GENERATION OF QUASI–FANO PROFILES IN THE LOW FIELD STARK SPECTRUM OF CAESIUM

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We present the experimental evidence for a few elementary processes occurring in Stark effect for non-hydrogenic atoms. They are likely to play an important role in the field redistribution of non-hydrogenic states onto the parabolic hydrogenic channels. We only consider the low field quasi-bound part of the spectra. We show in particular that the interaction between non-hydrogenic states and the incomplete hydrogenic manifold (exhibiting a linear Stark behaviour) leads to the generation of quasi-Fano interference profiles well described in the framework of "interaction of one *discrete* state with a quasi-continuum of *discrete* states". Some examples of accidental decoupling of states from the manifold are also shown leading to peculiar features throughout the spectrum.

1. Introduction

Although the mechanisms of Stark effect in non-hydrogenic atoms look complicated at first sight, the most basic features can indeed be understood from very simple models in quantum mechanics. Our purpose is here to experimentally illustrate some of these, in their most elementary form, for the quasi-bound Stark spectra at very low field strengths. Namely, we deal with the Stark redistribution of non-hydrogenic states onto the parabolic eigenchannels which roughly characterize the long range asymptotic behaviour of the one electron system in the *E* field. Such a redistribution is shown to obey general rules which are those of "one discrete state interacting with a quasi-continuum of discrete states" and consequently leads to the generation of quasi-Fano interference profiles. Similar phenomena have previously been reported in the bound-free spectra at high-field strengths [1,2]. We experimentally show here that this is quite general a behaviour [3] not necessarily involving the pre-ionized channels.

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2. Theoretical

The reference situation for the understanding of Stark effect in one-electron atoms is that of hydrogen. The solution of such a problem has long been known [4] owing to the separability of Schrödinger's equation in parabolic coordinates. There are three exact constants of the motion, which are the energy and the projections on the field axis of the angular momentum and the generalized Lenz vector [5]. The latter is associated with the dynamical symmetry in the problem which results in the general crossing behaviour of the energy levels.

Practically, each n^2 fold degenerate Coulomb manifold is split, for each $L_z = M$ value, into a Stark manifold with $n - |M|$ sublevels [6]. In the low field *linear Stark regime*, which holds on a large range of field strengths (up to the classical ionization field), the spacing of the sublevels is twice the linear Stark frequency, $\omega_S = 3nE/2$ (au).

How to extend such an analysis to one electron Rydberg atom (as alkali atoms) in which there is breaking of the dynamical symmetry due to non-coulombic corrections to the potential is described in general terms in ref. [7,8]. Such treatments aim at a generalization of the usual quantum defect theory in the parabolic basis, as the latter is still dominating the

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asymptotic behaviour of the system (while the symmetry is of spherical type near the core). The Stark spectrum of one electron atom is then similar to the one of hydrogen, for the most part of the states. But the intensity spectrum is usually different while a general non-crossing rule is obeyed [8,9].

For our present concern – the low field redistribution of a non-hydrogenic state onto the parabolic eigenchannels (see fig. 1) – we choose to develop a simpler model referring to the "interaction of one discrete state with a quasi-continuum of discrete states". This involves well-known and general physical mechanisms and can be fruitfully extended to the description of the one electron Rydberg atom in various external field conditions [3,10,11].

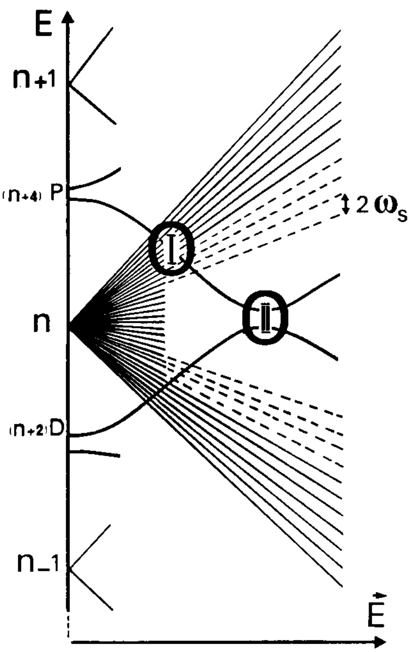

Fig. 1. Sketch of the experimental situations (M = 1 Stark spectrum). The incomplete manifold spectrum is composed of a set of equally spaced sublevels with a spacing $2\omega_S = 3nE$ and exhibits a linear Stark behaviour. The $(n + 4)$P and $(n + 3)$D states are out of the manifold. Upon crossing of their diabatic energy curves with the manifold components, the generation of Fano profiles takes place. (Region I) – Then at higher fields the curves diabatically issuing from the Stark mixed $[(N = 4)\mathrm{P}, (n + 3)\mathrm{D}]$ and $[(n + 3)\mathrm{P}, (n + 2)\mathrm{D}]$ states do anti-cross which results in the accidental decoupling of one state from the manifolds. (Region II) – Notice that the P and D states are situated approximately half distance between two manifolds.

The key concept is the one of "incomplete hydrogenic manifold". It introduces itself quite naturally from the remark that the quantum defects of alkali atoms are negligibly small for the states with angular momenta $l \geqslant 3$. Hence, such states are nearly degenerate in zero field and exhibit a linear Stark behaviour once (δ the quantum defects):

$$\delta(l > 3)/n^3 \ll \tfrac{3}{2} E n^2 . \qquad (1)$$

In contrast, the states with $l < 3$ are out of such manifold and do present a non-hydrogenic behaviour at low fields. As L_z is still a constant of the motion, the manifold is complete for states with $M \geqslant 3$ while incomplete for $M < 3$ (lacking 1, 2 or 3 "spherical" states).

Then, the problem of Stark effect in non-hydrogenic atoms amounts to determining the eigenstates and eigenvalues of the hamiltonian in terms of these two classes of zero-field eigenstates which do interact at higher fields. This leads to a "Stark redistribution" of the discrete non-hydrogenic states onto the incomplete hydrogenic manifold acting as a quasi-continuum of discrete states. Such a situation is then similar to the ones considered in Fano [12] or better in Cohen-Tannoudji and Avan [13].

This allows us to answer on general grounds the two key questions, the first one being "what are the properties of an *incomplete* manifold"?

From the model of ref. [13], one discrete state interacting with a set of N discrete states considered in the *strong coupling* limit, it is readily deduced that the spectrum of the incomplete manifold is bounded by the one of the complete manifold. Their (linear) behaviour with E field is similar but the absolute positions of the energy levels do not coincide. The spacings (here the linear Stark frequency) do coincide (to within $1/n$ order corrections. Obviously, the wavefunctions and hence oscillator strengths distributions should completely differ (some essential contributions from spherical harmonics are lacking in the parabolic channels). A detailed theoretical analysis based on the SO(4) Coulomb supersymmetry will be reported elsewhere, under various external field conditions [14,15].

The second key point is "how does the transition between the incomplete and complete manifold situation take place under the field action"? This is the problem of Stark redistribution which is now easy to understand from the results of refs. [12,13]. Following the strength of the coupling V compared to the spacing ω_S and mean extension $n\omega_S$ of the manifold, there are three types of regimes [3,12,13]. The weak coupling case ($V/\omega_S \ll 1$) leads to the familiar anti-crossing situation. The intermediate coupling case ($V/\omega_S \gtrsim 1$) is associated with the generation of quasi-Fano interference profiles [13] (provided the various channels are optically excited), as the non-hydrogenic state interacts with several sublevels in the manifold. At last, in the strong coupling regime ($V^2/\omega_S \gtrsim n\omega_S$), the redistribution of the discrete state onto the incomplete manifold is total. The coupling constant V is slowly varying a function on the manifold states and depends on E, n and on the non-hydrogenic state under consideration (e.g. the F states are completely redistributed at the lowest field values while the S, P, D are not).

In such a situation which only involves discrete states, the quasi-Fano phenomenon affects the intensity of the discrete lines, the envelop of which being Fano shaped. The experiments described hereafter being Doppler limited, the recorded signal will be the convolution of such quasi-Fano profiles with the Doppler one, which at low fields *restores some continuous aspect* to the phenomena.

3. Experimental

The experiments have been performed on high lying series of caesium atoms using c.w. dye laser excitation and thermoionic detection. The excitation scheme and main features of the experimental apparatus have been previously described [16,17]. The electric field values are in the range 0–14 V/cm. The "hybrid resonance" [17] excitation scheme allows to populate the $M = \pm 3$ Rydberg series using σ polarized laser light and merely the $M = \pm 1$ Rydberg series with π polarization. Owing to the very small quantum defect of the F state ($\delta = 0.033$), the $M = \pm 3$ states do act as a complete hydrogenic manifold in the $\boldsymbol{E}$ field. As reported in refs. [10,17], they exhibit a nearly perfect linear Stark effect (see fig. 3 of ref. [17]).

As to the π spectrum of $M = \pm 1$ states, the situation is not the same. The P and D series with highly non-hydrogenic behaviour (their quantum defects are $\delta(P) = 3.57$; $\delta(D) = 2.47$) are involved in their Stark effect. At low fields, the manifold of $M = \pm 1$ states is incomplete as it lacks the contribution of the P and D states. We will discuss two types of phenomena taking place during the field redistribution of these states on the incomplete manifold. This is sketched in fig. 1.

Region (I) provides one with a good example of interaction between a discrete non-hydrogenic state and the incomplete manifold, in the intermediate coupling case. At low fields, the $M = 1$, P and D states are out of the incomplete manifold and exhibit a non-hydrogenic Stark behaviour. But as a consequence of the quasi-degeneracy of the $(n + 4)$ P and $(n + 3)$ D energy levels, they are completely Stark mixed at low fields. The resulting eigenstates are then the proper linear combinations built on the P and D wavefunctions. In this region, the lines have the Doppler width. At higher fields, one of the components of these Stark mixed "P, D" states interacts with the n incomplete manifold. The coupling is large enough compared to the linear Stark frequency ω_S in order that the interaction take place with several sublevels in the manifold. Both channels being optically excited, such a situation gives rise to quasi-Fano interference profiles. This is shown in fig. 2 for the $M = 1$ component of the (54 P, 53 D) Stark mixed states interacting with the extreme components of the $n = 50$, $M = 1$ incomplete manifold.

Fig. 2. Generation of Fano profiles in the interaction of the 54P, 53D; $M = 1$) non-hydrogenic state with the incomplete ($n = 50$; $M = 1$) manifold (electric field value $E = 4.5$ V/cm). Although the components of the manifold are not resolved here, their interaction with the (P, D) state in the intermediate coupling regime generates both an enlargement of the line and an asymmetry (the experimental Pano parameters are $\gamma = 3.5$ GHz, $q = 2$). The numerical calculation (dotted curve) leads to $\gamma = 3.6$ GHz and $q = 2.3$ (upper trace – the molecular iodine spectrum). Such a phenomenon refers to region I in fig. 1.

The subcomponents of the manifold are not resolved for such small values of the E field (ω_S is smaller than the Doppler width).

Nevertheless, their presence is revealed through the *perturbation of the optical line involving the discrete state*. They are responsible for the enlargement and asymmetry of the optical line which can be studied from n = 40 to n = 100. In the situation of fig. 2, the experimental parameters of the Fano profile are q = 2.0 and γ = 3.5 GHz (the Doppler width being 1 GHz). The theoretical profile shown for comparison [14] (without correcting for the effects of the wings of the manifold on the optical profile nor doing any deconvolution) leads to q = 2.3 and γ = 3.6 GHz which is in reasonable agreement with the experiments.

Other examples of quasi-Fano profiles are displayed on the spectral trace in fig. 3. The broad patterns around the n = 60 and n = 61 zero-field positions are the M = 1 manifolds of states, the components of which are not resolved (ω_S is smaller than the Doppler width). The other (obviously periodic) structures are those associated with the non-hydrogenic lines. The two broad lines on the wings of the n = 60 manifold are associated with the Stark mixed (63 P, 62 D; M = 1) (low energy side) and (64 P, 63 D; M = 1) (high energy side) states (see fig. 1). The remaining sharp lines are associated with (M = 2, D) states (the result of a draw-back in the excitation process). The coupling of such states with others being weak, the latter lines have the Doppler width. This is contrasting with those associated with the M = 1 states. Due to the interactions with the manifolds states their widths are several times the Doppler one and the lines are asymmetric. Lacking the interpretation given above in terms of quasi-Fano profiles generation, such behaviours in the spectra would have been puzzling.

Such a quasi-Fano regime generalizes the familiar Stark anticrossings regime. Previous experiments on the quasi-bound part of the spectrum at high resolution bear the evidence of the phenomena which so far have not been interpreted this way (see e.g. the various discrete patterns in ref. [18]).

Region (II) in fig. 1 provides us with another example of interference effects occurring in the Stark spectrum of non-hydrogenic atoms which we can qualify as "accidental decoupling" of one state from the manifold. It may happen that the interaction of two non-hydrogenic states coupled to several incomplete manifolds leads to unexpected simple behaviours at their anticrossings. We here discuss a situation in which an eigenstate of the Stark hamiltonian turns out to be completely decoupled from the manifolds at *a non-*

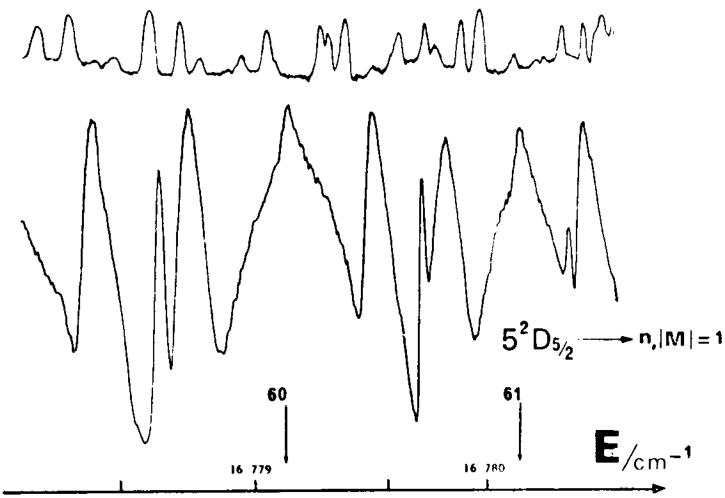

Fig. 3. Stark spectrum for M = 1 states showing several examples of quasi-Fano interference profiles (electric field E = 1.9 V/cm). The components of the manifold are not resolved (here ω_S < 1 GHz) and the manifold appears as a broad pattern around the zero-field positions (for n = 60, 61). The lines associated with the P and D non-hydrogenic states are asymmetric and broadened as a consequence of their interaction with the manifold states (intermediate coupling case with $V \gtrsim \omega_S$) (upper trace – molecular iodine spectrum).

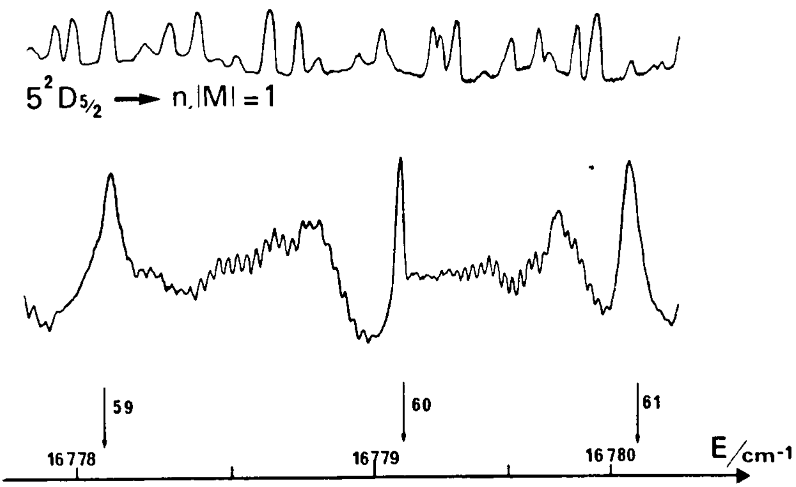

Fig. 4. Accidental decoupling in the M = 1 spectrum around n = 60 (electric field value E = 4.25 V/cm). The accidental decoupling of a Stark mixed "P, D" state from the manifold is clearly exhibited on the plot for n = 60 (while the decoupling is not complete for n = 59 and 61 which results in a broadening of the lines). Such a decoupling nearly occurs at the zero field position of the incomplete manifold (this refers to region II of fig. (1)). The components on the extreme wings of the incomplete manifold are clearly seen on the record. Their intensity distribution is Fano-shaped as a consequence of the strong interaction with the other ("P, D") component.

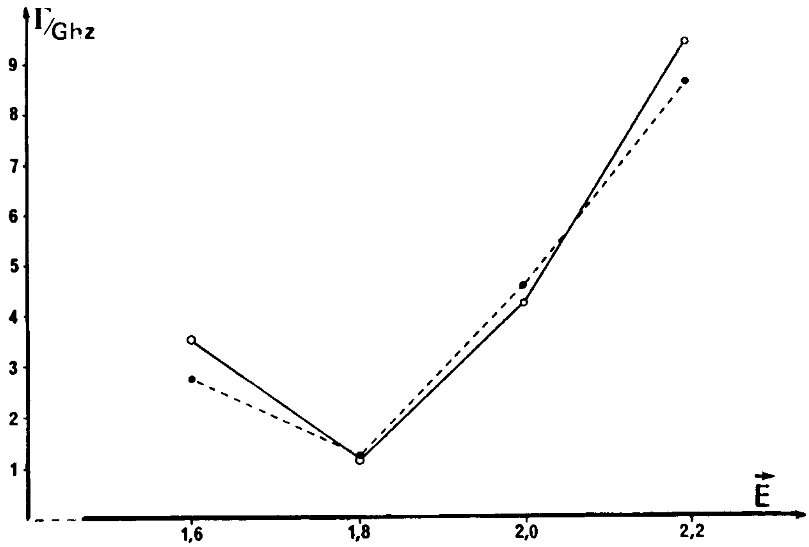

Fig. 5. Plot of the linewidth of the decoupled "P, D" state as a function of the field strength. Γ measures the degree of coupling with the incomplete manifold. This refers to region II of fig. 1 (also see fig. (4)). The accidental decoupling nearly occurs at the zero-field position of the incomplete manifold and fulfills a $n^5E \simeq 1$ law. This is shown for n = 60. The minimum value of Γ is 1 GHz (the Doppler width) which is associated with zero coupling with the manifold. The results of a numerical theory for n = 60 are shown for comparison (dotted line).

zero field value. If such a state is optically accessible, this results in the (local in field) appearance in the spectra of a line having the Doppler width and no asymmetry (as the coupling with the manifolds locally disappears).

Such a sharp phenomenon is shown in fig. 4 under conditions which correspond to region II of fig. 1. It is associated with the anticrossings of the Stark mixed $((n+4)P, (n+3)D)$ and $((n+3)P, (n+2)D)$ non hydrogenic states (conveniently combined). From fig. 1, such an anticrossing occurs for a field value E_0 such that $E_0 \sim n^{-5}$ (au). This law has been checked for n values between 40 and 100. The fact that the decoupling conditions are critical is clearly illustrated in fig. 4. The decoupling is perfect for $n = 60$ while not exactly achieved at $n = 59$ and 61, which results in a broadening and asymmetry of the lines due to the non-zero coupling with the manifold. The $n = 60$ "P, D" line has in turn exactly the Doppler width while its intensity is comparable to the zero-field one.

The extreme components of the manifolds are seen on the record of fig. 4. But the intensity patterns exhibit some striking features being again Fano-shaped. This is the result of the interaction of the manifold components with the second state (lower energy one) in region II (see fig. 1) which, to the contrast of the decoupled one, is still strongly coupled to the manifold (strong coupling situation).

A numerical theory [14] of the decoupling phenomenon allows us to deduce the width Γ of the "decoupled" line as a function of the field strength. This is shown in fig. 5 in comparison with the experimental values for $n = 60$. As the excess of the width Γ over the Doppler width (1 GHz) roughly measures the degree of coupling with the quasi-continua, this confirms the existence of a decoupling of the states which is local in the field. Similar phenomena are likely to occur when dealing with the bound-free Stark spectrum (they lead to the so-called stabilization effects [19]) or in molecules.

4. Conclusion

The experimental evidence for a few simple but basic mechanism in Stark effect for non-hydrogenic atoms has been carried out in its most elementary form, for the quasi-bound Stark spectrum. Although no preionized channels are involved, the redistribution process of non-hydrogenic states has been shown to generate quasi-Fano interference profiles. The experimental situation at low fields also turns out to be a nearly perfect playground for studying the interaction of one discrete state with a set of equally spaced discrete states acting as a quasi-continuum [13]. The concept of incomplete hydrogenic manifold which we use here appears to be essential for dealing with other situations in external field [10,11,17], the diamagnetic behaviour in $\boldsymbol{B}$ field or the Lenz–Pauli quantization [11,20] in crossed $(\boldsymbol{E}, \boldsymbol{B})$ fields for one-electron Rydberg atoms.

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