Direct Observation of the Momentum-Density Profile of Excited and Oriented Sodium Atoms

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The noncoplanar symmetric $(e,2e)$ cross section is obtained for sodium atoms pumped by σ^+ laser light tuned to the $3^2S_{1/2}(F=2)\leftrightarrow 3^2P_{3/2}(F'=3)$ transition. The excited pumped atoms are in the $m_{F'}=3$ state, or in terms of orbital momentum in the $m_l=+1$ state. The coordinate frame is chosen so that the momentum density of the excited state is probed in the k_x direction with k_z and k_y being fixed and essentially zero. The results are in excellent agreement with the momentum-density profile given by the $3p(m_l=1)$ Hartree-Fock wave function.

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Electron momentum spectroscopy (EMS), or $(e,2e)$ spectroscopy, is now a well developed spectroscopic tool for investigating the electronic structure of atoms and molecules in their ground states.[1] Such $(e,2e)$ experiments have contributed significantly to a better knowledge of the dynamical properties of electron motion and of the role of correlation effects in the valence electronic structure. This is achieved through the determination of electron separation energy spectra and momentum distributions for transitions at different separation energies. All previous EMS measurements on atoms and molecules have been restricted to targets in the ground state and to obtaining spherically averaged momentum distributions due to the fact that the targets have random alignment and orientation with respect to the incident beam.

The advent of high-power tunable dye lasers has made it possible to prepare the initial state in a specific way. The interaction of electrons with such states is of fundamental interest in atomic physics.[2] In EMS the preparation of a target state in a specific magnetic substate allows the possibility of measuring the momentum distributions of electrons in aligned and oriented states. With laser excitation it is also possible to measure the momentum distribution of states not occupied in the ground-state configuration. In addition, the excitation of an outer electron can significantly modify the effective potential experienced by the other electrons, with a resultant modification of their wave functions and momentum distributions.

The investigation of the dynamic mechanism in the presence of a laser field is also of extreme interest. The dressing of the atomic target by the laser field is predicted to strongly affect the triple-differential $(e,2e)$ cross section for high-laser intensities.[3,4] Maquet *et al.*[5] also find cases where this should happen at low-laser intensities. In the present experiment the laser intensities were quite low and we would not expect to see these "dressing" effects, except in as much that the atom is promoted to an excited aligned and oriented state. We report here the first $(e,2e)$ measurements on an excited state which is also oriented, and obtain the probability distribution for an electron having momentum component k_x for this state, namely, the $m_l=+1$ substate of the $\mathrm{Na}(3^2P_{3/2})$ state.

We give here only a brief description of the apparatus and experimental techniques, a detailed description can be found elsewhere.[6,7] An argon-ion laser pumps a cw ring dye laser, which is used to optically pump sodium atoms from the $3^2S_{1/2}(F=2)$ ground state to the $3^2P_{3/2}(F'=3)$ excited state using right-hand circularly polarized light. Noncoplanar symmetric $(e,2e)$ measurements are then made on the sodium beam consisting of both ground-state and excited-state atoms. The reaction can be schematically written as

$$h\nu+\mathrm{Na}(3^2S_{1/2},F=2)$$
$$e^-(E_0,\mathbf{k}_0)+\mathrm{Na}(3^2P_{3/2},F'=3)\rightarrow \mathrm{Na}^+ + e^-(E_1,\mathbf{k}_1) + e^-(E_2,\mathbf{k}_2)\,,$$

where $F=I+J$ is the maximum hyperfine-structure quantum number with $I=\frac{3}{2}$ the nuclear spin of $^{23}\mathrm{Na}$ and J the total electronic angular momentum. The dynamics of atomic collisions are generally independent of the nuclear spin, which enters only through statistics, and if no electron-spin analysis is made, one can generally factor it out as well.[8] Therefore as no spin analysis is involved in the present analysis it is appropriate to use directly expectation values constructed from angular momentum operators $\mathbf{L}$ for the orbital motion of the atomic electron.

The optical pumping of the sodium $^3P_{3/2}$ state has been studied in detail both theoretically and experimentally.[9-11] In the present work σ^+ light from a cw single-mode ring dye laser with very narrow linewidth ($\sim$3 MHz) is tuned to the $3^2S_{1/2}(F=2)\rightarrow 3^2P_{3/2}(F'=3)$ transition. In the stationary condition this pumping with σ^+ light produces a population of only the $F=2$, $m_F=2$ ground state and of the $F'=3, m_{F'}=3$ excited state. This means that in the excited state there is 0% of

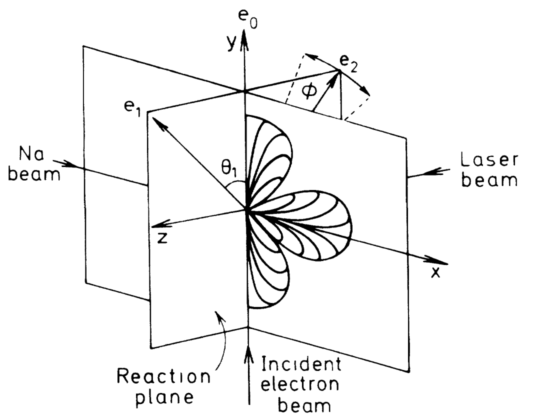

FIG. 1. Schematic diagram of the experimental geometry. The $3p_x$ and $3p_y$ charge-cloud distributions are indicated. The polar angles are $\theta_1=\theta_2=45°$. The out-of-plane azimuthal angle ϕ of detector 2 is varied to vary the momentum k_x.

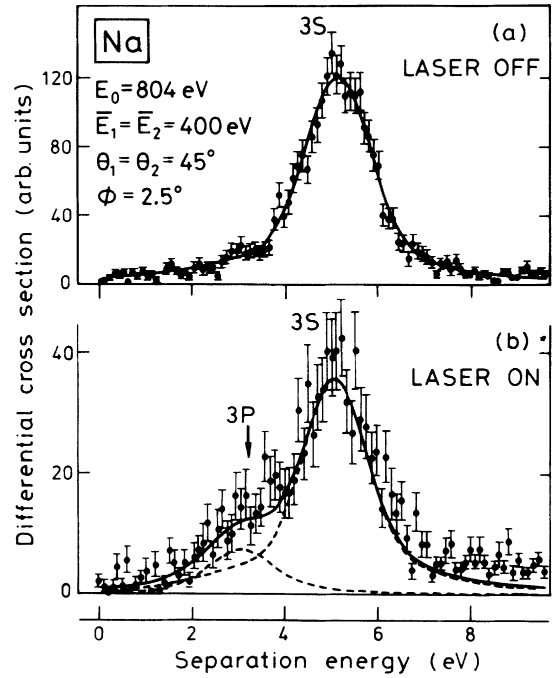

FIG. 2. The coincidence separation or binding-energy spectra of the $3s$ ground state of Na (laser off), and both the $3s$ ground state and $3p$ excited state (laser on) at $\phi=2.5°$. The mean outgoing energies are 400 eV and the incident energy is 804 eV.

the population in the $m_l=0$ (i.e., $3p_z$) orbital and 100% in the $m_l=+1$ orbital, corresponding to 50% in both the $3p_x$ and $3p_y$. The axis of quantization z has been chosen as being the direction of propagation of the circularly polarized pumping beam; the atomic sodium beam is incident in the x direction and the electron beam is incident in the y direction. This is shown schematically in Fig. 1, which also shows the $3p_x$ and $3p_y$ distribution of the charge cloud of the sodium in the $3p(m=+1)$ excited state. The scattering plane is the z-y plane and both outgoing electrons make a polar angle of 45° with respect to the incident (y) direction; the out-of-plane azimuthal angle ϕ of electron 2 is varied to probe the different momentum components of the target.[1] Thus the experiment measures the (e,2e) triple-differential cross section as a function of the x component of momentum, k_x. For symmetric noncoplanar kinematics,[1] which is employed in the present measurements, the z and y components of momenta are fixed and essentially zero for $\theta_1=\theta_2=\theta=45°$ and small binding energies. Because of the finite angular and hence momentum resolution of the spectrometers, the average values of $|k_y|$ and $|k_z|$ are not quite zero, being of the order of 0.06 a.u. The experiment probes the $3p_x$-orbital momentum density along the k_x axis in momentum space.

The experiment was carried out using a new coincidence spectrometer based on two hemispherical electron analyzers with position-sensitive detectors sensitive in the energy dispersing dimension.[6] It is similar in principle to the noncoplanar symmetric spectrometer described by McCarthy and Weigold.[1] The incident electron energy was 804 eV, the outgoing energies of both detected electrons being in the range 400 ± 4 eV. The experimental energy resolution function and the angular resolution of the spectrometer were established by careful measurements of the argon $3p$ separation or binding-energy spectrum and its angular correlation. The resultant angular resolution is $\Delta\theta=1°$ and $\Delta\phi=0.5°$. The binding energies for the $3^2S_{1/2}$ ground state and the $3^2P_{3/2}$ excited state are 5.1 and 3.0 eV, respectively. With the laser off only the ground-state transition could be observed, and with the laser on there is also ionization from the $3p$ excited state (Fig. 2). The ratio of the populations of the excited- and ground-state atoms can be estimated from the relation

$$N_{3p}/N_{3s}=I_{3p}\sigma_{3s}/I_{3s}\sigma_{3p}\ ,$$

where I_{3p} and I_{3s} are the measured intensities of the $3p$ and $3s$ transitions, respectively, at a given ϕ, and σ_{3s}/σ_{3p} is the ratio of the corresponding differential cross sections. Using calculated relative plane-wave impulse approximation (PWIA) cross sections, the resulting value of N_{3p}/N_{3s} is found to be 0.12, close to the stationary value of 0.15 given by the calculations of McClelland and Kelley.[10]

Figure 3 shows the measured momentum distribution for the ground-state $3s$ transition compared to that calculated using the PWIA and the Hartree-Fock $3s$-orbital wave function.[12] The agreement is excellent. The finite experimental angular resolution, i.e., momentum resolution, has been folded into the calculations. A distorted-wave impulse approximation (DWIA) calculation carried out by us gave a momentum distribution in complete agreement with the PWIA result.

Figure 4 shows the results obtained for the excited-

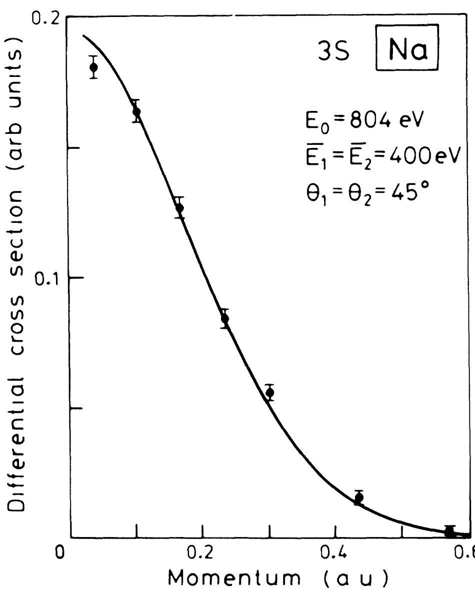

FIG. 3. The 800-eV noncoplanar symmetric momentum profile for the $3s^{-1}$ orbital of sodium, compared with the Hartree-Fock wave function of Clementi and Roetti (Ref. 12). The finite experimental angular resolution has been folded into the calculated momentum profile. The mean summed emitted electron energy is 800 eV, the incident energy being 804 eV.

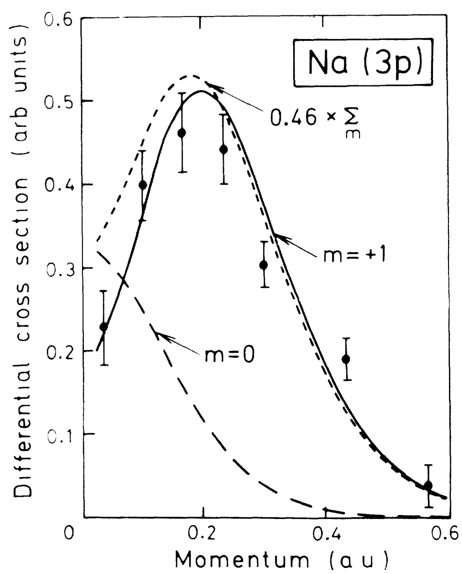

FIG. 4. The measured and calculated momentum profiles for the excited $3p$ state of Na. The solid curve is the calculated $3p(m_l=+1)$ profile, the short-dashed curve the total $3p(m_l=0\pm 1)$ profile, and the long-dashed curve that expected for the $m_l=0$ component. The PWIA calculations use a Hartree-Fock $3p$ wave function (Ref. 13) with the experimental angular resolution folded in. The momentum is probed along the k_x axis, with $\bar{k}_y=\bar{k}_z=0$.

state $3p(m_l=1)$ momentum distribution compared with the momentum distribution given by the Hartree-Fock $3p(m_l=+1)$ wave function.[13] The finite angular resolution has again been included in the calculations. The main effect of the finite angular (momentum) resolution is to fill in the momentum distribution at momenta close to zero. The momentum distribution peaks at very small momenta ($\sim$0.2 a.u.) because of the diffuse nature of the $3p$ orbital in coordinate space. Also shown in the figure for comparison are the momentum distributions expected for the $m_l=0$ substate of the $3p$ state and that for the unoriented $3p_{(0,\pm 1)}$ state. This latter distribution is similar to that for the $m_l=1$ substate, but in significantly poorer agreement with the data. The momentum distribution for the $m_l=0$ (i.e., $3p_z$) state is, of course, zero along the k_x axis ($k_y=k_z=0$), since the $3p_z$ (dumbbell) orbital is perpendicular to the nodal x-y plane, the node being at the origin. However, due to the finite momentum resolution, there is a finite probability of seeing some $m_l=0$ contribution at the origin, this probability decreasing as one moves away from the origin along the k_x (or for that matter the k_y) direction. This explains the "anomalous" shape of the expected $m_l=0$ ($3p_z$) momentum distribution along the k_x axis.

In conclusion, we have demonstrated that EMS can be used to obtain new information on the dynamic electronic structure of excited short-lived species, and of oriented targets.

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