Photoionization spectroscopy of Yb I from the $6s7s\ ^1S_0$ state in the presence of a static electric field

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Laser investigation of the photoionization spectra in the presence of an electric field has been performed in π and σ polarizations on an ytterbium atomic beam from the particular level $4f^{14}6s7s\ ^1S_0$ which was populated by a two-step excitation. Broad modulations (in π polarization) and Stark resonances in the vicinity of the zero-field ionization limit are interpreted satisfactorily as in the case of alkali atoms. However, the photoionization spectra are deeply affected by the presence of two perturbing levels belonging to the configuration $4f^{13}5d^26s$. This phenomenon is explained by strong interference effects between the structured Stark continuum and the field-induced autoionizing states corresponding to the doubly excited levels.

As is well known, the photoionization spectroscopy in the presence of an electric field is achieved by optically exciting atoms submitted to an external static electric field of intensity F for energies E higher than the classical ionization limit $E_c = E_0 - 2\sqrt{F}$ (in atomic units), where E_0 is the energy of the zero-field ionization limit.

The spectra obtained by scanning the frequency of the exciting light and recording the ion signal exhibit narrow resonances in the range $E_c < E \lesssim E_0$, and broad modulations in the range $E \gtrsim E_0$ which depend on the polarization of the excitation. Such spectra have been studied experimentally with alkali atoms (Li,[1] Na,[2–4] Rb[5–7]) and with Ba.[3]

Theoretical analyses have been performed by several authors for hydrogen[7–11] and recently for alkali atoms.[12] The observed spectra are interpreted either in terms of the density of final states or of the absorption cross sections from a given "starting" level. Presently, most authors seem to agree with the latter interpretation. For alkali atoms, the spectra are expected to depend on the l values as well as on the m_l values which are allowed in the final state when making an electric-dipole transition from a given starting level. However a direct comparison with experimental spectra is rather difficult for alkali atoms due to fine or hyperfine structure which introduces several m_l values in the final state even if conveniently polarized light is used to excite the atoms.

The purpose of our work is to investigate thoroughly the influence of the relevant parameters by comparing spectra of the same element obtained in conditions as well defined and as diverse as possible. For this purpose, two-electron atoms seem appropriate and allow us to get rid of fine structure (through the use of $6snl\ ^1L_L$ levels as starting levels) and of hyperfine structure effects (by selecting an even isotope).

Finally we have chosen ytterbium, whose spectrum is, to a large extent, that of a two-electron atom, on the one hand because its eigenstates are rather purely LS coupled and free of configuration interaction, and on the other hand in consideration of the convenience in operating the appropriate lasers at the useful frequencies.

In this Communication we report our results concerning the photoionization spectra from the excited state $4f^{14}6s7s\ ^1S_0$ of Yb I.

The experiment is performed on an atomic beam of natural ytterbium intersected at right angles by three colinear laser beams in the presence of a static electric field perpendicular to all beams. The first-step excitation, provided by a cw dye laser, populates the $4f^{14}6s6p\ ^3P_1$ level from the ground level and the isotopic selection is achieved by locking the laser wavelength on the ^{174}Yb fluorescence line ($\lambda_1 = 555.65$ nm). The two other lasers are pulsed dye lasers pumped by the same nitrogen laser (pulse duration 5 ns, repetition rate 10 Hz). The second-step laser (fixed wavelength $\lambda_2 = 611.13$ nm) excites the $4f^{14}6s7s\ ^1S_0$ level which is used as starting level for the photoionization process ($l = L = 0$ and $m_l = m_L = m_J = 0$). The third-step laser, 5 ns delayed, can be scanned by rotation of the cavity grating in the wavelength range $\lambda_3 = 605$–650 nm.

The static electric field ($F = 1$ to 15 kV/cm) is provided by two parallel plates. The photoions produced are accelerated by this field, they pass through a grid (transmission $\simeq 50\%$) in one of the plates, and are finally collected by an electron multiplier, the output of which is recorded by means of a gate integrator. The spectra are then averaged by means of a multichannel analyzer in order to improve the signal-to-noise ratio and to get rid of the slow drifts. Each spectrum thus corresponds to several scans (10 to 80).

The polarization of the third-step laser is linear, either parallel (π) or perpendicular (σ) to the electric field, allowing us to study, respectively, the $m_l = 0$ and $|m_l| = 1$ states of the photoionization continuum.

The general aspect of the photoionization spectra is illustrated in Fig. 1 in the particular case of π polarization for $F=10$ kV/cm. The photoionization background which is apparent in the range $E<E_c$ exists without the third-step laser, and is due to a reabsorption of the second-step laser from the level $6s7s\ ^1S_0$. The coincidence of the frequencies of the second- and third-step lasers causes also an increase of the photoionization which explains the narrow resonance located at $E-E_0=266$ cm^{-1}.

As in the case of alkali atoms, the π spectrum exhibits narrow Stark resonances ($E_c<E<E_0$) and broad modulations ($E>E_0$). But a striking feature, characteristic of many-electron atoms, is present in the π and σ spectra: a first photoionization dip is observed around $E_1-E_0\simeq-460$ cm^{-1}, where the signal from the $6s7s\ ^1S_0$ level vanishes completely. A similar feature has been observed in photoionization spectra of Ba.[3] This observed dip can be due to the autoionization of a discrete level embedded in a continuum, a level which was discovered by Camus and Tomkins[13] as the "perturber B" for the $4f^{14}6snp$ series and identified as a $J=1$ level of the configuration $4f^{13}5d^26s$.[14,15] Since the energy $E(B)$ of this level [given by $E(B)-E_0=-523$ cm^{-1}] is larger than E_c for $F=10$ kV/cm, the perturber B located inside the continuum becomes an autoionizing level by "forced autoionization."[3,16] In such cases, a Fano profile is expected,[17] but in the present case the picture is complicated by the presence of the sharp Stark resonances. However, the behavior of the photoionizing background (minimum for $E=E_1$ and asymmetric shape around E_1) is characteristic of such a profile.

In order to check this hypothesis we have performed a more quantitative comparison between our experimental curves and the expected autoionization profiles. The general form of the profile is given by[17]

$$y(\epsilon)=\frac{(\epsilon+q)^2}{\epsilon^2+1}$$

with

$$\epsilon=\frac{E-E(B)}{\Gamma/2},\quad q=\frac{E(B)-E_1}{\Gamma/2},$$

where Γ is the width of the autoionization resonance. The value of $E(B)$ is obtained from the study of the field-free spectrum [we assume that $E(B)$ is not changed by F]; the value of $\Gamma=100$ cm^{-1} is deduced from the MQDT (multichannel quantum-defect theory) analysis of the $4f^{14}6snp$ ($J=1$) series by simply taking the half-maximum width of the percentage function of perturber B in the Rydberg series.[15] The parameter $q=-1.26$ is determined with our experimental value E_1 for the minimum. Finally a scale intensity factor is chosen so as to fit approximately the experimental spectrum. One finally obtains the broken curve of Fig. 1. Below E_c, this calculated curve is no longer valid, as expected, due to the detection process. Owing to these considerations, the agreement between the experimental and theoretical profiles can be considered as quite satisfactory.

A similar comparison has been made for the second observed dip at $E\simeq E_0$ with the following values: $E(C)-E_0=-73$ cm^{-1}, $q=-2.44$, and $\Gamma=60$ cm^{-1} corresponding to the perturber C identified as a $J=1$ level of the configuration $4f^{13}5d^26s$.[15] The agreement is slightly worse, but in that case the energy of the perturber C is only deduced from the MQDT analysis of the spectrum (this level has not yet been observed in the field-free spectrum).

Figure 2(a) shows that the energies of the Stark resonances in the energy range -400 cm$^{-1}<E-E_0<0$ cm^{-1} are rather well described by the perturbation expansion for hydrogen at fourth order of the field.[18] The observed resonances in the π and σ photoionization spectra can be labeled (n,n_1,m_l), n and n_1 being the two usual parabolic quantum numbers, the other quantum number n_2 being given by $n_1+n_2+|m_l|+1=n$.

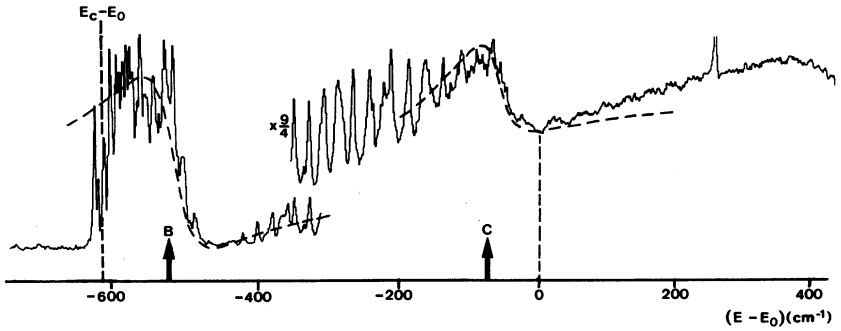

FIG. 1. Photoionization spectrum from the $6s7s\ ^1S_0$ level for $F=10$ kV/cm in π polarization ($m_l=0$). The gain of the second part of the spectrum is multiplied by a factor $\frac{9}{4}$. E_0 and E_c are, respectively, the zero-field ionization limit and the continuum limit for $F=10$ kV/cm. The arrows indicate the positions of the perturbing levels denoted B and C in the text. The broken curves are calculated Fano profiles (see text).

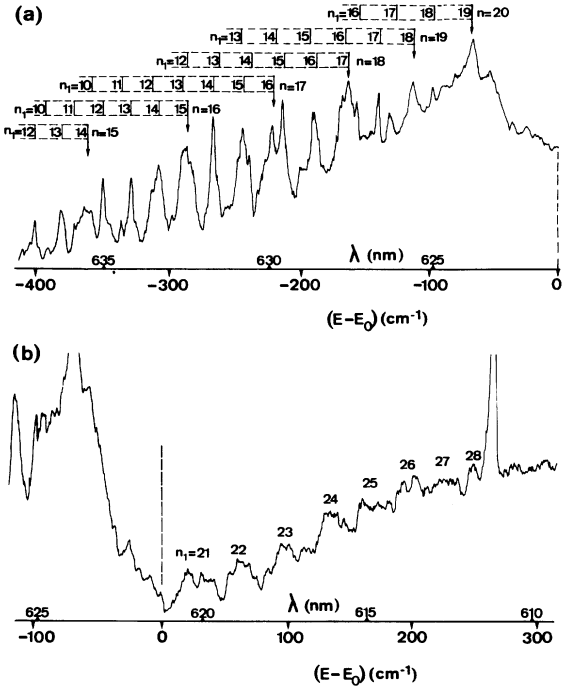

FIG. 2. Improved and expanded $\pi(m_l=0)$ photoionization spectra from the $6s7s\,{}^1S_0$ level for $F=10$ kV/cm. (a) (n,n_1) assignment and calculated energies of the Stark resonances in the region between the first dip and the zero-field ionization limit. The photoionization background has been subtracted from the total signal. (b) (n_1) assignment of the modulation peaks in the region above the zero-field ionization limit. The gain is multiplied by a factor 3 with respect to (a) and an offset 1.5 times the intensity of the 625.5-nm Stark resonance is introduced.

However, for $F=10$ kV/cm, the absolute values of the successive terms of the perturbation expansion decrease rapidly only for $n \leq 17$. In the range $n=18$–20 the third-order and fourth-order terms W_3 and W_4 are of opposite signs and comparable for $n_1-n_2 \gg 1$. Taking into account the expected oscillating, asymptotic behavior of the series,[19] we have found that a good agreement between experimental and calculated energies is obtained by replacing W_3+W_4 by the expression $W_3^2/(W_3-W_4)$. This is the so-called geometric approximation which has proved to give accurate results in many problems of atomic physics when the sum of a slow-converging or even nonconverging series has to be estimated from its first few terms.[20]

The photoionization spectra in the energy range $-650 < E-E_0 < -400$ cm^{-1} cannot be interpreted with this simple model. In fact, in this perturbed region, the photoionization cross section must be calculated by mixing the Stark resonances, the Stark continuum, and the field-induced autoionizing resonance corresponding to the perturber B.[4,16] The individual Stark resonances may have particular Fano profiles, and the assignment and labeling of the resonances require a complete calculation out of the scope of this Communication.

In the region $E \gtrsim E_0$, the expected broad modulations are clearly observed for π polarization in Fig. 2(b), but they disappear for σ polarization. This behavior is in agreement with the comparable previous results on Rb,[5,7] and with the theoretical predictions.[9,11] The amplitude of the modulation for π polarization is about 5% near the zero-field ionization limit and decreases regularly for higher energies. The n_1 values in Fig. 2(b) are attributed by continuity from the lower-lying Stark resonances. However the value of n_1 for $E=E_0$ is given by the following formula[11]:

$$n_1 \simeq -\tfrac{1}{2} + 37.5[F(\text{kV/cm})]^{-1/4} .$$

For $F=10$ kV/cm, one thus obtains $n_1 \simeq 20.6$, in agreement with the extrapolated value of Fig. 2(b).

The theoretical value for the spacing between adjacent peaks is given by[8,9]

$$\frac{dE}{dn_1} = 7.50[F(\text{kV/cm})]^{3/4}$$

and decreases for higher values of the energy. In order to verify this prediction, we measured the position of four or five successive modulation minima

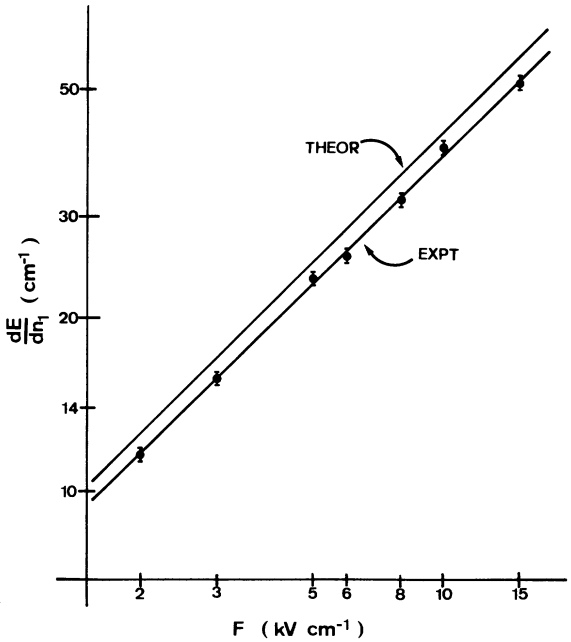

FIG. 3. Log-log plot of the spacing between adjacent modulation minima around the zero-field ionization limit as a function of the electric field F. The straight lines THEOR and EXPT correspond, respectively, to the theoretical formula and to the experimental results.

around E_0 as a function of n_1 and did a quadratic interpolation to determine the slope at $E = E_0$. The corresponding log-log plot for values of F ranging from 2 to 15 kV/cm is given in Fig. 3. The $F^{3/4}$ law is obeyed accurately but the experimental spacing appears to be 7% smaller than the theoretical one.

These specific results on the photoionization from the $6s7s\ ^1S_0$ level must now be compared with the results that we obtained on the photoionization in the presence of an electric field from other starting levels with values of $l = 0$ to 2, and corresponding values of m_l. This work will be published soon.

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