, which are combined in Fig. 2 to explain the measured photon yield. The match of this nonrelativistic result in moderate electric field is very good up to $n=11$, and a similar result is expected for the calculated autoionization and fluorescence decay rates. Indeed, as seen in Fig. 4, the inverse sum of the two is in good agreement with our lifetime measurements.

In conclusion, the predicted trend of $^1P^e$ lifetime shortening is well verified. A similar effect is expected in the $\mathbf{F}\|\mathbf{P}$ geometry; the lifetimes of optically allowed $2pnd$ $^1P^o$ states should decrease in the electric field, due to the admixture of strongly autoionizing $^1S^e$ states. The autoionization to fluorescence decay branching ratio of doubly excited states with zero (or relatively small) autoionization rate can be easily changed by application of a static electric field; for the $n=10$ $^1P^e$ state the ratio changes from 0 to 1 when the field strength is increased from 0 to 6 kV/cm. The precise treatment of such systems requires an extension of the Fano formalism [30], which makes them an ideal test ground for unified treatment of the photon field coupled to a discrete state in the continuum. Time resolved fluorescence is a sensitive tool to study correlated doubly excited states in helium.

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