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Up-down asymmetry of the electrons ejected from barium $6p_{1/2}nk$ autoionizing states

J. Nunkaew and T. F. Gallagher
Department of Physics, University of Virginia, Charlottesville, Virginia 22904, USA

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We have measured the ejected electron signals from Ba $6p_{1/2}nk$ autoionizing Stark states, of $n = 28$ and 29, produced by linearly polarized laser excitation in weak electric fields. These states do not have well-defined parities and as a result do not lead to angular distributions which are up-down symmetric with respect to the laser polarization direction. The weakness of the electric field makes the observation of the up-down asymmetry of the ejected electrons possible. We observe that the electrons from Ba $6p_{1/2}nk$ autoionizing red states are ejected in the upfield direction, while the electron from Ba $6p_{1/2}nk$ autoionizing blue states are ejected in the downfield direction.

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I. INTRODUCTION

Electric dipole photoionization of atoms by light linearly polarized in the z direction usually leads to angular distributions of electrons which are symmetric in the z and $-z$ directions. For simplicity, we shall term the equivalence of the z and $-z$ directions as up-down symmetry. The up-down symmetry has its origin in the fact that the atoms are usually in a state of good parity, and electric dipole photoionization simply reverses the parity if an odd number of photons are absorbed and leaves it unchanged if an even number of photons are absorbed. The symmetry can be broken if there is interference between different orders of above-threshold ionization. Such interference arises in multiphoton ionization by two-color, phase-related fields and in multiphoton ionization by temporally short, single- to few-cycle pulses in which there is overlap of ionization processes of different orders [1,2]. The symmetry can also be broken if the electrons are ejected from atomic states which do not have good parity, such as the Rydberg Stark states formed in the presence of an electric field. These states are linear superpositions of zero-field states of even and odd parity. In a static field in the z direction, the Rydberg electron in a Stark state can be localized primarily on the $+z$ or $-z$ side of the atom [3]. Photoionization or autoionization of Stark states can be expected to result in superpositions of even- and odd-parity continua, with the result that the up-down symmetry of the electron ejection is broken. As a simple example, we show in Fig. 1 the classical trajectory of an electron initially in an $\ell = 3$ orbit aligned along the $-z$ axis which autoionizes as it passes near the core and leaves the atom in the $-z$ direction. Here ℓ is the orbital angular momentum of the electron. In a zero-field ℓ state, electron orbits along the $+z$ and $-z$ axes are equally likely, resulting in no up-down asymmetry of the ejected electrons. However, if the electron's orbit is on the $-z$ side of the atom, as in Fig. 1 or in a Stark state, the electron is ejected preferentially in the $-z$ direction. While the existence of the up-down asymmetry in the photoionization and autoionization of atomic states which do not have a well-defined parity, such as the Stark states, seems obvious, it has not, to our knowledge, been observed. The impediment to its observation is that to form the Stark states generally requires such strong fields that it is impossible to determine the direction in which the electrons have been ejected from the atoms.

Here we report the observation of the up-down asymmetry of electrons ejected in the autoionization of Ba $6p_{1/2}nk$ Stark states in electric fields of less than 10 V/cm. We choose the label k so that a k state is labeled by the zero-field ℓ state to which it is adiabatically connected. We use atomic units, unless specified otherwise. In the sections which follow we describe our experimental approach, present our observations, and discuss their implications.

II. EXPERIMENTAL APPROACH

In the experiment, Ba atoms from a heated oven are collimated into a 0.5-mm-diameter beam, which passes between two copper plates 1.2 cm apart inside a vacuum chamber at a pressure of $\sim 10^{-7}$ torr. Laser excitation of the atoms occurs between the plates in a field $E_0 \sim 10$ V/cm. As shown in Fig. 2(a), ground-state Ba $6s^2\ {}^1S_0$ atoms are excited to the Ba $6s6p\ {}^1P_1$ state and then to the $6s(n+3)d\ {}^1D_2$ state by sequential 553- and ~419-nm, 5-ns laser pulses in an electric field of less than 10 V/cm. As shown in Fig. 2(b), starting 50 ns after the first two laser pulses, the electric field is reduced linearly to zero in ~20–30 μs. In the first 3 μs of the field ramp, a microwave field resonant with the $6s(n+3)d\ {}^1D_2$ to $6snk$ transition at the field $E_R < E_0$ is present, and as the field ramp passes through E_R, the atoms undergo adiabatic rapid passage from the $6s(n+3)d\ {}^1D_2$ state to the $6snk$ state. At a chosen time, and therefore field, after the microwave pulse, atoms in the $6snk$ state are excited to the autoionizing $6p_{1/2}nk$ state by a third 5-ns-long laser pulse at a wavelength of ~493 nm. This excitation, the isolated core excitation (ICE), is one in which the ion core is excited while the outer nk electron remains a spectator [4]. The Ba $6p_{1/2}nk$ states are degenerate with the Ba $6s_{1/2}\epsilon k'$ and the Ba $5d_{3/2}\epsilon k''$ continua. The autoionization thus leads to Ba$^+$ $6s_{1/2}$ or $5d_{3/2}$ ions and ejected electrons with energies of 2.5 or 1.9 eV, respectively. The $6p_{1/2}nk$ atoms autoionize rapidly, during the third laser pulse, and the electrons resulting from autoionization are forced through a 1-cm diameter hole in the top plate and fly to the 1.8-cm diameter microchannel plate (MCP) detector 3.2 cm above the top plate.

In a field of 5.83 V/cm, the 2.5-eV electrons which are ejected up reach the detector ~15 ns before those which are ejected down, and the two signals are resolved in time

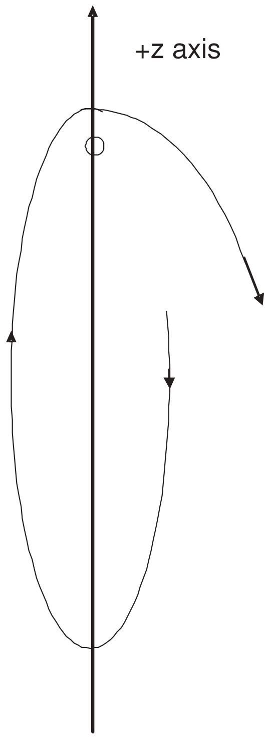
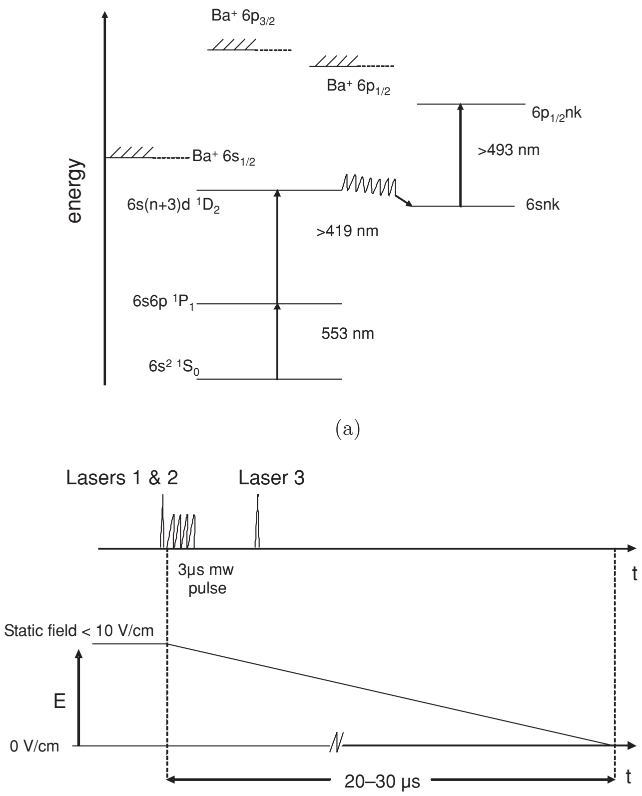


FIG. 1. The classical trajectory of an electron initially in an $\ell = 3$ orbit aligned along the $-z$ axis which autoionizes as it passes near the core and leaves the atom in the $-z$ direction.

as shown in Fig. 3(b). The up-down asymmetry is observed in the difference in the number of electrons ejected up and down. A slight complication is that the collection efficiency for electrons ejected up is greater than for electrons ejected down. However, the relative collection efficiencies can easily be calculated from the geometry of the apparatus. If θ is the angle at which electrons are ejected relative to the vertical axis, we define θ_{up} and θ_{down} such that all angles $0 < \theta < \theta_{\text{up}}$ and $\pi > \theta > \theta_{\text{down}}$ define the angular regions of acceptance for electron detection. Since the electrons ejected down travel a longer time, $\pi - \theta_{\text{down}} < \theta_{\text{up}}$. In Fig. 3(a) we show the trajectories of atoms ejected with equal angles relative to the $+z$ and $-z$ vertical axes. As shown, the electrons ejected down travel farther horizontally, which is why $\pi - \theta_{\text{down}} < \theta_{\text{up}}$. In Fig. 4, the calculated values for θ_{up} and $\pi - \theta_{\text{down}}$ for 2.5 eV electrons are plotted as a function of electric field. While raising the field reduces the difference in the collection efficiencies, it also reduces the difference in the flight times shown in Fig. 3(b), making it harder to temporally separate the up and down signals.

Microwaves in the 75–115 GHz frequency range are required, and we generate them in the following way. The continuous-wave 13- to 20-GHz output of a Hewlett-Packard 83 550A sweep oscillator is formed into 3-μs pulses by using a General Microwave DM862B switch. The microwave pulse is frequency doubled by a Phase One SX40-220 active doubler and travels through a WR28 waveguide into the vacuum chamber where it is tripled by a Pacific Millimeter W3WO passive tripler. The high-frequency microwave pulses then travel through a WR10 waveguide and propagate from a WR10 microwave horn to the region between the field plates. The lasers and microwaves are linearly polarized vertically, in the same direction as the static field, so we always have states of $|m| = 0$. We use Helmholtz coils to reduce the Earth's magnetic field to less than 50 mG. To produce the temporal profile of the field shown in Fig. 2(b), we use a Hewlett-Packard 8112A pulse generator to apply a negative voltage to the bottom plate. The voltage is initially from -7 to -12 V, and its magnitude is reduced linearly in time to zero in 20–30 μs. The top plate is grounded.



(a)

(b)

FIG. 2. (a) Energy levels and (b) timing sequence used in this experiment. Lasers 1 and 2 (553 and 419 nm) are in a static field of less than 10 V/cm. 50 ns later, the field is then reduced at the same time as the 3-μs-pulse microwave is applied. The field is reduced to zero in 20–30 μs. The third laser is typically fired $\sim$2 μs after the microwave pulse.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Symmetric case, Ba $6p_{1/2}22s$ autoionizing state

To verify that we are correctly accounting for the difference in collection efficiencies of electrons ejected up and down, we observed the electrons from the Ba $6p_{1/2}22s$ state in weak electric fields, $\leqslant 16.67$ V/cm. In such fields, the the wave function of the $6p_{1/2}22s$ state remains up-down symmetric to within 5% due to the large quantum defect, $\delta_s = 3.58$ [5]. As a measure of the up-down asymmetry we define R, the ratio of the time-integrated up electron signal to the total signal. In general, R is given by

$$R = \frac{\int_0^{\theta_{\text{up}}} P(\theta,\phi)\sin\theta\, d\theta}{\int_0^{\theta_{\text{up}}} P(\theta,\phi)\sin\theta\, d\theta + \int_{\theta_{\text{down}}}^{\pi} P(\theta,\phi)\sin\theta\, d\theta}, \quad (1)$$

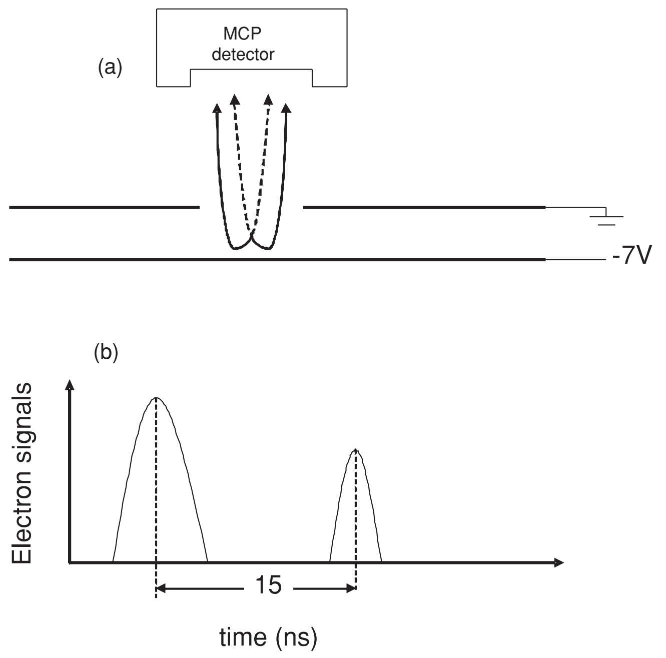


FIG. 3. (a) Arrangement for detecting the angular distribution of electrons ejected from Stark states. Trajectories are shown for electrons ejected at the same angle relative to the positive and negative z axes. (b) Expected time-resolved electron signals. The earlier signal is due to electrons ejected upward to the detector, and the later signal is due to electrons ejected away from the detector.

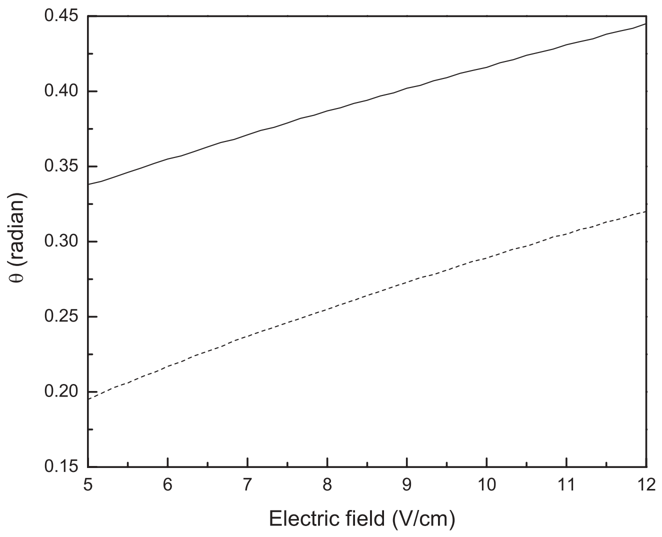


FIG. 4. θ_{up} (solid line) and $\pi - \theta_{\mathrm{down}}$ (dashed line) for 2.5-eV electrons as a function of electric field. Note the offset of the vertical scale.

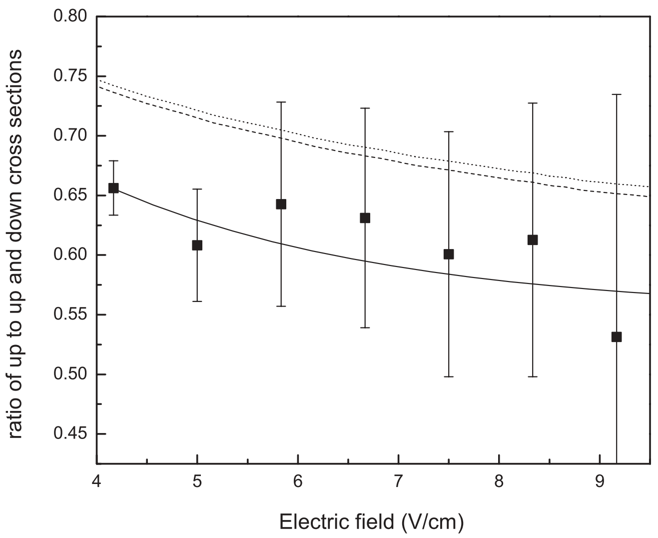


FIG. 5. The ratios of up to the total up and down integrated electron signals (R) of $6p_{1/2}22s$ in electric fields. (■) represent the experimental values of R. The broken line (—) represents calculated values of weighted average R of 1.9- and 2.5-eV electrons with $\beta = 1.5$ and 1.05, respectively, from Kachru *et al.* [7]. Note the offset of the vertical scale arises from the fact that we fit the data in a way which systematically gives values of R which are low. The dotted line ($\cdots$) represents calculated values of R with $\beta = 0$. The solid line (—) is the smooth curve through the experimental values.

where θ and ϕ are the polar and azimuthal angles at which the electron is ejected, and $P(\theta,\phi)$ is the probability of ejection in this direction. We assume that the probability of ejection does not depend on ϕ. We define R in this way, since it is an easily measured number and contains no assumption about the angular distribution. It is straightforward to calculate R if θ_{up}, θ_{down}, and the angular distribution of the ejected electrons are known. For the Ba $6p_{1/2}22s$ state, we know all of these parameters, and R is given by [6]

$$R = \frac{\int_0^{\theta_{\mathrm{up}}} [1+\beta P_2(\cos\theta)] \sin\theta \, d\theta}{\int_0^{\theta_{\mathrm{up}}} [1+\beta P_2(\cos\theta)] \sin\theta \, d\theta + \int_{\theta_{\mathrm{down}}}^{\pi} [1+\beta P_2(\cos\theta)] \sin\theta \, d\theta}, \tag{2}$$

where β is the asymmetry parameter, $P_2(\cos\theta)$ is the second-order Legendre polynomial, θ_{up} is the angular acceptance of upward ejected electrons, and θ_{down} is the angular acceptance of the downward ejected electrons. In a field higher than $\sim$3 V/cm, we cannot resolve the 1.9- and 2.5-eV ejected electron signals. Therefore, the calculation is carried out using weighted average R of 1.9- and 2.5-eV electrons with $\beta = 1.05$ and 1.5, respectively, for $6p_{1/2}22s$ from Kachru *et al.* [7]. The calculated and measured values of R are shown in Fig. 5. From Fig. 5 it is apparent that the experimental and calculated values of R agree reasonably well, although the measured values of R are systematically slightly lower. They are lower because we fit the observed signals to a sum of two Lorentzians, one for the electrons ejected up and one for the electrons ejected down. The experimental signals are not perfect Lorentzians but have tails at a later time, so our procedure gives values of R which are systematically too small. Since we use the measured values of R shown in Fig. 5 to define symmetric up-down ejection of electrons, this small systematic error does not affect our final results. The uncertainties shown in Fig. 5 are those obtained in fitting the data. Finally, we note that a completely isotropic distribution of ejected electrons ($\beta = 0$) would lead to a larger R for the same θ_{up} and θ_{down}, and, an angular distribution more strongly peaked along the $+z$ and $-z$ axes would lead to R closer to 1/2.

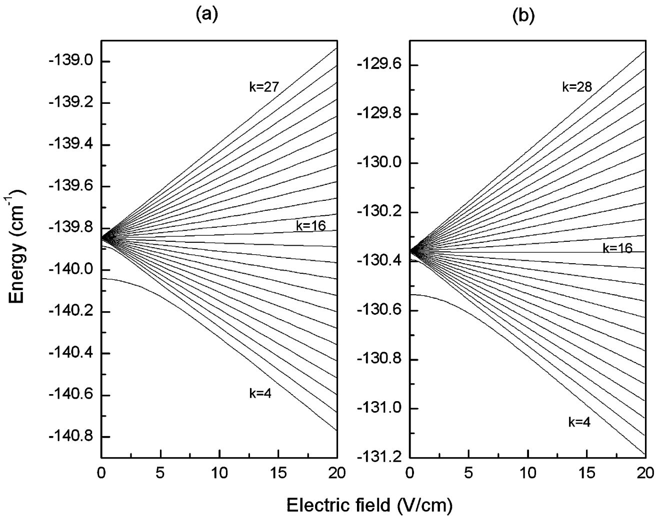
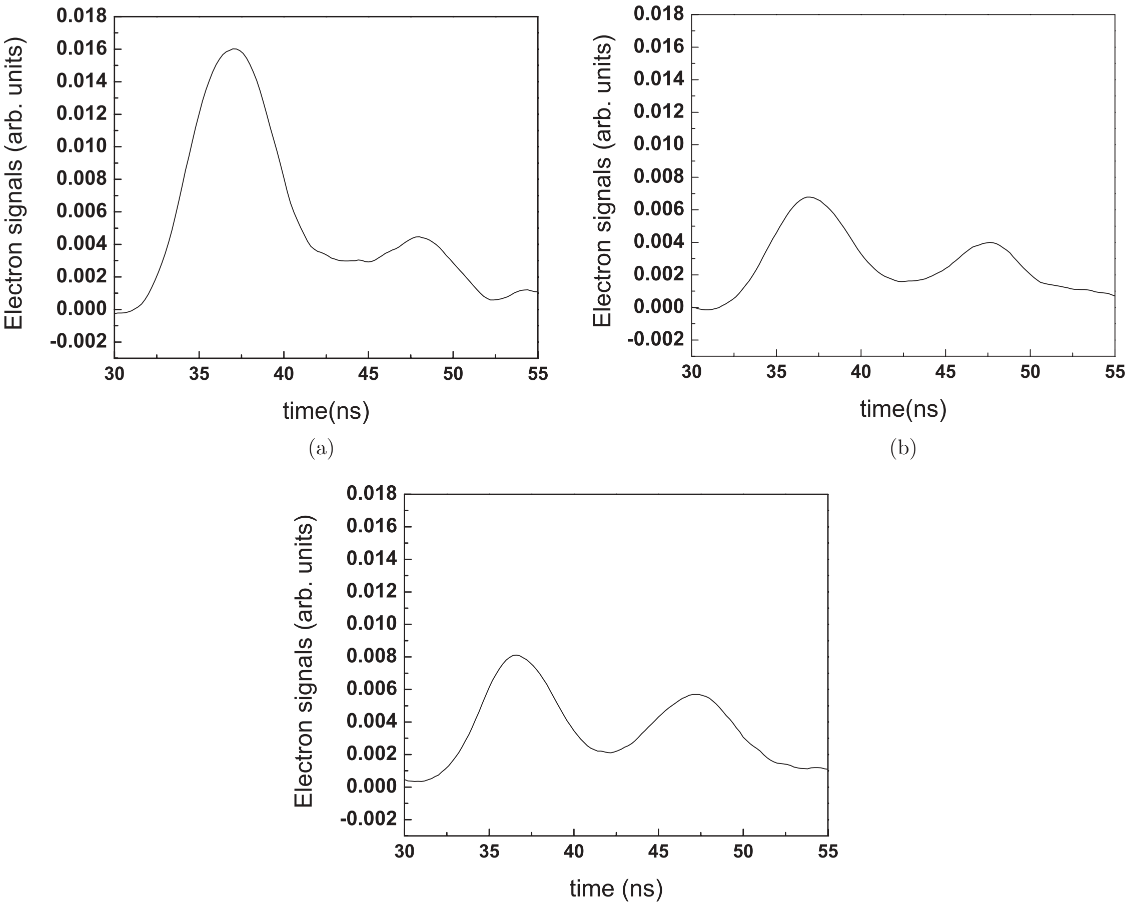


FIG. 6. Calculated Stark map of the (a) $6p_{1/2}28k$ and (b) $6p_{1/2}29k$ states, $4 \leqslant k \leqslant n-1$.

B. Ba $6p_{1/2}nk$, $n = 28$ and 29, $5 \leqslant k \leqslant n-1$

We have observed the ejected electrons from Ba $6p_{1/2}nk$ states of $n = 28$ and 29, $5 \leqslant k \leqslant n-1$ in electric fields less than 10 V/cm. For both n, $k = 15$ and 16 are in the middle of the Stark manifold as shown in Fig. 6. These states have small Stark shifts, and the wave function of the Rydberg electron is not localized above or below the ion, and we would expect the electrons to be ejected with up-down symmetry. States of $k < 15$ are red states, and the wave functions are localized on the upward side of the atom, while states of $k > 16$ are blue states, and the wave functions are localized on the downward side of the atom. We intuitively expect electrons from the red states to be ejected preferentially in the up direction, while ejection from the blue states should be preferentially in the down direction. Figure 7 shows the ejected electron signals as a function of time in $|E| = 7.9$ V/cm from Ba $6p_{1/2}28k$ states of $k = 5$, 16, and 21, which are red, intermediate, and blue states, respectively. In the figure, $t = 0$ corresponds to the time of the third laser pulse, so the times are the flight times of the electrons. In Fig. 7, the earlier peak is due to the electrons ejected up, and the later peak is due to the electrons ejected down. In Fig. 7(a), for the Ba $6p_{1/2}28k$, $k = 5$ state, there is clearly much more signal in the early peak at $t = 37$ ns than in the later peak at 48 ns; i.e., more electrons ejected in the upward direction are detected. In Fig. 7(b), for the Ba $6p_{1/2}28k$, $k = 16$ state, the earlier electron signal is only slightly larger than the later one. Finally, in Fig. 7(c) for the Ba $6p_{1/2}28k$, $k = 21$ state, the earlier and later signals are comparable in amplitude, but the time-integrated later signal is larger. Although we have not

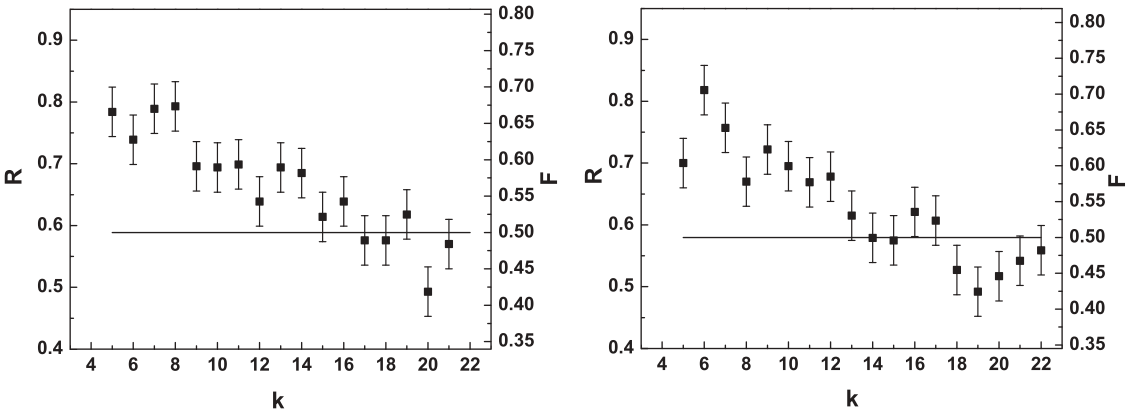


(a)

(b)

(c)

FIG. 7. Ejected electron signals from (a) $6p_{1/2}28k$, $k = 5$, (b) $6p_{1/2}28k$, $k = 16$, and (c) $6p_{1/2}28k$, $k = 21$ autoionizing states as a function of time in $|E| = 7.90$ V/cm.



(a) (b)

FIG. 8. R, the ratio of up to the total up and down integrated electron signals, as a function of k for the $6p_{1/2}28k$ states (■) in (a) $|E| = 7.10$ and (b) $|E| = 7.90$ V/cm. The line is the R value for up-down symmetric ejection of electrons from Ba $6p_{1/2}22s$ states obtained from the experimental values in Fig. 5. The values of R above (below) the lines indicate electron ejection in the up (down) direction. F is the fraction of the electrons ejected up.

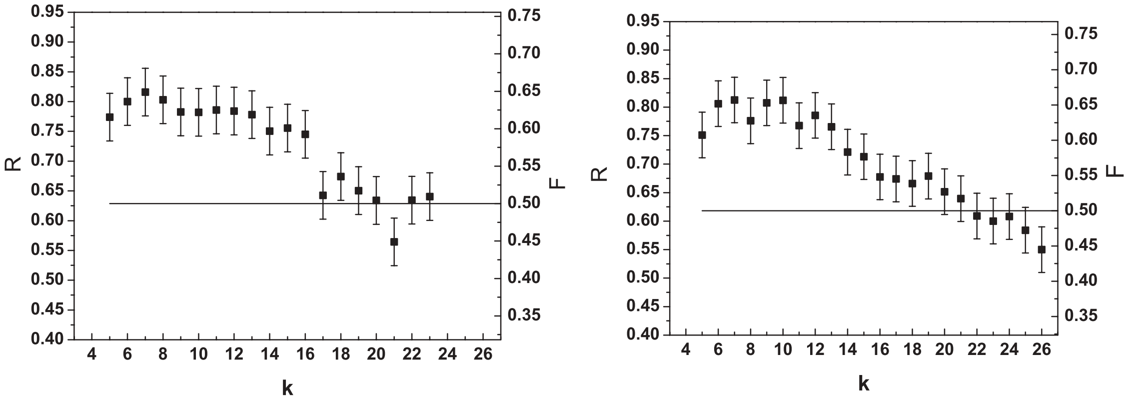
taken into account the greater collection efficiency for atoms ejected up, it is already apparent that the red state preferentially ejects the electron in the upward direction and the blue state in the downward direction.

In Figs. 8 and 9 we show the measured values of R as a function of k in different electric fields. For reference, the lines in both figures show the values of R produced by the up-down symmetric ejection of electrons from the $6p_{1/2}22s$ state. The wave functions of Stark states in the center of the manifold suggest that electrons ejected from these states should have angular distributions not too different from those described by $\beta = 0$ or 1 [8]. Assuming this to be the case, from the solid line of Fig. 5, values of R above (below) the lines indicates electron ejection in the up (down) direction. We can approximately convert the measured values of R into the fraction of the electrons ejected up, F, in the following way. On the right-hand sides of Figs. 8 and 9 we have shifted the scales from the left-hand sides so the lines occur at $F = 0.5$ in all cases. In cases for which $F \sim 0.5$, F is the fraction of electrons ejected up, although for $F \sim 1$ or $F \sim 0$ this procedure is not necessarily correct. In sum, we conclude that Stark states of $k < 16$ autoionize by ejecting atoms in the up direction, and states of $k > 16$ autoionize by ejecting electrons in the down direction, in reasonable agreement with our naive expectation.

IV. DISCUSSION

The observed up-down asymmetry is not large, but it should not be. Our intuition came from the classical picture of Fig. 1, which is drawn for $\ell = 3$, in which case the electron is ejected preferentially on one side, the $-z$ side, of the atom. In higher angular momentum states, the electron does not come as near the core due to the centrifugal potential, and the electron is no longer ejected with as strong a preference for the $+z$ or $-z$ direction. In our experiment, we have studied the Stark states which are composed of states of $\ell \geqslant 5$ for which there is less asymmetry in the direction of the ejected electron.

From a quantum-mechanical point of view, the asymmetry arises from the interference between partial waves of even and odd ℓ. Autoionization rates of the Ba $6p_{1/2}n\ell$ states fall



(a) (b)

FIG. 9. R, the ratio of up to the total up and down integrated electron signals, as a function of k for the $6p_{1/2}29k$ states (■) in (a) $|E|$=5.02 and (b) $|E| = 5.43$ V/cm. The line is the R value for up-down symmetric ejection of electrons from Ba $6p_{1/2}22s$ states obtained from the experimental values in Fig. 5. The values of R above (below) the lines indicate electron ejection in the up (down) direction. F is the fraction of the electrons ejected up.

quickly with ℓ, roughly a factor of 5 per ℓ, so the interference terms cannot be too large.

Calculating the probability of ejection in a specific direction, $P(\theta,\phi)$, poses a theoretical challenge, and we hope these data will inspire a theorist to undertake it.

V. CONCLUSION

We have observed the up-down asymmetry in electrons ejected from an asymmetric atomic state, specifically from Ba $6p_{1/2}nk$ autoionizing Stark states. The red autoionizing states are more likely to eject the electrons in the upfield direction, while the blue autoionizing states are more likely to eject the electrons in the downfield direction.

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