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RYDBERG ATOMS INTERACTING WITH INTENSE NON RESONANT E.M. FIELD

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Résumé. - Les déplacements de niveaux atomiques faiblement liés (États de Rydberg) dus à une perturbation par un champ électromagnétique intense, non résonnant et de faible fréquence, ont été étudiés expérimentalement. Cet effet peut être interprété par la mécanique classique de l'électron comme un accroissement de son énergie cinétique moyenne dû à ses vibrations dans le champ E.M.. La comparaison des résultats expérimentaux avec les calculs sont satisfaisants.

Abstract. - Shifts of weakly bound states of atoms (Rydberg states) due to perturbation by an intense non resonant low frequency electromagnetic field has been experimentally studied. This effect can be interpreted in terms of classical mechanics as an increase of the kinetic energy of the electron vibrating in the E.M. field. Comparison of experimental results with theoretical calculations has been found quite satisfactory.

Interaction of an atom with a strong electromagnetic field has been widely studied for a long time both theoretically and experimentally, especially since the discovery of laser. The main result of these interaction is the well known light shift which has given rise to numerous experiments and calculations particularly in the case where only few levels are involved in the interaction. In that case, the light shift may be calculated using the perturbation theory and the few oscillator strengths connecting the levels of interest. The corresponding experiments have been done on the deep levels of atoms which can be considered as sufficiently isolated.

However, during the past few years, the spectroscopic studies of Rydberg states has been extensively developed. For such studies it has been necessary to take into consideration the perturbation of these particular states by E.M. field. As an example, the effect of blackbody radiation mainly on the lifetime of the states has already been studied [1,2]. In the present work we are interested in studying perturbations due to intense non resonant laser radiation.

In any case the problem is not so simple than the above mentioned one due to the fact that the interaction involves an infinite set of levels including the continuum. In fact in a theoretical paper [3] C. Cohen-Tannoudji and his co-workers has treated a quasi-similar problem namely the interaction of a free or weakly bound electron with high frequency E.M. field. The fundamental hypothesis was that the extension ΔE of the infinite set of levels of interest is much smaller than the energy of the perturbing photons $\hbar\omega$: $\Delta E \ll \hbar\omega$. In this particular case, using an effective hamiltonian an exact quantum treatment can be done. In such a treatment, terms corresponding to effects due to vacuum fluctuations as well as those due to an external E.M. field appear simultaneously. The first order term of the interaction is $\mathcal{E}_v = \frac{e^2 E^2}{4m \omega^2}$ depending on the electron charge and mass and on the amplitude of the electric field of frequency $\frac{\omega}{2\pi}$. This term can be easily understood using classical mechanics : if one considers an

electron moving slowly in a coulomb potential (weakly bound electron) the net effect of the high frequency E.M. field is to subject the electron to rapid forced oscillations and then the non perturbed kinetic energy is increased by an amount $\mathcal{E}_v$ which is nothing but its vibrational kinetic energy ; it is , in fact , this term that we are mainly considering here, it is quite different from the term corresponding to the fact that the oscillating electron does not see the value of the potential at the mean position but rather a mean value of it, the corresponding correction is much lower than the previous one . However , contrary to the main term which affects equally each energy level this one depends on the potential gradient and thus on the characteristics of the electron trajectory : it will be different for an s state, for which the electron probe the field at the nucleus, than for a p state. This effect has a particular interpretation in the sense that the relative shift between the s and p state can be considered as a stimulated Lamb shift.

As mentioned above we were mainly concerned to give an experimental evidence of the increase $\mathcal{E}_v$ of the electron kinetic energy. The corresponding shift of its energy level is not very small and can be easily experimentally detected using high resolution spectroscopy technics : one can calculate that with a laser at 1.06μ delivering a power density of 1 MW/cm^2 it is of the order of 25 MHz. However the high frequency condition : $\hbar\omega \gg \Delta E$ is rather difficult to achieve in atomic systems. An ideal condition would be fulfilled with atoms in a high value of the quantum number ℓ such as $\Delta E = \frac{R}{\ell^2} \ll \hbar\omega$; but, in this case, the analysis of the perturbed state from the ground ℓ^2 state would require multiphoton excitation and thus much higher laser power on a single mode at relatively low frequency. The experiment that we have attempted was done on atomic rubidium perturbed by the 1.06μ radiation of a NdYag laser. The studied Rydberg level was a p one directly excited and analysed from the ground state using UV radiation. On the level scheme shown Figure (1) one can see that only few levels to not fulfil the high frequency condition and that mainly two phenomena may perturb the observation of the level shift $\mathcal{E}_v$ through a change in the UV transition : first of all the 1.06μ radiation induces a shift of the ground state due to the resonance 5p level this shift has an opposite sign compared to the shift $\mathcal{E}_v$. Unfortunately it cannot be measured experimentally by studying the shift of

the frequency of the resonance transition ; the shift of the 5p level being itself strongly dependent of other levels such as the 6s and 4d . The shift of the ground state has therefore to be evaluate theoretically.

Figure 1

Shifts of the atomic energy levels of rubidium under 1.06μ excitation.

On the other hand the shift of the Rydberg level integrates the shift we are dealing with to eventual other shifts coming from nearly resonant coupling. In fact only few valence levels may have some significant role in the presence of the $1.06\ \mu$ radiation. They are : the $6s$, $7s$, $4d$ and $5d$, which all are quite far from resonance. Nevertheless we have to give an estimate of their influence.

The experiment has been performed on an atomic beam of rubidium atoms especially mounted for high resolution experiments [4]. As shown in Figure 2 the Rb atoms cross an interaction region in which they experience two laser fields : an UV one which can be tuned to the frequency of the atomic transition between the ground state $5s$ and the Rydberg state, and an intense non resonant perturbing laser field at $1.06\ \mu$. The highly excited atoms are detected by means of electric field ionization. The interaction region is actually located between capacitor plates on which a voltage pulse is applied immediately after the laser excitation. For a high enough voltage, ionization occurs and the ions are deflected towards an electron multiplier and counted.

Figure 2

Experimental set-up.

In order to overcome all synchronization difficulties the two laser fields are both generated by the same pump. The UV source at $\sim 2950\ \text{\AA}$ is built from a single mode tunable pulsed dye laser using a doubling crystal (A.D.A.). It has been described in details previously [5] ; it combines in some way the properties of the CW laser and those of a pulsed one excited by a NdYag laser doubled in frequency (530 nm) . Its main characteristics lie in that it delivers single mode light pulses of 20 ns duration with a repetition rate of 10 Hz and a spectral linewidth of about 50 MHz (at 295 μm). The intense IR source is provided by the residual infrared output energy at 1.06 μm available at the output of the NdYag laser : the light pulse is sent into a double pass NdYag amplifier in which the use of a Glan prism together with a quater wave plate permits to extract all the energy and to decouple the amplifier from the oscillator. Such an arrangement has made it possible to deal with available IR light pulses of 140 ns duration and peak power ranging between 0 and 12 MW/cm^2 . Both light beams (the UV one and the IR one) propagate collinearly but in opposite directions. Special care has been taken in order to make sure that the UV beam actually probed the centre of the infrared one. On the other hand a synchronous photograph of the two pulses displayed on a scope

screen insured us that the UV pulses was probing the highest energy part of the IR pulses. As the frequency shift measurements are depending on the power density of the IR beam and, in addition, as they have to be done in an absolute way, the previously described adjustment as well as the determination of the power density in the probed region are certainly the most delicate parts of the experiment. Actually the laser power has been measured using a power meter (Scientech) outside the atomic beam apparatus and correction has been brought to take into account the absorption of the windows. The peak power has been deduced using the pulse duration measured on the scope screen, whereas the diameter of the laser spot has been measured using a detector mounted on a precision translator. It has been found to be .8 mm at half energy points.

The measurements have been done on the transition $5s\ ^2S_{1/2} - 22p\ ^2P_{3/2}$ of the most abundant isotope of Rb (which is the ^{85}Rb in the natural mixture). Each recorded spectrum shows a doublet structure which corresponds to the hyperfine splitting of the $5s$ ground state. Its value has been measured precisely long time ago and found to be 3035 MHz ; it allows us to calibrate the frequency scale in each recorded spectrum. Figure 3 shows a typical recording of the hyperfine structure of the line $5s \leftrightarrow 22p$ in the absence ($P_{\text{IR}}=0$) or in the presence of the infrared power. All resonances have been recorded during the same frequency scan which makes it easy to have an absolute measurement of the frequency shift. As we can see the spectral resolution is less than 60 MHz in the UV range, which is much lower than the shift. We may observe that in the presence of the infrared power the resonances are slightly broadened : their widths are about 120 MHz large. This broadening has been interpreted as due to both residual inhomogeneity of the probed part of the infrared beam and to fluctuations of the infrared peak power. In any case all these effects give a relative broadening with respect to the frequency shift of the order of 10 %.

On the Figure 4 we have plotted the result of several measurements of the frequency shift of the transition for various infrared peak power densities. As it was expected the experimental points lay on a straight line. Which passes through the origin and has a slope equal to 61 MHz/MWcm^{-2} .

Figure 3 : Typical recording obtained by switching on and off the IR power during the same frequency scan.

As it has been mentioned above, this frequency shift is in fact the sum of two terms, the shift of the ground state Δv_g and the shift of the 22p level Δv_e which integrates the shift we are dealing with to eventual perturbations coming from nearly resonant coupling. In order to compare the experimental results with the theory we have calculated all of these shifts. This has been done using appropriate wavefunctions which describe the energy spectrum of Rb atom [6]. All these data are reported in Table I.

Level		Frequency shift in MHz/MWcm ⁻²
22p	ξ_v	25
	Δv_{5d}	$3 \cdot 10^{-3}$
5s		-26.3
5p		107

Table I

As we can see the contribution of the 5d state to the shift of the 22p is completely negligible and thus a fortiori those of the 6s, 7s and 6d states. On the other hand the shift of the 5s ground state is of the same order of magnitude than the expected shift; we have also indicated the calculated shift of the 5p state, one can remark that it is completely different from the 5s one because it is strongly perturbed by the 5d and 6s states. For these reason the shift of the 5s state cannot be simply measured by studying the transition $5s \leftrightarrow 5p$.

The dotted line on Figure 4 corresponds to what one can calculate using the data of Table I. Comparison between experimental and theoretical results may be qualified as satisfactory.

Figure 4 : Experimental measurements of the frequency shift of the UV transition. Comparison with calculated shift (dashed line).

In conclusion we have clearly demonstrated that in the presence of a strong non resonant E.M. field, Rydberg states are shifted and that this shift may be seen as resulting from the stimulated radiative corrections induced by the field on a free or weakly bound electron.

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