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To cite this version:

S. Feneuille. INTRODUCTORY TALK ON EFFECTS OF ELECTRIC AND MAGNETIC FIELDS ON HIGH RYDBERG ATOMS. Journal de Physique Colloques, 1982, 43 (C2), pp.C2-13-C2-18. <10.1051/jphyscol:1982202>. <jpa-00221811>

HAL Id: jpa-00221811
https://hal.archives-ouvertes.fr/jpa-00221811
Submitted on 1 Jan 1982

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INTRODUCTORY TALK ON EFFECTS OF ELECTRIC AND MAGNETIC FIELDS ON HIGH RYDBERG ATOMS

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Résumé. - Durant les cinq dernières années, de nombreuses études expérimentales et théoriques ont été menées sur des atomes très excités en présence d'un champ statique extérieur, électrique ou magnétique. Cet exposé introductif a pour but de montrer, qu'en conséquence, notre connaissance de ce sujet a considérablement progressé mais que, néanmoins, de nombreuses questions restent encore ouvertes.

Abstract. - During the last five years, numerous experimental and theoretical studies have been carried out on highly excited atoms in the presence of an external static field, electric or magnetic. The aim of this introductory talk is to show that, consequently, our knowledge of the subject has considerably increased, but that, nevertheless, many questions still remain open.

1. Introduction.

The last meeting specially devoted to Rydberg atoms was held in Aussois (1), during the same month, but five years ago. Many participants of the present conference were already there and they will certainly remember that several papers were already given on highly excited atoms in the presence of an uniform static field, electric or magnetic. Therefore, one may ask the following question: was it really opportune to organize in 1982 a new meeting on the same subject and still more specialized? The aim of this introductory talk is to show that, during the last five years, our knowledge of the subject has significantly increased owing to numerous experimental and theoretical studies and that, nevertheless, many problems remain completely unsolved. Therefore, it is effectively the right time to take stock of the question and we must thank the organizers of this meeting for providing us such an opportunity.

2. Rydberg atoms in electric fields.

Five years ago, the study of the effects of electric fields on high Rydberg atoms was already very active (2). More precisely, many efforts were devoted, at that time, to the understanding of field ionization properties of highly excited atoms. In the corresponding experiments, a given atomic energy level, or sometimes a given atomic state, is selectively excited and the electric field is applied only a certain time after the excitation pulse, the number of created ions or electrons being counted as a function of the strength of the electric field. However, in that case, the corresponding results do not depend only on the atomic structure in the presence of the electric field; their interpretation requires also the introduction of dynamic or transient effects depending, in particular, on the manner of applying the field (3). In fact, it appears now that it is more informative to keep the field at a fixed value during the excitation and to count the number of ions or electrons as the function of the excitation frequency which is continuously tuned. Numerous experiments of this type have been carried out during the last five years on many atoms, mainly on alkali metals, alkaline earths and noble gas, for various field intensities and excitation energies. All of them utilize atomic beams and lasers.
Perturbation Theory for the hydrogen atom -

In 1977, our theoretical knowledge of the considered problem was essentially limited to the hydrogen atom. The corresponding description can be found in the famous book of Bethe and Salpeter (4). Of course, the separability of the Hamiltonian in parabolic coordinates and its various consequences have been recognized for a long time. The same holds true in respect of the predictions of perturbation theory to lowest orders which is valid in the low electric field limit and of the asymptotic character of perturbation expansion. However, during the last five years, many papers appeared on this perturbative approach (5-8). In particular, one is now able to calculate extremely high orders (the present record seems to be 250 (8)). Moreover, since the discovery of a dispersion relation between the state energy shift and its ionization rate (9-10), perturbation theory appears now to be an usable method in also calculating ionization rates, but the corresponding calculations remain tractable for rather small field strengths only.

Ionization properties of the hydrogen atom -

Ionization properties of the hydrogen atom in the presence of an electric field were already investigated in the book of Bethe and Salpeter (4) who start from the kinetic energy curves related to each of the two parabolic coordinates. In fact, the corresponding analysis contains implicitly a very important result which has been pointed out in 1978 only (2,11): according to the excitation energy E, and the field strength F, three situations can be distinguished: for negative energy, there is a low electric field limit (F<6.25) in which only quasi stable Stark levels exist (of course, their observation in excitation spectra requires that an intense electric field be applied for ionizing them, a certain time after the excitation pulse); in an intermediate region, quasi stable Stark levels are superimposed to ionization continua while in the third region (F<-21), only broad ionization continua can be found.

Photoionization -

Quantitatively, ionization properties of the hydrogen atom in the presence of an external electric field have been shown, before 1977, for quasi stable Stark states only. The first approaches used WKB approximations (12-13), but the first "exact" calculations appeared as far back as 1969 (14). During the last few years, one of the most important contributions to the subject was provided by exact calculations (15-17) not only of state densities (16) in the three regions described above, but also of photoionization cross sections from the ground state (15,16) or from weakly excited states (17). These calculations show that the photoionization cross section (or the excitation spectrum) is not directly related to the spectral density of the atomic states because of possible cancellation effects in the oscillator strength. They provide the first complete explanation of the equally spaced resonances observed, above the zero field ionization limit, in atomic photoionization in the presence of an electric field (18-20). However, it must be noticed that such regularities have been first predicted from one dimensional models (18, 21-23) within the framework of classical mechanics of WKB approximation.

Alkali atoms -

In fact, only one experiment has been actually performed up to now on two quasi stable Stark levels of hydrogen (5), and most of the experimental data concern alkali metals. Since 1977, many new phenomena have been observed, showing the influence of the atomic core or, of the spin-orbit interaction, which both break symmetry and destroy separability. In particular, anticrossings of Stark levels have been extensively studied mainly in the low field limit (24) but also in the intermediate region defined above (25). Very narrow resonances appearing on photoionization spectra have been interpreted in terms of autoionization of quasi stable Stark states superimposed to ionization continua (11,26,27). Strong perturbations of oscillator strengths for transitions starting either from the ground state or from
weakly excited states have been clearly demonstrated both in the low field limit (28) and in the vicinity of the field free ionization limit (17). Quantitatively, these phenomena have been approached by various methods including diagonalization of the Hamiltonian within a truncated spherical basis (24), configuration interaction and quantum defect theory in parabolic coordinates (17,29), but much remains to be done specially for computing the width of the various resonances which have been observed.

3. Rydberg atoms in magnetic fields.

In 1977, our theoretical knowledge of Rydberg atoms in the presence of an uniform magnetic field was essentially limited to the hydrogen atom although all experiments had been carried out either on alkali metals or on alkaline earths. The remark is still valid today.

Situation before 1977.

The main characteristics of the considered situation has been known for a long time (4): evolution of the various energy terms with increasing the principal quantum number, $n$; constants of motion (parity, magnetic quantum numbers); existence of a critical field $B_0 = 2.35 \times 10^5$ T; non separable character of the Hamiltonian. In particular, the determinant role of the diamagnetic interaction, called sometimes the quadratic Zeeman effect, on the magnetic structure of highly excited atoms ($B/B_0 \gg n^{-1}$) has been recognized, theoretically from the very beginning of quantum mechanics, and experimentally since the beautiful experiments of Jenkins and Segré in 1939. The very low field limit ($B/B_0 \ll n^{-1/2}$) has been well understood owing to first-order perturbation theory. In particular, because of a $z$-mixing induced by the diamagnetic contribution, it has been clear for a long time that an additional label, $k$, must be introduced to distinguish the various levels belonging to the same hydrogenic manifold.

Another important feature had been obtained by Garton and Tomkins (30) who observed, in 1969, equally spaced structures in the $\sigma$ absorption spectrum of barium in the presence of a rather weak magnetic field varying between 0 and 2.4 T. These structures, called quasi-Landau resonances extend across the zero field series limit into the continuum with a spacing of about $1.5 \hbar \omega_C$, $\omega_C$ being the cyclotron frequency. They have been explained in the beginning of the 70's, by more or less simplified Bohr models (31), and, in particular, by very simple considerations on the $n$-dependence of the various terms of the Hamiltonian (32).

New observations of quasi-Landau resonances.

During the last five years, many contributions, both experimental and theoretical have been brought to the problem. First quasi-Landau resonances have been observed in many other spectra (33-37), in particular, in strontium and in alkali metals, from lithium through cesium. In all these spectra, the quasi-Landau phenomenon occurs in $\sigma$ polarization but not in $\pi$ polarization, a result which has been predicted by Fano (1).

Quasi crossings and diabatic levels.

However, one of the most important results which have been recently obtained concerns the $k$-mixing induced by the diamagnetic interaction in the intermediate field region ($B/B_0 \approx n^{-2/2}$). In despite of this inter-$n$ mixing, it appears that anticrossings between levels with same parity and same magnetic quantum numbers become very weak for highly excited hydrogenic states (38,39). This experimental feature has two main consequences. On one hand, it could suggest the existence of an approximate dynamical symmetry and actually, some papers using $SO(4)$ symmetry recently appeared on this problem (40), but, it seems that the question still remains open, at least partially. Anyhow, it is now possible to give an interpretation of $k$ in terms of a generalized angular momentum (41).
On the other hand, one knows that one can easily follow "diabatic" levels labelled by the same quantum numbers as in the low field limit.

**Interpretation of the quasi-Landau phenomenon**

Concerning the interpretation of quasi-Landau resonances, the most significant contribution comes from two observations published two years ago (36, 37) and both showing that such resonances emerge by concentration of oscillator strength into the "diabatic" level which, in the low field limit, rises fastest in energy with increasing the magnetic field (by convention, this particular level is labelled by the minimum value of k). Moreover, these levels obey some scaling laws valid for the whole range of the magnetic field (35, 42) and simple analytical expressions for the spacing of quasi-Landau resonances, covering the entire energy range, have been recently derived (43). Anyhow, the appearance of the quasi-Landau phenomenon can be seen as a direct consequence of optical excitation. Moreover, the minimum value of k plays a major role and since the corresponding wave functions calculated in the low field limit appear to be essentially localized in a plane perpendicular to the magnetic field, one now understands why two dimensional Bohr models (31, 32) or more refined WKB approximations in two dimensions (35, 37, 44) are able to provide a correct interpretation of the observed spacing between quasi-Landau resonances.

**Concluding remarks**

Therefore, one can have the feeling that the situation of Rydberg atoms in the presence of a static magnetic field is now well understood but actually, many basic problems remain partially or totally open. For example, the ionization properties of Rydberg atoms in magnetic fields, the construction of three-dimensional solutions or the influence of the atomic core are subjects that should deserve a lot of attention in a near future.

4. Rydberg atoms in crossed fields.

One must finally notice that a few recent studies concern Rydberg atoms in crossed fields. In particular, one begins to well understand the situations in which one of two fields is much stronger than the other one.

When the magnetic field is much more intense than the electric one (Bn^3 \gg Fn^4 in a.u.), new resonances appear because of the mixing between different magnetic states induced by the electric field. This mixing leads, in the vicinity of the field free ionization limit, to the observation of quasi-Landau resonances with a spacing now equal to 0.5 \( h \omega \) (45). In the opposite case (Fn^4 \gg Bn^3 in a.u.), the observed phenomena are basically the same ones but the spacings are now essentially governed by the electric field (46).

In the intermediate case, very amazing predictions can be qualitatively derived from the potential energy surface which exhibits, in addition to the inner Coulomb valley, an outer well whose position, depth and width depend essentially of the respective strengths of the two fields. Therefore, a possibility appears of creating atoms with an electron temporarily trapped in an eccentric orbit far from the nucleus (47). Observation of such asymmetry atoms has not yet been achieved however.
References.

This list of references is not intended to be exhaustive but only representative.


