Observation of Fano Profiles in Photoionization of Rubidium in the Presence of a dc Field

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We report the observation of Fano profiles in the π-photoionization spectrum of atomic rubidium in the presence of a dc field. Such an observation has been made possible thanks to the use of very high-resolution techniques. It can be considered as an experimental proof of the validity of the "nonhydrogenic" process suggested recently to explain ionization in a static electric field for all atoms except hydrogen.

A lot of attention has been recently paid to the Stark structure of highly excited atoms and to the processes by which free atoms ionize in a static electric field.[1] Various experiments using atomic beams and pulsed lasers have been carried out and finally, it appears that the spectrum of an alkali atom in the presence of a dc field, F, is well separated in two quite different regions by a limit given, in atomic units, by[2]

$$E_c = E_0 - 2F^{1/2},$$

E_0 being the ionization energy of the unperturbed atom. E_c is nothing else but the energy of the saddle point which appears in the potential energy surface of an electron interacting simultaneously with a Coulomb field and a dc field. In fact, it was recently pointed out by Cooke and Gallagher[3] that E_c must be replaced for states with $|M_L| \neq 0$ by $E_c' = E_0 - 2F^{1/2} + |M_L| F^{3/4} + (3/16) M_L^2 F$. This formula being valid only for the case $|M_L| \ll n$. Since F is always much smaller than 1 a.u., E_c' is very close to E_c for $|M_L| = 1$. In any case, below this saddle-point limit, the outer electron is quasibounded and the spectrum consists in quasistable Stark levels which ionize, of course, but with very small rates (smaller than 10^5 s^{-1}). Above the saddle-point limit, such quasistable Stark levels disappear but the spectrum exhibits sharp ionization resonances superimposed to ionization continua. Such sharp resonances were first observed in rubidium and it was shown that, in this case, they are sufficiently narrow to be isotopically selective at least if σ light (exciting states with $M_L = \pm 1$) is used.[4] With π-polarized light (exciting states with $M_L = 0$), some resonances still appear but they are much broader. A more extensive study at lower resolution was recently performed on lithium by Littman, Kash, and Kleppner.[5] The results are quite similar and their main interest is to show that these resonances exist on a very large spectral range and appear as soon as the excitation energy becomes higher than E_c. Moreover, by following the evolution of the Stark states with an increasing strength of the dc field, Littman, Kash, and Kleppner were able to show that the Stark states not only ionize significantly as soon as they reach the saddle-point limit, but also suddenly broaden at a much higher value of the field before merging with the continuum.

Starting from their experimental data on lithium, Littman, Kash, and Kleppner identified two different processes by which a free atom ionizes in a static electric field. The first one is a pure tunneling effect which can be quantitatively studied using a pure hydrogenic model. It is well known that, because of separability in parabolic coordinates, the problem of a hydrogen atom in a dc electric field can be exactly solved.[2] Many exact calculations are now available[6] and all of them show the presence, far above the saddle-point limit, of quasistable Stark levels. This can be easily understood if one notices that a potential barrier (which depends not only on F but also on M_L and on the separation constant Z_2) appears for the η parabolic coordinate. For example, for $|M_L| = 1$, the energy of this barrier is given by[2]

$$E_c^H = E_0 - 2(Z_2 F)^{1/2}.$$

Since Z_2 depends on F in a complicated way, it is no longer possible to express E_c^H as a simple function of F. However, one knows that, for highly excited states, Z_2 can take very small values (of the order of $1/n$). Therefore, for some Stark states, E_c^H can be much higher than E_c and some quasistable Stark levels can exist above the saddle-point limit. Of course, because of tunneling some levels can have a nonnegligible ionization probability for E values slightly lower than E_c^H but there is no connection between this phenomenon and the saddle-point limit. Thus, another process must be taken into account to fully explain the Stark structure of a nonhydrogenic atom. This second process was recently proposed by Littman, Kash, and Kleppner as a consequence of symmetry breaking.[5] In fact, even in hydrogen, above the saddle-point limit, quasistable Stark levels are enbedded in ionization continua coming from upper levels but, but, because of separability in parabolic coordinates which introduces supplementary good quantum numbers (and therefore, selection rules) there is no interaction between the quasistable Stark levels and the ionization continua. In contradistinction, as soon as separability disappears, and this is the case for any nonhydrogenic atom, such an interaction takes place and Stark levels ionize through a mechanism similar to autoionization in many electron atoms or to predissociation in molecules. Such an explanation is, of course, quite convincing since it allows one to understand qualitatively all the characteristics of the Stark structure of an alkali atom but an experimental proof is highly desirable nevertheless. Now, it is well known that the signature of the interaction between a discrete level and a continuum is the appearance of interference effects giving rise to characteristically asymmetric peaks (so-called Fano profiles[7]) in excitation spectra.[8] The aim of this Letter is to report the first observation of such Fano profiles in the photoionization spectrum of an alkali atom in the presence of a dc field.

From previous observations, it was clear that such an observation was required to study atomic states as nonhydrogenic as possible and to use very high-resolution laser techniques. Therefore, the best choice was to excite $M_L = 0$ states of a heavy alkali atom (since S states have the largest quantum defect, Stark states with $M_L = 0$ are obviously the most nonhydrogenic) by using a single-mode pulsed tunable laser and a collimated atomic beam. These conditions were already fulfilled in our previous experiment on rubidium and, in fact, with a π excitation, we already recognized some structures presenting some similarities with a Fano profile, but the signal-to-noise ratio was too poor to lead to a definite conclusion.[4] The principle of our new experiment is thus the same as the previous one but some technical improvements have been made. The main elements of the experimental setup are a moderately collimated rubidium thermal beam (collimation ratio, 90) and a pulsed dye laser working in single-mode operation.[9] The laser pulses are frequency doubled and with this arrangement, they have a duration of 60 ns, a peak power of 200 W, and a width less than 40 MHz in the uv range. However, because of residual Doppler effect, the resolution limit is finally 50 MHz.

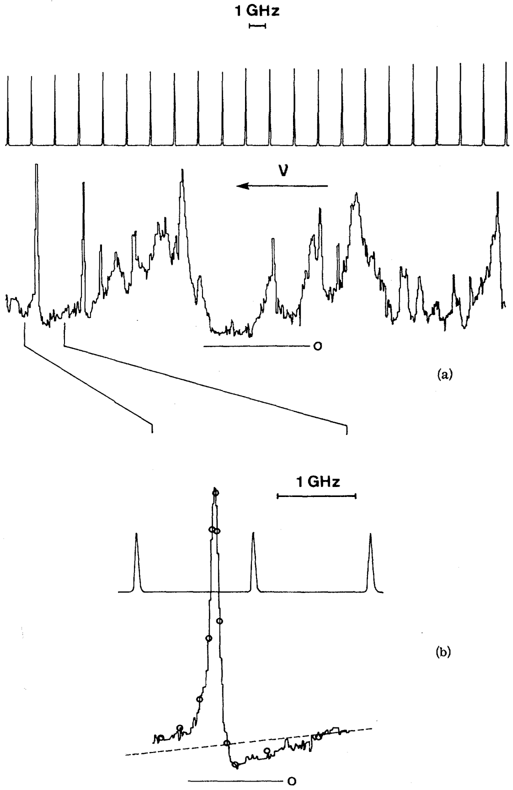

FIG. 1. (a) Photoionization spectrum of rubidium in the presence of a dc field, $F = 158$ V/cm, for an excitation energy close to 33.614 cm^{-1}, and a light polarization parallel to the field. (b) Example of characteristically asymmetric profile. Dots represent the best fit to the Fano formula leading to the parameters given in the text. This fit has been obtained by assuming a linear variation of the ionization background vs the excitation energy (dotted line). (Fabry-Perot fringes provide the frequency scale.)

The static electric field is premanently applied to the atoms interacting with the laser light. The produced ions are counted just after the end of the laser pulse and during a few microseconds. With the use of this setup, systematic studies have been undertaken for various field strengths and different excitation energies. Many asymmetric resonances have already been observed and, by slowly varying the field strength, we have verified that this asymmetric character is not coming from an unresolved multiplet Stark structure. Furthermore, the only multiplet structure independent of the field which could be put forward is the hyperfine structure of the ground state, but this structure is much larger[4] than the observed width of the resonances. A typical profile recorded with a field equal to 158 V/cm and for an excitation energy close to 33.614 cm^{-1} (corresponding to an energy located between the $40p$ and $41p$ levels for an unperturbed atom) is shown in Fig. 1, which also gives the best fit to the Fano theoretical formula. The corresponding parameters are

$$\Gamma = 0.13 \text{ GHz and } q = 3.3.$$

Of course, before entering upon a quantitative interpretation of such parameters, a more extensive study on a broader spectral range and for many different values of the applied field must be carried out. However, our present results already demonstrate unambigously the validity of the process suggested by Littman, Kash, and Kleppner and show that it is possible to obtain experimentally precise values of the parameters which characterize the interaction between a quasistable Stark level and the underlying ionization continua. In addition, it must be noticed that the resonances are superimposed to strong oscillations of the continuum, the period being approximately 13 GHz. We have not yet found a definite explanation of such oscillations but it must be emphasized that the phenomenon exhibits some similarities with the broad resonances observed by Freeman *et al.*[10]

Further experiments are certainly needed.

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[1] For references see, for example, Proceedings of a Conference on Atomic and Molecular States Coupled to a Continuum, Colloq. Int. CNRS No. 273, pp. 55–317.

[2] H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Springer, Berlin, 1957).

[3] W. Cooke and T. Gallagher, Phys. Rev. A 17, 1226 (1978).

[4] S. Feneuille, S. Liberman, J. Pinard, and P. Jacquinot, C. R. Acad. Sci., Ser. B 284, 291 (1977); P. Jacquinot, S. Liberman, and J. Pinard, Colloq. Int. CNRS 273, 215–226 (1977); S. Liberman and J. Pinard, Phys. Rev. A (to be published).

[5] M. G. Littman, M. M. Kash, and D. Kleppner, Phys. Rev. Lett. 41, 103 (1978).

[6] See, for example, R. J. Damburg and V. V. Kolosov, J. Phys. B 9, 3149 (1976), and Phys. Lett. 61A, 233 (1977), and Opt. Commun. 24, 211 (1978).

[7] U. Fano, Phys. Rev. 124, 1866 (1961).

[8] S. Feneuille and P. Jacquinot, Atomic Physics-6 (Plenum, to be published).

[9] J. Pinard and S. Liberman, Opt. Commun. 20, 344 (1977).

[10] R. R. Freeman, N. P. Economou, G. G. Bjorklund, and K. T. Lu, Phys. Rev. Lett. 41, 1463 (1978).

Polymorphism in a Lamellar Liquid-Crystal Bilayer System

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The lamellar liquid-crystal phase, L_α, of the potassium-palmitate–water system is shown to be actually polymorphic. Using deuterium magnetic resonance as well as birefringence observations, two sharp phase transitions are observed in the simple neat soap bilayers. Optical conoscopy shows the high-temperature phase to be uniaxial and the other two to be biaxial. Deuteron magnetic resonance shows that each phase transition is accompanied by a change in orientational order.

The lamellar liquid-crystal phase, L_α, in bilayer systems has not generally been regarded as being polymorphic[1] although polymorphism in membrane bilayers has been observed[2] and peculiar behavior suggestive of phase transitions has been observed in nuclear magnetic resonance (NMR) measurements of self-diffusion,[3] spin-lattice relaxation,[4] and quadrupolar splitting[5] in some simple soap systems. In this Letter, we wish to show both NMR and optical evidence for two distinct phase transitions within the lamellar L_α (soap boilers' neat soap) region of the potassium-palmitate–water system as the temperature is varied. This region is at temperatures above the gel phase and below the superneat phase of this system. The NMR studies were made from perdeuterated samples of potassium palmitate and the optical studies from nondeuterated sam-