Tunnel ionization of potassium and xenon atoms in a high-intensity CO_2 laser radiation field

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The experimentally determined ion and electron yields of the tunnel ionization of two very different atoms—potassium and xenon—were compared with different versions of the Ammosov–Delone–Kraïnov theory. Good agreement between the theoretical and experimental results was obtained.

1. INTRODUCTION

Single and multiple ionization of atoms in low-density gases by high-intensity laser pulses has recently been observed experimentally in the spectral range from infrared to ultraviolet.[1-8] These experiments have been carried out under tunnel conditions when the Keldysh parameter γ is less than or much less than unity. We recall that γ is the ratio of the ionization potential of an atom to twice the ponderomotive potential. In particular, multiple ionization of atoms in low-density gases by infrared laser radiation is reported in Refs. 2 and 4. The duration of the CO_2 laser pulses was 1 ns and the wavelength was 10.6 μm.

The same laser was used by Xiong et al.[5] to determine the energy distributions of electrons formed as a result of ionization of xenon and neon.[1] Corkum et al.[6] determined the energy spectra of the electrons emitted as a result of ionization by linearly and circularly polarized CO_2 laser pulses of 2 ps duration. Augst et al.[8] observed multiple tunnel ionization of rare-earth vapor atoms by laser pulses of 1 ps duration and of 1.05 μm wavelength. The results of this experiment clearly agreed best with the model of above-barrier Coulomb dissociation of an atom.

Both of these experiments were concerned either with the formation of ions or with the energy distribution of the emitted electrons, but not with both these aspects. In the present study we tackled the formation of ions and the electron spectra resulting from the ionization of a many-electron system such as the xenon atom, and also the potassium atom, which is effectively a one-electron system. Moreover, we compared the experimental results with different versions of the Ammosov–Delone–Kraïnov (ADK) tunnel theory.[9] This theory was used also in a comparison with the model of above-barrier Coulomb dissociation caused by multiple ionization of xenon.

Experiments of two types were carried out. In one of them we used CO_2 laser pulses ($\lambda = 10.6 \mu$m) of 2 ns duration. An effusion beam of potassium atoms intersected the laser beam and this produced K^+ ions and electrons. In the second experiment we used CO_2 laser pulses of 1.1 ns duration to ionize a gas of xenon atoms at rest. The detection technique was familiar: it was based on a time-of-flight mass spectrometer, which was used to count the ions, and on the retarding potential method, which was used to count high-energy electrons. We had published the experimental data on the ionization of xenon earlier,[1,2,4,5] but the results of potassium are completely new.

2. EXPERIMENTS ON THE IONIZATION OF POTASSIUM ATOMS

Figure 1 shows the dependence of the number of the K^+ ions on the laser radiation intensity, while Fig. 2 gives the energy distribution of the emerging electrons (in the range of energies from 5 to 10 eV) when the laser radiation intensity was 2.7×10^{12} W/cm^2. The continuous curves represent the results based on the ADK theory.[9] We describe the tunneling probability by different versions of the expression obtained from the ADK theory:[9]

$$W = \left(\frac{3e}{\pi}\right)^{3/2} \frac{Z^2}{n^{4,5}} \left(\frac{4eZ^3}{Fn^4}\right)^{2n-1,5} \exp\left(-\frac{2Z^3}{3n^3F}\right). \quad (1)$$

Here, W is the probability of ionization per unit time; e is the base of the natural logarithms; F is the amplitude of the intensity of the laser electric field; Z is the charge of the atomic residue; n is the effective principal quantum number of the ground state. The atomic system of units is used. The continuous curve in Fig. 1 gives the results of a calculation carried out on the basis of Eq. (1) and involving the usual integra-
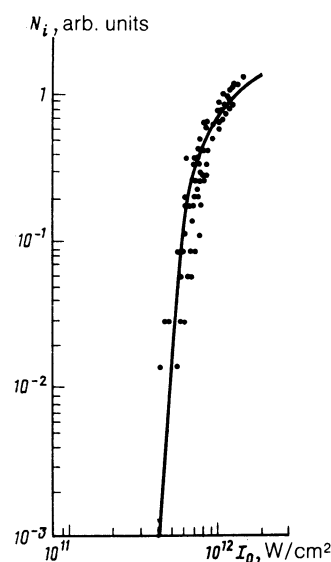

FIG. 1. Dependence of the number N_i of the K^+ ions on the laser radiation intensity I_0 plotted on a double logarithmic scale. The points are the experimental results. The continuous curve is based on a modified version of the ADK theory.[9]

tion of the probability over the spatial and temporal distributions of the radiation intensity.[4] No fitting parameters were used to find the laser radiation intensity scale.

The theoretical energy spectrum of electrons (continuous curve in Fig. 2) was obtained from Eq. (1) on the assumption that all the electrons were accelerated by the ponderomotive potential in the focal plane and the initial kinetic energy was zero. Since the energy of the detected electrons was quite high (up to 10 eV), it was reasonable to assume that electrons escaped from the focal region (with a diameter of the order of 100 μm) in a time much shorter than the laser pulse duration. We thus allowed for the spatial distribution of the ponderomotive potential in the focal plane; it was assumed that the distribution of the laser radiation intensity was Gaussian (because the experimentally determined spatial and temporal distributions of the radiation intensity in the focal plane were roughly Gaussian). We see from Figs. 1 and 2 that the agreement between the theoretical and experimental results is good.

3. EXPERIMENTS ON THE IONIZATION OF XENON ATOMS

We also compared the ADK theory with our earlier experimental results on the ionization of xenon.[1-5] The procedure in our calculations and the assumptions were exactly the same as in the case of the potassium atom. We can see from Fig. 3 that the agreement between the theoretical and experimental data is good for Xe^+ and Xe^{2+}. The cascade (multistage) nature of the ionization is assumed. It is also clear from Fig. 3 that the amplitude of the weakest recorded ion signal is three orders of magnitude less than the value causing saturation of the ionization of atoms (ions): in this case the ion yield curve began to bend smoothly toward the abscissa. We used the ADK theory to determine the density I_A of the signal needed to generate ions with different charges: the calculated value of the ion signal was found to be three orders of magnitude less than the signal corresponding to saturation. The experimental value of I_A was deduced from the minimum detectable signal for an ion with a given
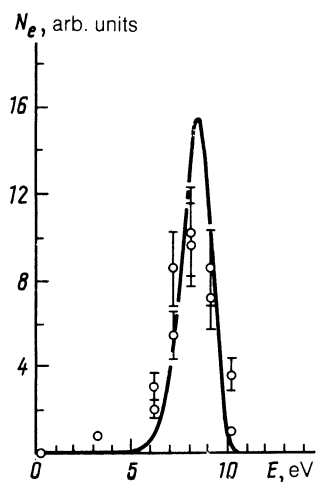

FIG. 3. Comparison of the experimental values of the number of ions N_i formed by the tunnel ionization of Xe producing Xe^+ (O) and Xe^{2+} (●), plotted as a function of the laser radiation intensity I_0, with various theoretical models. Cascade ionization is assumed. The chain curve is based on perturbation theory; curve 1 is the ADK theory; curves 2 and 3 are the ADK theory allowing for the intensity-dependent shift of the ionization potential of the original atom or ion.

charge. In view of the very steep slope of the ion yield curves, the error in the experimental determination of the intensity corresponding to the appearance of an ion was negligible.

We calculated the experimental values of I_A for the xenon ions up to and including Xe^{6+} (Ref. 2). We compared (Fig. 4) the theoretical (open circles) and experimental (black points) appearance intensities for the xenon ions carrying charges from 1 to 6. This figure includes also the corresponding appearance intensities found using the model of above-barrier Coulomb dissociation (triangles).[8] We can see that the tunnel ADK theory[9] agrees much better with the experimental results than the model of above-barrier Coulomb dissociation.

4. SPECTRA OF HIGH-ENERGY ELECTRONS GENERATED BY THE IONIZATION OF Xe

In this section we compare the theoretical predictions with the experimental data on high-energy (200 eV or higher) electron spectra. In calculations of the energy spectra we

FIG. 2. Energy spectrum of electrons formed by tunnel ionization of potassium with radiation of 10^{12} W/cm² intensity. The points are the experimental values and the continuous curve is the ADK theoretical result[9] calculated on the assumption that the emitted electrons have zero kinetic energy and that they are accelerated by the ponderomotive potential.
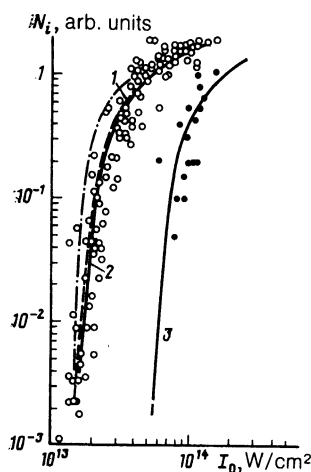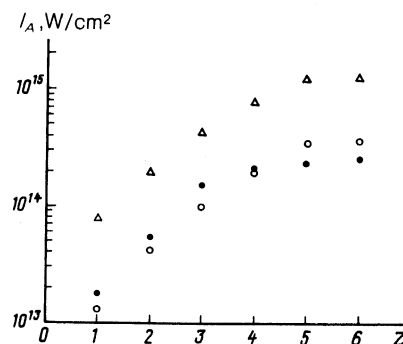

FIG. 4. Comparison of the experimental values of the intensity at which the I_A signal appears (●) with the results calculated on the basis of the ADK theory (O) and on the basis of the model of above-barrier Coulomb dissociation (△) of xenon ions with different charges Z.

used the same method as described above for the ionization of potassium. A comparison of the theoretical and experimental data[5] obtained for the intensity 7.5×10^{13} W/cm^2, when only the Xe$^+$ ion is formed, is made in Fig. 5. The comparison of two wide energy spectra in Fig. 6 applies to the laser radiation intensity increased to 1.5×10^{14} W/cm^2 before the Xe$^+$ and Xe^{2+} ions can form. The dashed curves in Figs. 5 and 6 are theoretical.

Analysis of these figures shows that whereas the theoretical spectra (dashed curves) coincide with the rising parts of the experimental spectra (points), on the high-energy side the experimental spectra are wider than the theoretical results. This means that some electrons with very high energies are formed in our experiments. If we retain the hypothesis that the electrons are accelerated by the ponderomotive potential in the focal plane,[8] then electrons with a higher energy should be created at a higher laser radiation intensity in the course of time evolution of a laser pulse. If the ionization mechanism proposed above is valid, this means that the ionization potential of the original atom or ion clearly increases, so that a higher laser radiation intensity is needed to detach an electron. We therefore assume that the ionization potential E_i depends on the intensity I as follows:

$$E_i = E_i^{(0)} + \sum_{n=1}^{\infty} C_n I^n, \qquad (2)$$

where E_i is the ionization potential of the original atom or ion and $E_i^{(0)}$ is the unperturbed ionization potential. The last term in Eq. (2) is a series in powers of the intensity I.

Retaining only the first six terms of the series and fitting the coefficients C_n, we can ensure agreement between the theoretical and experimental spectra in Fig. 6 by employing the intensity-dependent value of E_i from Eq. (2). The corresponding theoretical spectrum is represented by the continuous curve, which agrees much better with the experimental data. It should be pointed out that an increase in the ionization potential [i.e., the last term in Eq. (2)] for the majority of the Xe atoms and the Xe$^+$ ions is very small: it amounts to 1 and 0.5 eV, respectively. The different values of the coefficients C_n in Eq. (2), obtained by fitting to the first peak in Fig. 6 (representing electrons formed by single ionization), were used to calculate the energy spectrum of electrons generated by laser radiation of 7.5×10^{13} W/cm^2 intensity. The
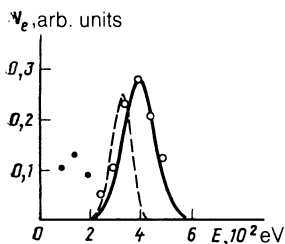

FIG. 6. Energy spectrum of electrons N_c formed by single (1) and double (2) ionization of xenon using laser radiation of 1.5×10^{14} W/cm^2 peak intensity. The dashed curve represents calculations carried out on the assumption that the unperturbed ionization potential is 12.13 eV. The continuous curve allows for the field-induced shift of the ionization potential. The dotted line describes the results obtained in the lowest order of perturbation theory when $W = (I/I_s)^N$ for the saturation intensities $I_s = 1.88 \times 10^{13}$ W/cm^2 in the case of Xe$^+$ (1) and $I_s = 8 \times 10^{13}$ W/cm^2 in the case of Xe^{2+} (2).

results are represented by the continuous curve in Fig. 5. We can see that the agreement between the theory and experiment is again very satisfactory.

To complete the picture of the observed effects, we used the intensity-dependent ionization potentials and new calculations of the intensity dependence of the ion yield. The results are represented by the continuous curve in Fig. 3. The agreement between the theory and experiment is as good as in the case when the unperturbed ionization potential is used. In the case of potassium the results are not affected at all because the shift of the ionization potential is negligible in view of the low radiation intensity.

5. CONCLUSIONS

We conclude by noting that we investigated the tunnel ionization of two very different atoms, potassium and xenon, by measuring the ion and electron yields. The ion and electron results for potassium were in very good agreement with the modified tunnel ADK theory allowing for the spatial and temporal distribution of the intensities. On the other hand, in the case of xenon the ion and electron yields could be described simultaneously in a satisfactory way if the shift of the ionization potential by the laser field was described within the framework of the ADK theory. The physical reason for this shift is the static Stark effect affecting the ground state.

The results can be compared also with other theories. In particular, the original theory of Keldysh[10] does not ensure a good agreement with the results on xenon, so there is no point in trying to carry out a similar comparison for potassium. Moreover, the modified theory of Reiss[11] is unsatisfactory. The chain curve in Fig. 3 represents the results of application of perturbation theory to the case of Xe$^+$. The agreement is satisfactory in the case of the ion yield, but the electron spectra agree very poorly with the predictions of this theory (dotted curves in Fig. 6). This demonstrates that simultaneous fitting of the theory to the data on ions and electrons is a much more reliable procedure than only fitting to the results on ions.

Moreover, we would like to draw attention to the following. While our experiments and the work of Augst et al.[8] correspond to the tunnel ionization case ($\gamma < 1$ or $\gamma \ll 1$), our

FIG. 5. Energy spectrum of electrons N_c formed by single ionization of xenon using laser radiation of $I_0 = 7.5 \times 10^{13}$ W/cm^2 intensity. The dashed curve represents calculations, based on the ADK theory with the unperturbed ionization potential of 12.13 eV, carried out on the assumption that electrons are accelerated by the ponderomotive forces. The continuous curve is the result of a similar calculation, but after allowing for the shift of the ionization potential (see text). The black dots represent the contribution of impurities.
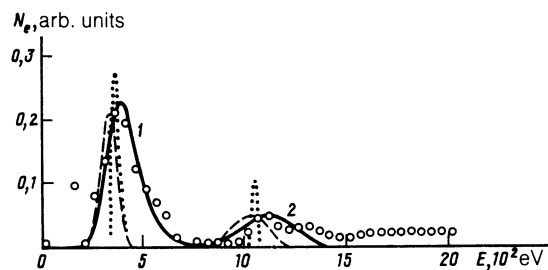
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results are closer to the tunneling model, whereas the experiments of Augst et al.[8] are closer to the model of above-barrier Coulomb dissociation. The difference between the two sets of results is mainly in the laser pulse duration, since the difference in respect of the wavelength or frequency is allowed for in the definition of the tunnel regime based on the Keldysh parameter γ. Hence, it follows that in the process of slow rise of a long CO_2 laser pulse (when the pulse duration is 1 ns) the electric field of the laser radiation passes through many oscillation cycles before the total tunneling probability becomes of the order of unity. However, if the laser pulse is much shorter (1 ps) and the wavelength is 1 μm, then during the fast rise time of the laser pulse the number of such oscillations may be insufficient to ensure a sufficiently high tunneling probability. Therefore, tunneling does not occur and a further increase in the laser radiation intensity suppresses the Coulomb barrier, so that an electron is simply "pushed out" from an atom.

A more detailed investigation of the tunnel ionization probability based on the ADK theory shows[2] that in the case of the laser wavelengths under consideration here the intensity I_A needed for the formation of a particular charge state (for example, Xe^+) is less than that calculated using the model of above-barrier Coulomb dissociation.[8] This can be seen qualitatively from the suppression of the Coulomb barrier. The tunneling occurs before the Coulomb barrier drops to the energy level of the ground state. This means that the tunneling should predominate over the process of above-barrier Coulomb dissociation. It should also be noted that during a pulse of 1 ps duration there are still many optical oscillations (when the wavelength is 1 μm), so that the tunneling should still be significant.

This conflict clearly demonstrates that the length of the laser pulse is an important factor in the study of the process of ionization of atoms with ultrastrong laser pulses. Another experimental confirmation of the influence of the pulse length is an increase in the intensity I_A when atoms are ionized by picosecond pulses.[3,8]

It is therefore necessary to develop new theories in the physics of strong fields characterized by ultrashort durations of interaction with atoms.

Note added in proof (December 17, 1990). In our calculations the width of the electron energy distribution was entirely due to the change in the laser intensity in space and time, since we assumed that at the time of escape from the tunneling region the initial kinetic energy is zero. Delone and Kraĭnov recently calculated [J. Opt. Soc. Am. B (January 1991)] the energy spectrum of electrons in the course of tunnel ionization before the ponderomotive acceleration. Therefore, in a comparison with the experimental data it was necessary to apply a "convolution" of the two distributions. The distribution of Delone and Kraĭnov is in better agreement with the experimental data for potassium than the present distribution.

[1] There is a small error in Ref. 5 in the calibration of the intensity, but it is corrected in the present paper.
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