Photoionization Stark spectra of $m_l=1$ excited sodium atoms

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We report experimental results on the photoionization spectra of excited sodium atoms in a well-defined $m_l=1$ substate in the presence of a dc electric field. The various spectra corresponding to a definite polarization of the ionizing light exhibit characteristic features. It appears in addition that they are strikingly perturbed by the Earth's magnetic field.

Since the first observation[1] of narrow resonances in the photoionization spectra of atomic rubidium in the presence of a static electric field, a lot of attention has been paid to photoionization Stark spectra of alkali atoms either in their ground state or in an excited state. Various experiments using atomic beams and pulsed lasers have been carried out, and most of the characteristics of such spectra seem to be understood at least qualitatively. The photoionization Stark spectra in the vicinity of the field-free ionization limit ($E=0$) have been extensively studied both experimentally and theoretically.[2–8] Large undulations as well as more or less broadened structures have been observed. The first is characteristic of hydrogenic behavior, whereas the latter is essentially attributed to symmetry breaking due to the non-Coulombic part of the atomic potential.

It is to be emphasized that in all the above-quoted experiments very little attention has been paid to a rigorously pure definition of the lower state. The aim of this Communication is to report new experimental results concerning the photoionization Stark spectrum of the sodium atom in its $3^2P_{3/2}$, $F=M_F=3$ excited state.

The experiment was performed on an atomic beam of sodium atoms which are subjected to two laser excitations in the presence of a dc field (see Fig. 1). The latter is provided by two parallel grids which have a relatively good transmission for ions as well as for light beams. After being produced in the interaction region, the ions are accelerated by the dc field and extracted through the grid; then, they are deflected by an electrostatic prism towards an electron multiplier. This particular configuration is suitable for excitation of the atoms by a well-defined σ^+ or σ^- polarized light. More precisely, a cw single-mode dye laser, right-hand-circularly polarized, is tuned and servo-locked on the transition $(3^2S_{1/2}, F=2) \rightarrow (3^2P_{3/2}, F=3)$. For this particular transition, only optical pumping between magnetic sublevels may occur, and actually, the use of a cw laser permits the excitation of sodium atoms in the pure state $(3^2P_{3/2}, F=3, M_F=3)$.[9] It is to be emphasized that this particular state corresponds to a well-defined magnetic orbital quantum number, namely, $m_l=1$. The ionizing light is provided by a nitrogen-pumped dye laser having a frequency bandwidth of 0.2 cm^{-1}, and it is continuously tunable over the range extending from $\lambda=408$ to 413 nm. In addition, it can be σ^+, π, or σ^- polarized, so that it permits the study of three different m_l projections of the photoionization spectrum. The experimental configuration is set in such a manner that the $m_l=0$ and 2 photoionization spectra are obtained with the two laser beams counterpropagating collinearly, whereas the $m_l=1$ photoionization spectrum is obtained with the pulsed laser beam propagating perpendicular to the cw one.

In order to check the good definition of the orbital magnetic quantum number $m_l=1$ of the intermediate

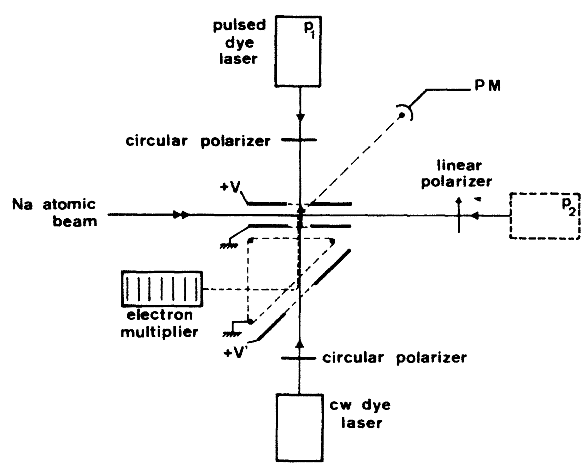

FIG. 1. Experimental setup. Notice the two possible positions of the pulsed dye laser: p_1 corresponding to σ^+ or σ^- excitation, p_2 corresponding to π excitation. The photomultiplier (PM) is to be seen as collecting the resonance fluorescence light in a direction perpendicular to the figure plane; the CW dye laser is servo-locked on the resonance transition.

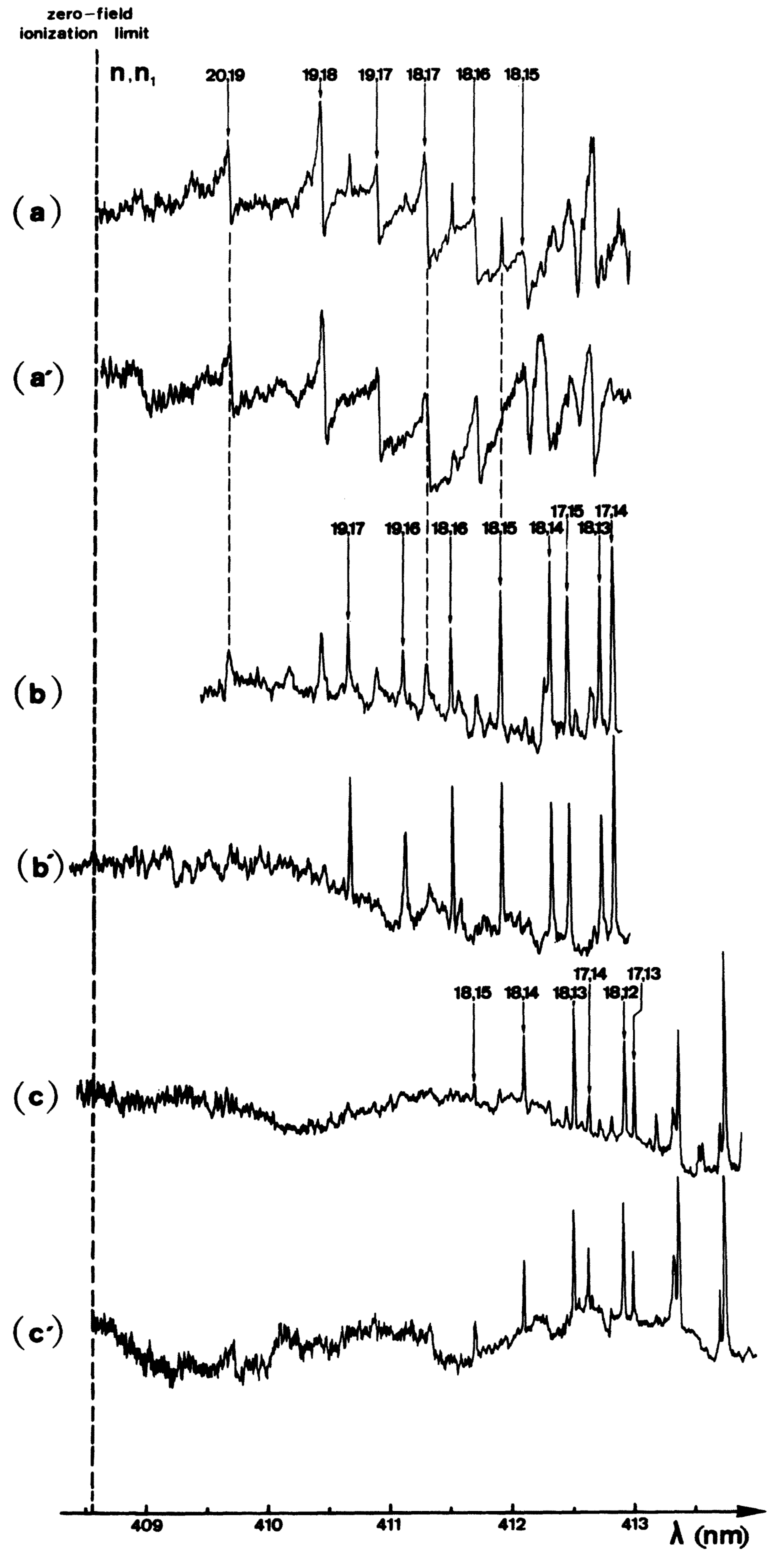

FIG. 2. Recording photoionization spectra for various light polarizations ($F = 9950$ V/cm). For each polarization, two recordings are given, one without canceling the Earth's magnetic field (a)–(c), the other one by canceling the Earth's magnetic field (a')–(c'). Note the disappearance of spurious resonances in the second case. (a), (a') σ^+, $m_l = 0$, $m_j = \frac{1}{2}$; (b), (b') π, $m_l = 1$ $m_j = \frac{3}{2}$; and (c), (c') σ^-, $m_l = 2$, $m_j = \frac{5}{2}$. n and n_1 are parabolic quantum numbers labeling the resonances.

state, two experiments have been performed. On the one hand, we have recorded the unperturbed Rydberg spectra using the field-ionization-detection technique for the two polarizations σ^+ and σ^- of the pulsed laser. In the σ^+ configuration, only d states were observed, whereas in the σ^- configuration both s and d states were recorded. On the other hand, we have recorded the photoionization Stark spectra in the vicinity of the critical ionizing energy value for the three polarizations σ^+, π, and σ^- of the pulsed laser. As previously established these critical energy values do depend on the m_l value of the final state. In accordance with these results we have found three distinct ionization limits for the three values of $m_l = 0, 1, 2$, separated by a total amount of 22 cm^{-1} in a dc field strength of 9950 V/cm (results which are in perfect agreement with the analysis of Cooke and Gallagher[10]). In the studied spectral range it is quite clear that no significant pollution of one spectrum by any of the others is noticeable. After these observations we are able to assert that the intermediate state is a nearly pure $m_l = 1$ state, at least in the experimental limit of accuracy which is of the order of 1%.

Using the above described setup, the photoion current has been recorded versus the pulsed laser frequency for the three polarizations of this laser beam corresponding to the three m_l values. Typical recordings are shown on Figs. 2(a)–2(c). First, one can see that the $m_l = 2$ spectrum [Fig. 2(c)] is quite different from the two others: This proves again that the intermediate state is a pure $M_F = 3$ ($m_l = 1$) state, otherwise the $m_l = 2$ spectrum would have been polluted by resonances similar to those observed in the $m_l = 0$ [Fig. 2(a)] and $m_l = 1$ [Fig. 2(b)] spectra. A second remark concerns the big difference between the resonance shapes in the three spectra: Among others, the $m_l = 0$ spectrum exhibits strongly dissymmetric resonances which look like Fano profiles whereas less dissymmetric resonances mainly appear in $m_l = 1$ spectrum and almost symmetric and narrow ones characterize the $m_l = 2$ spectrum. Such an evolution of the resonance shapes can be interpreted qualitatively as discussed below. In the hydrogen atom, for the considered energy range ($E \lesssim 0$) and static field strength ($F = 9950$ V/cm), quasidiscrete levels (corresponding to high values[11] of the parabolic quantum number n_1) are superimposed to continuum states (corresponding to smaller valus of n_1). The quasidiscrete levels have a very small ionization width, related to tunneling effect, which can be estimated to a few tens of MHz approximately.[9] In a sodium atom, the Rydberg electron sees a non-Coulombic potential, and therefore a coupling appears between the quasidiscrete levels and the underlying continua having the same m_l value. This coupling leads to a broadening of the quasidiscrete levels and, because of interference effects, it manifests in the appearance of asymmetric profiles. Now, the greater the l value, the more hydrogenic is the character it corresponds to, and, therefore, the coupling is expected to strongly decrease as m_l increases. More quantitatively, the width induced by this coupling in the sodium atom has been recently estimated[12] to a few hundreds GHz for $m_l = 0$ levels and to a few GHz for $m_l = 1$ levels located close enough to the field-free ionization limit. In conclusion, the resonances observed in the $m_l = 0$ and spectra are mainly nonhydrogenic, whereas in the $m_l = 2$ spectrum, they are nearly hydrogenic. In this latter case, no resonance appears near $E = 0$ since, in this energy range, the quasidiscrete states interact too weakly with the $m_l = 2$ continua and they cannot be ionized. In the $m_l = 0$ spectrum the resonances vanish rapidly beyond $E = 0$ and are replaced by undulations.[8]

Now, one can try, by using a hydrogenic model, to label the observed resonances by the parabolic quantum numbers n and n_1. For this purpose, we have used perturbation theory to fourth order,[13] and we have shown that only one resonance out of two can be identified; the remaining structure cannot be labeled. More precisely, in the $m_l = 0$ spectrum only the very asymmetric profiles can be identified, while in the $m_l = 1$ spectrum, only the less asymmetric profiles can be labeled. Actually, some resonances appear in the three spectra at the same frequency; this corresponds to an apparent degeneracy in m_l. However, there is no way to understand such a degeneracy in the strong mixing region under study and therefore, one must assume that, in one way or another, the orbital cylindrical symmetry around the dc field direction is broken. Simple evaluations show that this symmetry breaking cannot come out from Stark mixing in the intermediate state: $M_F = 3$ is an extremum value, and in any case, the mixing between 3^2P and other states remains smaller than 10^{-4} for the considered field strength 9950 V/cm. One could also invoke spin-orbit interaction in the ionizing states for breaking the symmetry, but this explanation can be rapidly discarded. First, in a sodium atom, the spin-orbit mixing between the two m_l values ($m_l = m_j \pm \frac{1}{2}$) corresponding to a well-defined m_j value (which remains, of course, a good quantum number) is very small near $E = 0$; it has been recently shown[14] that it does not exceed 10^{-6}. Second, the dipole transition $m_l = 1 \rightarrow m_l = 3$ is obviously forbidden, and therefore, all the resonances observed in the $m_j = \frac{5}{2}$ spectrum should have a pure $m_l = 2$ character and one should be able to identify all of them.

At the present step of the discussion, there is no way to explain cylindrical symmetry breaking from the internal properties of the sodium atom. Thus, the origin of this symmetry breaking must be external. In other words, there is no longer a good magnetic quantum number. Since the polarizations of the two laser beams are very well defined with respect to the electric field direction, the only phenomenon which can break the symmetry is the presence of an external magnetic field (for example, the Earth's magnetic field) in a direction different from that of the electric field. In order to check the validity of this explanation, we have canceled the Earth's magnetic field in the interaction region by using a pair of Helmholtz coils which reduce the magnetic field from 0.8 G (Ref. 15) to zero with an accuracy of 0.02 G. Using this new setup, three spectra have been recorded in exactly the same conditions as previously. The results are displayed on Figs. 2(a′), 2(b′), and 2(c′), and one can be easily convinced of the validity of our previous assumption since all the resonances not identified in the hydrogenic model have now completely vanished. In particular, there are no longer strongly asymmetric profiles in the $m_l=1$ spectrum.

Of course, these results are very surprising considering the weakness of the Earth's magnetic field. However, one must remember that diamagnetic effects become dramatic in highly excited atomic spectra. Moreover, the diamagnetic interaction being a very long-ranged interaction it induces a mixing of states which is more important between continuous states than between bound states. Therefore, all the arguments derived from considerations on unperturbed bound states to evaluate the order of magnitude of the observed effects must be utilized with great caution. Quantitative interpretation is rather difficult, but it is in progress and it will be published elsewhere. Anyhow, we have now an experimental evidence that very weak phenomena can strongly perturb photoionization Stark spectra.

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[14]The m_l mixing induced by the spin-orbit interaction has been evaluated through a diagonalization procedure similar to that already used to analyze the Stark effect in rubidium [S. Liberman, E. Luc-Koenig, and J. Pinard, Phys. Rev. A 20, 519 (1979)]. A 145-states basis has been utilized to diagonalize an energy matrix including non-Coulomb potential energy, Stark operator, spin-orbit interaction, and relativistic effects induced by the presence of the electric field.

[15]The measured magnetic field perpendicular to the electric field is a bit larger than the value expected from the Earth's magnetic field. This means that some residual magnetic fields were present in the atomic beam. One could suspect that such a magnetic field perturbs the atomic alignment obtained in the $3p$ level by the optical pumping process. In this low magnetic field a calculation has shown us that the alignment is better than 99%.