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Multiphoton Ionization of Potassium Atoms in Femtosecond Laser Fields

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We study the multiphoton ionization of potassium atoms in 800 nm and 400 nm femtosecond laser fields. In the 800 nm laser field, the potassium atom absorbs three photons and emits one electron via one photon resonance with the $4p$ intermediate state with the help of the ac-Stark shift. The resonance feature is clearly shown as an Autler–Townes (AT) splitting and is mapped out in the electron kinetic energy spectrum. In a 400 nm laser field, although one photon resonance is possible with the $5p$ state, no splitting is observed. The different transition amplitudes between $4s$–$4p$ and $4s$–$5p$ explain the observed results. Due to the AT effect, an unexpected peak in the photoelectron energy spectrum that violates the dipole transition rule is observed. A preliminary explanation involving the spin-orbit interaction in the p state is given to account for this component. The observed AT-splitting in the electron kinetic energy distribution can be used as an effective method to calibrate the intensity of a laser field.

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Multiphoton ionization of atoms and molecules has been an active subject for almost a century.[1,2] The multiphoton ionization of an atom reflects both the characteristics of the laser pulse and the properties of the atom perturbed by the intense laser field, and thus constitutes a very favorable method for studying the response of an atom in the presence of an intense laser field.

Photoelectron spectroscopy coupled with the resonance-enhanced multiphoton ionization (REMPI) has proven to be a powerful tool for studying the photoionization dynamics of neutral atoms.[3] Kinetic energy and angular distributions of outgoing electrons are important physical quantities in the photoelectron spectroscopy. The kinetic energy distribution provides information about the internal state of the remaining ion core, while the angular distribution includes the partial wave character of outgoing electrons and the alignment of the resonant intermediate state.

Specifically, the multiphoton ionization of potassium atoms has been studied for around 30 years.[4] Initially the interest was focused on obtaining a complete measurement of the photoionization process including both information of transition amplitude and the involved phase.[5–7] Later, it was found that the outgoing photoelectron can be coherently manipulated by shaped femtosecond laser fields.[8–12] Interesting phenomena has been revealed, such as electron vortices.[13,14]

In this study, we compare the photoelectron momentum spectrum in the femtosecond laser field with different central wavelengths. The role of the involved resonantly coupled excited states on the final electron momentum spectrum is also discussed. An unexpected photoelectron kinetic energy spectrum that violates the dipole approximation is observed and a preliminary explanation which involves the spin-orbit coupling is given.

Experiments. We investigated the multiphoton ionization of potassium atoms in a femtosecond laser field with a velocity map imaging (VMI) detection system. The femtosecond laser used in our experiment is commercially available with a repetition rate of 1 kHz, pulse duration of around 50 fs (Coherent Libra-HE). We use either the fundamental of 800 nm or the frequency doublet of 400 nm. The laser is focused with a 275 mm lens and results in a beam diameter of around 40 μm in the interaction regime. The VMI detection system is home-built with a typical three-electrode geometry.[15–17] The potassium atom in an effusive beam are produced by heating up an oven containing pure bulk potassium metal to around 407 K. Experimentally, we overheat the oven for around half an hour and then reduce the temperature to the operation condition to stabilize the atomic beam. The oven is enclosed by a 1-mm-thick stainless steel plate with

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a 0.2 mm hole in the center. The atomic beam is confined transversely by a skimmer (2 mm in diameter) 5 cm downstream from the front plate of the oven. After the skimmer the atomic beam continues free flying for about 55 cm to the interaction spot where the laser beam hits the atoms with an angle of 90◦. Due to the long free-flight distance the atomic beam will spread considerably. Further confining the interaction volume and to shield the straight and the earth magnetic fields is realized by placing a 3-mm-thick permalloy tube around the VMI electrodes. A 1 mm slice on the magnetic shielding ensures that the interaction region along the propagation direction of the laser is less than 2 mm.

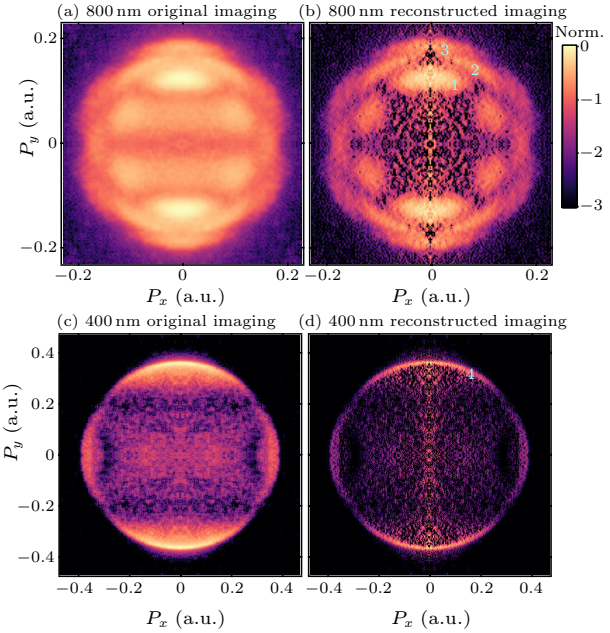

Fig. 1. Velocity map images of photoelectrons of potassium interacted with an 800 nm (a)–(b) and a 400 nm (c)–(d) femtosecond laser field. The left-hand column represents the original images while the right column are the 2D images after the Abel-inversion. The labeled numbers indicate the resolved peaks resulting from distinct ionization channels.

Results and Discussions. The main results of our experiment are the momentum spectra of photoelectrons generated from multiphoton ionization of potassium atoms in an 800 nm or a 400 nm femtosecond laser field. We keep the laser intensities below 3×10^{11} W/cm^2 such that the Keldysh parameter is larger than 1 (for 800 nm in the intensity range used in our experiment the Keldysh parameter is around 20, for 400 nm laser it is above 40). The electron momentum distribution is shown in Fig. 1, where only the low-energy parts are shown but not the above-threshold-ionization (ATI) parts. The left-hand column corresponds the original images obtained in the 800 nm (a) and 400 nm (c) laser fields, respectively. However, the right column is the reconstructed 2D images.

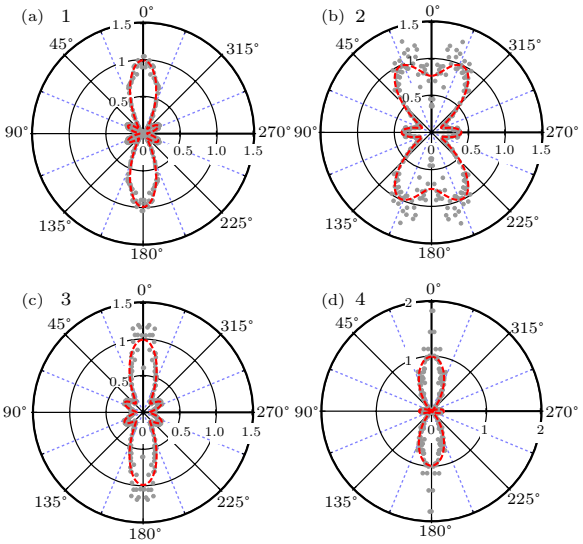

Fig. 2. Angular distribution of photoelectrons by radial integration of images shown in Fig. 1. The integration interval is around 0.1 eV, which reflects the typical FWHM of the kinetic energy distribution of each components. Labels 1–4 corresponds to the ones in Figs. 1(b) and 1(d) which indicate individual kinetic energy components. Grey points are the experimental results and the red lines correspond to different spherical harmonics; that is, $Y_3^0(\theta,\phi)$ for (a) and (c), $Y_3^{\pm1}(\theta,\phi)$ for (b), and $Y_2^0(\theta,\phi)$ for (d).

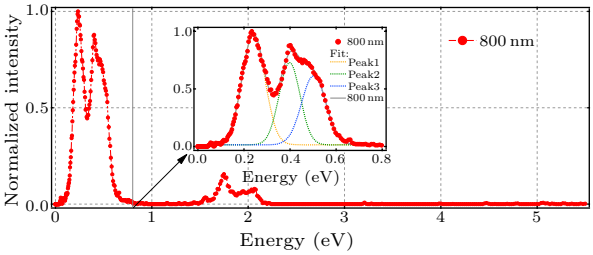

Fig. 3. Kinetic energy spectrum of the photoelectrons generated in an 800 nm femtosecond laser field with the intensity of around 1×10^{11} W/cm^2. Red dots are the experimental result after the angular integration. The yellow, green and blue dotted lines are the Gaussian fittings from which the information of the peak positions can be extracted.

The ionization potential of the potassium atom is around 4.34 eV. In the 800 nm femtosecond laser field the K atom absorbs at least three photons to get ionized. From the energy conservation law the kinetic energy of the ionized electron should be $kin_{\rm ele} = 3h\nu - I_{\rm p} = 3\times1.55-4.34 = 0.31$ eV. The $h\nu$ represents the single photon energy of the 800 nm laser, and the $I_{\rm p}$ is the ionization potential of the K atom. In the above formulae we do not take into account the effect of the ponderomotive energy because under the experimental conditions it amounts only a few meV. However, as shown in Fig. 1(b), instead of a single peak, three well resolved components labeled as peaks 1–3 are observed. The positions of those three peaks are

derived as 0.23 eV, 0.40 eV and 0.52 eV, respectively, after the angular integration of the reconstructed image. The results are shown in Fig. 3. To understand the origin of the three-separated peaks in the photoelectron momentum distribution we check the angular distribution of each peak and fit the results with spherical harmonic functions as shown in Fig. 2. Peaks 1 and 3 show identical angular distribution featured a $Y_3^0(\theta,\phi)$ spherical harmonic distribution. This coincides with the character of the three-photon ionization of the K atom initially prepared in the $^2S_{1/2}$ state. The kinetic energy separation of peaks 1 and 3 is around 0.29 ± 0.04 eV. This energy splitting is due to the well-known Autler–Townes effect which results from the resonance coupling between two states.[18] In our case, this resonance coupling is mediated by the ac-Stark shift of the ground $4s$ and the $4p$ state.[5,9,19] The ac-Stark shift of an atom in a static electric field can be expressed as

$$\Delta E_{\rm ac} = -\frac{1}{2}\alpha_0 E^2, \tag{1}$$

where α_0 is the state specified polarizability and E is the electric field strength. For potassium atoms, α_0 has the value of around 45 and 90 (in atomic units) for the $4s$ and $4p$ states, respectively, which gives rise the net ac-Stark shift of around -20 meV at the intensity of 1×10^{11} W/cm^2. The field free energy separation between $4s$ and $4p$ states is around 1.61 eV, which is around 50 meV blue-detuned from the single 800 nm laser photon energy. Our femtosecond laser has a duration of 50 fs and the bandwidth is of around 25 meV. Taken together, the ac-Stark effect will bring exactly one 800 nm photon into resonance with the $4s$–$4p$ transition. The resonance will split each involved level into two with the separation given by the Rabi frequency:

$$\Omega_{\rm rabi} = \frac{e\langle a|r|b\rangle E}{\hbar}, \tag{2}$$

where e is the elementary charge, $|a\rangle$ and $|b\rangle$ are the two coupled states, $\hbar$ is the reduced Planck constant; and $\langle a|r|b\rangle$ is the transition matrix element. The value of the transition matrix element can be found from Ref. [20]. By plugging this value into Eq. (2), we can obtain the Rabi frequency, which reflects the energy splitting of peaks 1 and 3 in the observed electron momentum distribution as 0.27 eV, well in agreement with the experimental result. The origin of the peak 2 is still somehow puzzling. The angular distribution shows a clear $Y_3^{\pm1}(\theta,\phi)$ nature. Under the dipole approximation, we would expect that in the linear polarized laser fields the magnetic quantum number should be conserved during the ionization process. The initial state of the potassium atom is $^2S_{1/2}$ with the magnetic quantum number $m_l = 0$. From this state we would expect that, under the perturbation condition, the ionized electron angular distribution features only spherical harmonic $Y_N^0(\theta,\phi)$, where N reflects the number of absorbed photons leading to the ionization. One possible explanation addressing the observed peak 2 is due to the spin-orbit coupling in the $4p$ state. The spin-orbital coupling can cause the mixing between different Zeeman levels. In this case the magnetic quantum number is not a good quantum number anymore but a superposition of different m_l's which mix the $m_l = \pm1$ levels and the $m_l = 0$ level. This mixing will give rise the observed $Y_3^{\pm}$ feature in the photoelectron angular distribution due to the present of the $m_l = \pm1$ components. The spin-orbit interaction will give the photoelectron with $Y_3^{\pm1}(\theta,\phi)$ angular distribution but not the energy splitting, as for the case of those with $Y_3^0(\theta,\phi)$ distributions. The reason for this can be understood from Eq. (2) in a laser dressed-state picture. The observed photoelectron energy splitting between peaks 1 and 3 shown in Figs. 1 and 3 can be viewed as the energy splitting of the ground state $^2S_{1/2}$ due to the resonance coupling between the $4s$ and $4p$ states. The quantity of the splitting is expressed as the Rabi frequency which in tune is linked to the dipole matrix element. The Rabi frequency has contribution solo from the matrix element with $\Delta m_l = 0$, whereas the matrix elements with $\Delta m_l = \pm1$ are 0. This is why the component with the $Y_3^{\pm1}(\theta,\phi)$ angular distribution does not split and shows only a single peak in the kinetic energy spectrum. Though plausible, the mechanism for the observed peak 2 in Fig. 1 needs further investigation. Currently, a numerical approach for solving the time-dependent Schrödinger equation taking into account the relativistic effect is undertaking, aiming to reveal the nature of the observed $Y_3^{\pm1}(\theta,\phi)$ component.

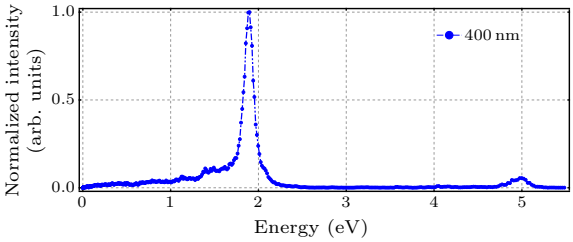

Fig. 4. Kinetic energy spectrum of photoelectrons generated in a 400 nm femtosecond laser field with the intensity of around 1.5×10^{11} W/cm^2.

We also studied the multiphoton ionization of potassium atoms in the 400 nm femtosecond laser field with the intensity of around 1.5×10^{11} W/cm^2. The velocity map imaging measurement results are shown in Figs. 1(c) and 1(d). The peculiar energy level structure of the potassium atom offers a near resonance with a single 400 nm photon between the $4s$ and $5p$ states. Via this resonance, the K atom needs only one more photon to get ionized. Naively, we would expect

similar results compared to the results with the 800 nm laser pulse; that is, the kinetic energy of the photoelectron will split into two components with similar angular distribution featuring the $Y_2^0(\theta,\phi)$ spherical harmonics. The angular distribution fits well with the expected $Y_2^0(\theta,\phi)$ spherical harmonic function as shown in Fig. 2(d) due to the two-photon ionization. However, the kinetic energy distribution shows only one peak (shown in Fig. 4) labeled as peak 4 in Fig. 1(d). The electron peaks at around 1.85 ± 0.05 eV, which is exactly the energy difference between two-400 nm photons and the ionization potential of K. Two possible reasons account for the lacking of the energy splitting in the kinetic energy distribution. The first is that one 400 nm photon couples the $4s$ and $5p$ states near resonantly resulting the Rabi frequency proportional to the matrix element $\langle 5p|r|4s\rangle$, which is one order of magnitude smaller than the matrix element $\langle 4p|r|4s\rangle$. This fact leads to one order of magnitude smaller Rabi frequency compared to the case in the 800 nm laser field. This smaller Rabi frequency will fall into the bandwidth of our pulsed laser, and thus only broadens the kinetic energy distribution without any splitting. A second possibility is again linked with ac-Stark shift. Note that the field free energy difference between the $4s$ and $5p$ states is around 3.06 eV, whereas one single 400 nm photon energy is 3.10 eV. Differing from the case of the 800 nm, the 400 nm laser is blue-detuned relative to the energy levels of $4s$ and $5p$ states. Similar to the case of the $4p$ state, the ac-Stark produces a net negative energy shift that will increasingly take the interaction out of resonance, and thus decrease the effective population in the $5p$ state. In the current status, we are not able to confirm which mechanism is the dominated one or if both lead to the experimental observations. Experiments with a wavelength-tunable femtosecond laser are planed in the near future with hope of fully understanding our experimental results. Meanwhile, a time-resolved experiment aiming at revealing the photoionization dynamics of the dressed states coupled by the resonance laser pulse is being undertaken in our laboratory.

In summary, we have performed experimental investigation of multiphoton ionization of potassium atoms in both 800 nm and 400 nm femtosecond laser fields. In the 800 nm laser field, the photoelectrons clearly show the 1+2 resonance multiphoton ionization feature. The one-photon-dressed energy levels of either $4s$ or $4p$ states are directly mapped into the photoelectron kinetic energy distribution. This energy splitting lifts the energy degeneracy, which allows us to observe an unexpected peak component in the kinetic energy spectrum. A seemingly plausible but still preliminary explanation taking into account the spin-orbit interaction addressing the origin of this peak is given, which explains the measured energy and the angular distribution. We hope that our results will stimulate further theoretical research where the spin-orbit interactions are fully taken into account. Moreover, a comparison study with a 400 nm laser has been carried out. However, near the $4s$ and $5p$ resonance, no energy splitting is found in the kinetic energy distribution. Two possible mechanisms are proposed to explain the observed differences. We propose that the kinetic energy splitting of photoelectron can be used as an alternative way of calibrating the intensity of a pulsed laser. The typical energy resolution of a velocity map imaging spectrometer used is a few tens of eV in our experiment. From the simple relation $\frac{\Delta E_k}{E_k} = \frac{1}{2}\frac{\Delta I}{I}$, for the kinetic energy of an electron typically around 1 eV, the relative accuracy of the laser intensity calibration can achieve 10%. Providing that the transition matrix element is precisely known in some atoms, the absolute value of the laser intensity can be derived in a wide range.

References

[1] Mainfray G and Manus G 1991 *Rep. Prog. Phys.* **54** 1333
[2] Joachain C J 2014 *Europhys. Lett.* **108** 44001
[3] Reid K L 2003 *Annu. Rev. Phys. Chem.* **54** 397
[4] Sharma A, Bhale G L and Razvi M A N 1990 *Pramana-J. Phys.* **35** 95
[5] Wollenhaupt M, Krug M, Köhler J, Bayer T, Sarpe-Tudoran C and Baumert T 2009 *Appl. Phys. B* **95** 647
[6] Hockett P, Wollenhaupt M, Lux C and Baumert T 2014 *Phys. Rev. Lett.* **112** 223001
[7] Hockett P, Lux C, Wollenhaupt M and Baumert T 2015 *Phys. Rev. A* **92** 013412
[8] Wollenhaupt M, Präkelt A, Sarpe-Tudoran C, Liese D and Baumert T 2005 *J. Mod. Opt.* **52** 2187
[9] Wollenhaupt M, Engel V and Baumert T 2005 *Annu. Rev. Phys. Chem.* **56** 25
[10] Wollenhaupt M, Präkelt A, Sarpe-Tudoran C, Liese D and Baumert T 2006 *Appl. Phys. B* **82** 183
[11] Bayer T, Gräfing D, Kerbstadt S, Pengel D, Eickhoff K, Englert L and Wollenhaupt M 2019 *New J. Phys.* **21** 033001
[12] Kerbstadt S, Eickhoff K, Bayer T and Wollenhaupt M 2019 *Adv. Phys.: X* **4** 1672583
[13] Pengel D, Kerbstadt S, Englert L, Bayer T and Wollenhaupt M 2017 *Phys. Rev. A* **96** 043426
[14] Pengel D, Kerbstadt S, Johannmeyer D, Englert L, Bayer T and Wollenhaupt M 2017 *Phys. Rev. Lett.* **118** 053003
[15] Liu Y, Hu W, Luo S, Yuan K J, Sun Z, Bandrauk A D and Ding D 2019 *Phys. Rev. A* **100** 023404
[16] Hu W, Li X, Zhao H, Li W, Lei Y, Kong X, Liu A, Luo S and Ding D 2020 *J. Phys. B: At. Mol. Opt. Phys.* **53** 084002
[17] Song L, Sun Y, Wang Y, Wang X, He L, Luo S, Hu W, Tong Q, Ding D and Liu F 2019 *Chin. Phys. B* **28** 063201
[18] Autler S H and Townes C H 1955 *Phys. Rev.* **100** 703
[19] Wollenhaupt M, Krug M, Köhler J, Bayer T, Sarpe-Tudoran C and Bauert T 2009 *Appl. Phys. B* **95** 245
[20] Nandy D K, Singh Y, Shah B P and Sahoo B K 2012 *Phys. Rev. A* **86** 052517