Laser detection of ^{40}K by isotopically selective ionization in a beam of accelerated atoms

Yu. A. Kudryavtsev, V. S. Letokhov, and V. V. Petrunin
Institute of Spectroscopy, Academy of Sciences of the USSR

(Submitted 12 May 1985)
Pis'ma Zh. Eksp. Teor. Fiz. **42**, No. 1, 23–26 (10 July 1985)

The isotope ^{40}K has been detected by a two-step laser photoionization in an accelerated atomic beam with collinear excitation. A selective ionization of 10^5 has been achieved.

1. The development of laser methods for detecting single atoms[1] has made it possible to take up the problem of directly determining ultralow concentrations of isotopes without making use of radioactive decay. This problem is currently being solved by using megaelectronvolt-range accelerators as highly sensitive mass spectrometers.[2] Laser methods are potentially much simpler and cheaper.[3] However, the selectivity of the laser excitation of rare long-lived isotopes in the presence of stable abundant isotopes is limited by the small isotopic shift for the isotopes of most interest. It has been suggested that this difficulty could be overcome by using a method of multistep excitation and ionization of atoms accelerated beforehand in a given potential U (Ref. 4). In this acceleration, the isotopes with different masses acquire different velocities, so that a kinematic isotopic shift arises for any spectral line of any isotope:

$$\frac{\Delta\nu_{12}}{\nu_0} = \frac{1}{c}\sqrt{2eU}\left(\frac{1}{\sqrt{M_1}} - \frac{1}{\sqrt{M_2}}\right), \qquad (1)$$

 0021-3640/85/130026-05$01.00

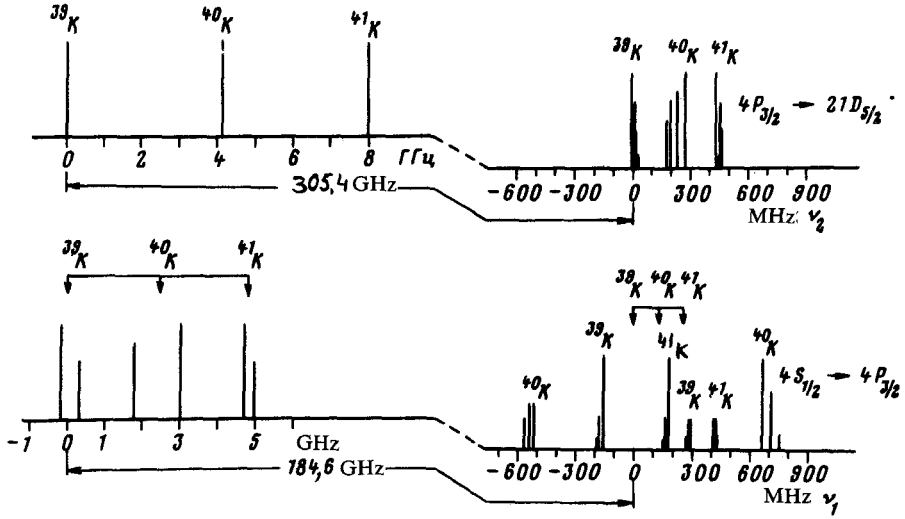

FIG. 1. Calculated absorption spectrum of thermal-energy potassium isotopes (at the right) and of the same isotopes after acceleration to 4 keV (at the left) for the transitions $4S_{1/2} \rightarrow 4P_{3/2}$ and $4P_{3/2} \rightarrow 21D$. The hyperfine-splitting constants are taken from Ref. 7. The arrows mark the centers of gravity of the transitions.

where M_1 and M_2 are the masses of the two isotopes. At $U = 10$ kV, this induced shift is about an order of magnitude larger than the ordinary mass-related isotopic shift. Furthermore, during electrostatic acceleration of atoms (in the form of ions, which subsequently undergo charge exchange and form neutral atoms) there is a bunching of longitudinal velocities, which leads to a pronounced contraction of the Doppler-broadened spectral lines in the case of collinear excitation. As a result, all of the accelerated atoms of the rare isotope interact with monochromatic laser light.

In the present letter we report the first successful experiments on the detection by this method of the relatively rare isotope ^{40}K, which has a very small mass-related isotopic shift. A selective ionization of 10^5 has been achieved by virtue of isotopically selective excitation in only one step of a two-step excitation into a Rydberg state.

2. The potassium atom has two stable isotopes, ^{39}K and ^{41}K, with abundances of 93.2% and 6.8%, respectively, and the radioactive isotope ^{40}K ($T_{1/2} = 1.3 \times 10^9$ yr) with an abundance of 0.012%.

Figure 1 shows the calculated absorption spectra of the thermal-energy isotopes of potassium (at the right) and the same isotopes after acceleration to 4 keV (at the left) in the first and second steps of the excitation. Since the excitation by the laser beam occurs in the direction opposite that in which the atoms are moving, the absorption spectra of the accelerated atoms are displaced in the red direction with respect to the spectra of thermal atoms. The spectra shown here for the thermal atoms reflect the hyperfine splitting of the $4S_{1/2}$ ground state and of the $4P_{3/2}$ first excited state, while the spectra of the accelerated atoms (in the scale used here) reflect the hyperfine splitting of only the ground state. When the atoms are accelerated to 4 keV, the

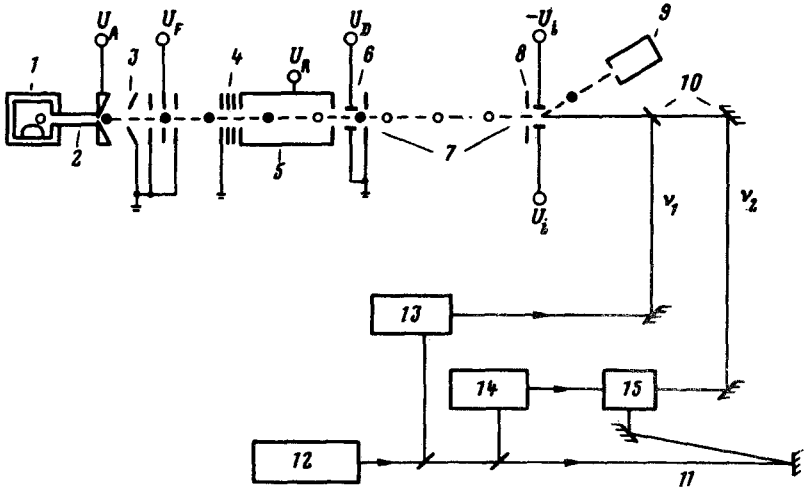

FIG. 2. The experimental apparatus. 1—Atomizer; 2—ionizer; 3—extraction electrode; 4—ion retardation system; 5—charge-exchange cell; 6—filtering capacitor; 7—field-free region; 8—field ionizer; 9—secondary electron multiplier; 10—mirrors; 11—optical delay line; 12—XeCl excimer laser; 13—laser of first step; 14, 15—laser oscillator and laser amplifier of the second step.

isotopic shift in the second step increases by a factor of 20, from 220 MHz (Ref. 6) to 4.1 GHz (Fig. 1), and the Doppler width of the absorption line decreases by a factor of $2\sqrt{eU/kT}$, to $\Delta\nu_D = 5.9$ MHz at a source temperature $T = 1073$ K. In the experiments, the atoms of all three isotopes are sent from the $4S_{1/2}$ ground state into the $4P_{3/2}$ first excited state by the beam from the wide-band laser in the first step ($\lambda_1 = 765$ nm, $\Delta\nu_1 = 0.8$ cm^{-1}). The atoms of the one selected isotope are then excited by the beam from the narrow-band laser in the second step ($\lambda_2 = 460$ nm, $\Delta\nu_2 = 0.015$ cm^{-1}) into the $21D$ Rydberg state, from which they are ionized in the static electric field.

3. The beam of accelerated potassium atoms is produced by resonant charge exchange of accelerated potassium ions; the experimental arrangement is shown in Fig. 2. After metallic potassium is atomized in atomizer 1, the potassium is ionized on the 800 °C surface of the tantalum foil in ionizer 2. The resulting ions are accelerated in the electric field U_A of accelerator 3, and a single lens shapes these ions into a parallel beam. The ions then enter the charge-exchange cell, 10 cm long and filled with potassium vapor, in which a temperature of 160 °C is maintained. Here 80% of the ions are converted into atoms; those ions that do not undergo charge exchange are stripped off in filtering capacitor 6. The effective current of the atoms in the beam is 3 nA at a beam diameter of 3 mm. The atoms then pass through field-free region 7, in which the fast atoms are excited into the Rydberg state by the isotopically selective two-step process. The Rydberg atoms are ionized in field ionizer 8 and sent to a secondary electron multiplier 9. The output signal from this multiplier is processed by a strobed voltmeter and sent to a chart recorder. It is more convenient to vary the energy of the atoms rather than the laser frequency to achieve exact resonance with the absorption lines of the various isotopes. For this purpose, a retarding system 4 is

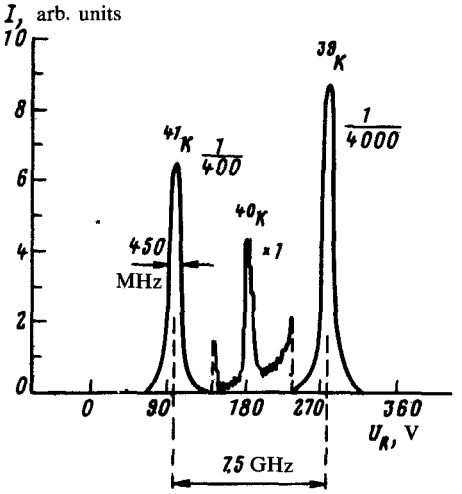

FIG. 3. Yield of photoions from the ionization of potassium atoms versus the retarding voltage.

placed in front of the charge-exchange cell, and a positive retarding voltage $U_R = 0$–320 V is applied to it. At a potassium-atom energy of 3.8 keV a 1-eV change in the energy corresponds to a 40-MHz change in the resonant frequency in the second step. When the retarding voltage is scanned, the various isotopes of potassium reach resonance in succession.

The laser beams at ν_1 and ν_2 are directed by mirrors 10 opposite the atomic beam. The width of the output spectrum of the laser in the first step, 13 is five times the spectral width required for the excitation of all of the potassium isotopes. The laser in the second step consists of a laser oscillator 14 and a laser amplifier 15. The amplifier is pumped with a delay of 12 ns with respect to the time at which the light arrives at the laser amplifier from the laser oscillator. Both of the lasers are pumped with the beam from a XeCl excimer laser 12 operating at 20 Hz.

4. Figure 3 shows the ionization spectrum of the potassium isotopes accelerated to 3.8 keV. The higher retarding voltage corresponds to the peak of the lighter isotope ^{39}K. The intensity ratios of the ^{39}K, ^{40}K, and ^{41}K peaks are equal to the ratios of the natural abundances of these isotopes. The width of the lines observed in the ionization spectrum is 450 MHz, determined by the effective width of the laser line in the second step. The spacing between the ^{39}K and ^{41}K lines, which is 7.5 GHz (185 V), agrees with the calculated value (Fig. 1). The selectivity of the ionization, defined as the ratio of the ^{39}K signal to the background signal, is 10^5.

In summary, laser photoionization of accelerated atoms with collinear excitation makes it possible to achieve a high isotopic selectivity of ionization in a single excitation step. An additional isotopic shift for any spectral line makes it possible to implement the idea of multiplying selectivities through multiple-step excitation.[8] Collinear photoionization could also be used at the exit from a mass separator to radically improve the resolution.

[1]V. I. Balykin, G. I. Bekov, V. S. Letokhov, and V. I. Mishin, Usp. Fiz. Nauk **132**, 293 (1980) [Sov. Phys. Usp. **23**, 651 (1980)].
[2]A. E. Litherland, R. P. Beukens, L. R. Kilius, J. C. Rucklidge, H. E. Gove, D. Elmore, and K. H. Purser, Nucl. Instrum. Methods **186**, 463 (1981).
[3]V. S. Letokhov, Comments At. Mol. Phys. **10**, 257 (1981).
[4]Yu. A. Kudriavtsev and V. S. Letokhov, Appl. Phys. **B29**, 219 (1982).
[5]K.-R. Anton, S. L. Kaufman, W. Klempt, G. Muzzi, R. Neugart, E. W. Otten, and B. Schinzler, Phys. Rev. Lett. **40**, 642 (1978).
[6]L. R. Pendrill and K. Niemax, J. Phys. B **15**, L147 (1982).
[7]E. Arimondo, M. Inguscio, and P. Violino, Rev. Mod. Phys. **49**, 31 (1977).
[8]V. S. Letokhov and V. I. Mishin, Opt. Commun. **29**, 168 (1979).

Translated by Dave Parsons