Direct Observation of Hot-Electron Spectra from Laser-Excited Sodium Vapor

J. L. Le Gouët and J. L. Picqué
Laboratoire Aimé Cotton, Centre National de la Recherche Scientifique, F-91405 Orsay, France

and

F. Wuilleumier, J. M. Bizau, and P. Dhez
Laboratoire de Spectroscopie Atomique et Ionique and Laboratoire pour l'Utilisation du Rayonnement Electromagnétique, Université Paris-Sud, F-91405 Orsay, France

and

P. Koch
Yale University, New Haven, Connecticut 06520

and

D. L. Ederer
Radiation Physics Division, National Bureau of Standards, Washington, D. C. 20234

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A sodium atomic beam with density 10^{13} cm^{-3} was illuminated by cw dye laser radiation (a few watts per square centimeter) tuned to the D_2 resonance line. In the energy spectrum of the emitted electrons, several lines were observed between 4 and 7 eV. Their positions and intensities indicate that seed electrons are produced either via associative ionization or via collisional ionization from excited states populated by energy-pooling collisions. These electrons are then heated through successive superelastic collisions with excited $3p$ atoms.

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There is a crucial interest in fields as different as plasma physics, astrophysics, or photochemistry to obtain a better knowledge of collisional and ionization processes in a medium containing ground-state atoms, excited atoms, and ions. Within the past few years, following the pioneering work of Lucatorto and McIlrath,[1] a number of experiments have studied collisional mechanisms which produce ionization in a sodium vapor illuminated by a laser tuned to the $3s$-$3p$ resonance line.[2-6] Three classes of processes have been put forward to account for laser ionization.[7,8a] The first is multiphoton ionization, which requires high laser intensity to observe significant effects. The second is collisional ionization involving atom-atom processes, such as associative ionization, energy pooling, and laser-induced collisions. The third group involves electron-atom processes, such as superelastic collisions and electron impact ionization. Up to now, accurate experimental data were very scarce, and a new type of information was obviously needed to obtain a clear identification and understanding of the dominant processes.

In this Letter, we provide (i) a direct observation of superelastic collisions[8b] in the laser-excited medium, and (ii) the unambiguous identification of the mechanisms for the production of seed electrons. While the previous experiments were investigations of either the total ion yields[3-5] or the mass-resolved ion spectra,[2,6] the present experiment introduces electron spectrometry to observe, for the first time, the energy spectrum of electrons emitted from such a medium. In our experimental conditions, it was possible to observe hot electrons of up to 6.3 eV kinetic energy in a medium (density $n_a \sim 10^{13}$ cm^{-3}) interacting with photons of energy (2.11 eV) far below the atom ionization threshold (5.14 eV). We provide strong evidence that the low-energy primary electrons are created by purely collisional processes involving excited atoms (the intensity of our cw dye laser was low enough, about 3 W/cm^2, so that the multiphoton or laser-assisted processes are completely negligible). The energy of these seed electrons is then increased through superelastic collisions with excited $3p$ atoms.

From the analysis of the observed electron energy spectra, it follows that seed electrons are produced through two main mechanisms under our experimental conditions. The first one is associative ionization:

$$\mathrm{Na}(3p) + \mathrm{Na}(3p) \rightarrow \mathrm{Na}_2{}^{+} + e(E < 0.1\ \mathrm{eV}). \qquad (1)$$

This process, already considered as the seeding mechanism in the previous ionization studies on sodium,[2-6] has a cross section $\sigma_{\mathrm{AI}} \simeq 5\times10^{-18}$ cm^2.[3] The second mechanism is ionizing collisions occurring between $3p$ atoms and atoms in higher excited states nl:

$$\mathrm{Na}(3p) + \mathrm{Na}(nl) \rightarrow \mathrm{Na}(3s) + \mathrm{Na}^+ + e(2.11\ \mathrm{eV} - E_{nl}), \qquad (2)$$

where E_{nl} is the binding energy of the nl electron, and $E_{3p} - E_{3s} = 2.11$ eV. Process (2) was not mentioned as a possible electron seeding mechanism in the previous sodium ionization studies.[9] Actually, it is just being studied in rubidium, where very large cross sections σ_{IC} between 10^{-13} and 10^{-12} cm^2 have been measured for collisions between $5p$ atoms and nl atoms ($nl \equiv 9s, 8s, 7d, 6d$).[10] In our experiment, the population of the high-lying nl states involved in process (2) is due to energy-pooling collisions, according to

$$\mathrm{Na}(3p) + \mathrm{Na}(3p) \rightarrow \mathrm{Na}(3s) + \mathrm{Na}(nl) + \Delta W. \qquad (3)$$

This process was observed in studies of the fluorescence of sodium vapor excited by a cw dye laser[2,11]; the cross section σ_{EP} is probably of the order of 10^{-15}–10^{-14} cm^2.[12] Our results confirm that levels $nl \equiv 3d, 4d, 5s$ are the most populated.[11] Finally, the energy of the primary electrons produced by mechanisms (1) and (2) is raised to the observed values through two superelastic collisions of the type

$$e(E) + \mathrm{Na}(3p) \rightarrow \mathrm{Na}(3s) + e(E + 2.11\ \mathrm{eV}). \qquad (4)$$

The cross section of this process, $\sigma_{\mathrm{SE}}(E)$, can be obtained from experimental data on electron-impact excitation,[13] by using the detailed balancing principle.[8] Its values are in the range 10^{-15}–10^{-14} cm^2 for kinetic energies E lower than 4 eV.

In our experiment (Fig. 1), a laser beam irradiates a weakly collimated effusive sodium beam at right angles to its mean direction. The electrons emerging from the interaction zone are detected by a cylindrical mirror analyzer (CMA).[14] The laser beam is adjusted to fit the shape of the source volume ($V \simeq 0.05$ cm^3) seen by the CMA, located close to the circular exit (5 mm diam) of the sodium oven. The oven temperature, 580 K, corresponds to an average relative velocity of the atoms in the beam of $v_a \simeq 4\times10^4$ cm/s. We measured[15] the atomic density in the active volume to be $n_a \simeq 1.5\times10^{13}$ cm^{-3}.

The laser is a cw single-mode ring dye laser (Spectra Physics 380A), which we locked to the $3^2S_{1/2}\,F=2 \longrightarrow 3^2P_{3/2}\,F=3$ hyperfine component of the D_2 resonance line (at 589.0 nm). During the experiments, it delivered typically 400-mW power within a 10-MHz bandwidth. We measured[15] that about 15% of the atoms present in the interaction volume were in the excited state in a steady-state regime, i.e., $n_a^{3p} \simeq 2\times10^{12}$ cm^{-3}.

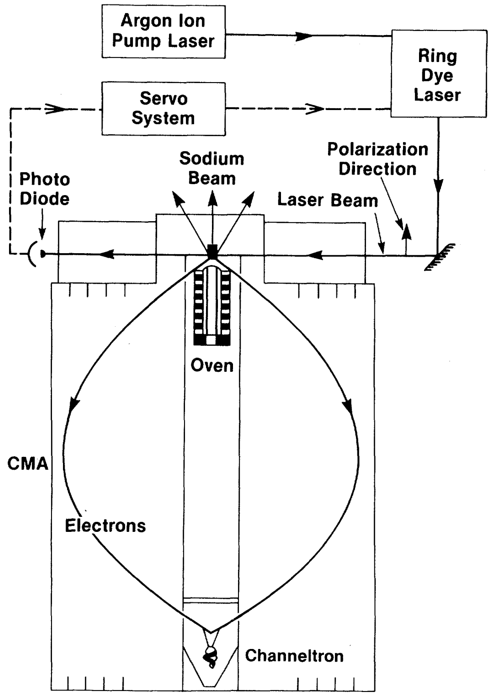

FIG. 1. Experimental setup.

A typical electron energy spectrum is shown in Fig. 2. The absolute energy scale could be accurately established.[15] Under our experimental conditions, low-energy electrons are not transmitted by the CMA. Above 4 eV, the CMA efficiency is proportional to the electron energy.

A number of peaks are observed in the spectrum above 4 eV kinetic energy. The two main ones are measured at 4.25(5) eV (labeled 1 in Fig. 2) and 5.45(5) eV (labeled 2 in Fig. 2). We attribute the origin of peak 1 to very-low-energy electrons produced by associative ionization [process (1)], then heated by two superelastic collisions [process (4)]. Peak 2 should arise from electrons produced by ionization of $4d$ atoms through collisions with $3p$ atoms [process (2)]; these electrons, with energy $2.11\ \mathrm{eV} - E_{4d} = 1.24$ eV, are then heated by two superelastic collisions [process (4)]. The positions of the small peaks at about 4.8 and 5.25 eV correspond well to the energies of electrons issuing from atoms in $3d$ and $5s$ states, respectively, through the same processes (2) and (4). These levels $4d$,

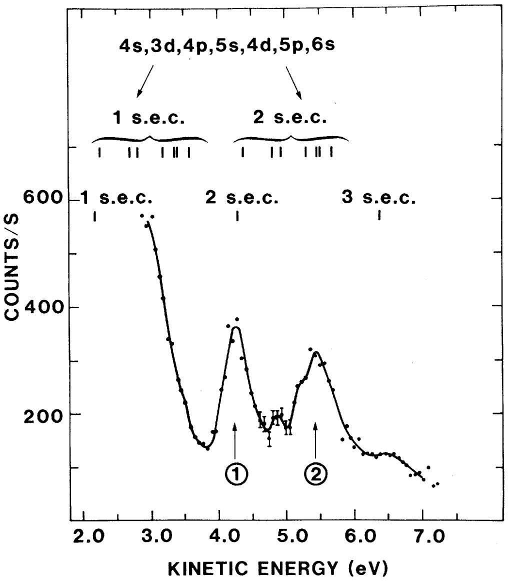

FIG. 2. Typical energy spectrum of electrons ejected from the laser-irradiated sodium vapor. The upper group of bars indicates the energy positions of the electrons created by collisional ionization from the various excited states and heated by either one or two superelastic collisions (s.e.c.); the lower group of bars shows the energy positions of the electrons created by associative ionization and having undergone one, two, or three superelastic collisions.

3*d*, and 5*s* are populated by energy-pooling collisions [process (3)]. The small bump around 6.35 eV is consistent with 4.25-eV electrons having undergone a third superelastic collision. The counting rate rises sharply below 3.5 eV, even though the CMA efficiency drops rapidly to zero in this energy range. This suggests the existence of a huge number of low-energy electrons in the laser-irradiated medium.

The above interpretation primarily relies upon the energy position of the electron lines. But, in the following, we give it strong support by estimating the expected counting rates, using the previously quoted values of the effecitve cross sections and experimental parameters. The number of seed electrons produced per second by associative ionization is $N_e(E_0 < 0.1\text{ eV}) = (n_a^{3p})^2\sigma_{\text{AI}} v_a V \simeq 4\times10^{10}$ electrons/s. The number of seed electrons produced via collisional ionization from the energy-pooling-populated 4*d* states (natural lifetime $\tau^{4d} \simeq 40$ ns) is

$$N_e(E_0' = 1.24\text{ eV}) = [(n_a^{3p})^2\sigma_{\text{EP}} v_a \tau^{4d}](n_a^{3p}\sigma_{\text{IC}} v_a V)$$
$$\simeq 2.5\times10^{10}\text{ electrons/s},$$

using 10^{-14} and 10^{-13} cm^2 for σ_{EP} and σ_{IC}, respectively. The mean free path associated with the various processes which may consume electrons of a given energy (superelastic or inelastic collisions, electron impact excitation or ionization, etc.) is more than 100 times larger than the dimension of the active volume $l \simeq 0.2$ cm. Within a good approximation, the number density of electrons of energy E and velocity $v_e(E)$ is thus simply given by $n_e(E) = [l/v_e(E)][N_e(E)/V]$. Applying this result to the electrons of energy E_{n-1} and to the electrons of energy $E_n = E_{n-1} + 2.11$ eV resulting from an *n*th superelastic collision, we obtain easily the rate of emission of electrons of energy E_n as $N_e(E_n) = N_e(E_{n-1})n_a^{3p} l\sigma_{\text{SE}}(E_{n-1})$. Accounting for the measured transmission of the CMA, this yields calculated values for the areas under peaks 1 and 2 of about 100 counts eV/s ($E_2 = 4.25$ eV) and 60 counts eV/s ($E_2' = 5.45$ eV), respectively, in good agreement with the observed signals (see Fig. 2). This confirms the interpretation we have proposed and the order of magnitude of the cross sections for energy pooling and collisional ionization. Preliminary results concerning the effect of a variation in the excited-state density also support our interpretation: We find that a higher density of 3*p* atoms favors the intensity of peaks 1 and 2 compared with the tail of lower-energy electrons.

In conclusion, we have demonstrated that electron spectrometry brings further insight into the processes occurring in a laser-irradiated medium. In the present case of resonantly excited sodium, our analysis has revealed that, under our experimental conditions, electrons are produced through a small number of dominant processes of purely collisional nature.

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[15] The experimental setup shown in Fig. 1 is part of a setup developed for the study of photoionization out of laser-excited atoms using synchrotron radiation from the ACO storage ring. [A preliminary report was presented as a postdeadline (unpublished) poster session at the Twelfth International Conference on Physics of Electronic and Atomic Collisions.]

Photoelectron Polarization in Hg $6s^2$ Subshell Ionization with Unpolarized Light: New Aspect of the Fano Effect

G. Schönhense and U. Heinzmann

Fritz-Haber-Institut der Max-Planck-Gesellschaft, D-1000 Berlin 33, West Germany, and Physikalisches Institut der Universität Münster, D-4400 Münster, West Germany

and

J. Kessler

Physikalisches Institut der Universität Münster, D-4400 Münster, West Germany

and

N. A. Cherepkov

A. F. Ioffe Physical-Technical Institute, 194021 Leningrad, U. S. S. R.

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Experimental evidence is presented for the effect of photoionization of ns^2 atomic subshells with *unpolarized* radiation that leads to *highly polarized* photoelectrons in angular-resolved measurements. Like the Fano effect, the observed phenomenon is caused by spin-orbit interaction in the continuous spectrum. The spin polarization essentially depends on the phase-shift difference between the $\epsilon p_{1/2}$ and $\epsilon p_{3/2}$ continua.

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In a well-known paper Fano[1] predicted high spin polarization of photoelectrons which are ejected by circularly polarized light from alkali atoms near the Cooper minimum of the cross section. One year later the Fano effect was established experimentally.[2] The polarization of photoelectrons from *ns* subshells is direct evidence of spin-orbit interaction in the continuous spectrum, which leads to a difference between wave functions corresponding to the $\epsilon p_{1/2}$ and $\epsilon p_{3/2}$ outgoing partial waves. For the same reason the angular asymmetry parameter β in the Cooper minimum varies from +2 to −1 as a function of photon energy while in the *LS*-coupling scheme it should be equal to +2 irrespective of photon energy.[3]

The Fano effect also appears in ns^2 subshells,[4-6] since its cause, the spin-orbit interaction in the continuous spectrum, evidently exists there, and there is usually a Cooper minimum, also. But while in alkalis the minimum appears at energies between the first and the second ionization threshold, in ns^2 subshells it can occur at energies above the second threshold. Therefore inter-