2. Symposium on atomic and surface physics.
Hintermoos, Austria, Febuary 10 - 16, 1980.
CEA - CONF 5075

MULTIPHOTON PROCESSES

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Abstract :

The main features of multiphoton processes are described on a somewhat elementary basis.The emphasis is put on multiphoton ionization of atoms wnere the influence of resonance effects is given through typical examples. The important role played by the coherence of light is shown to produce a very dramatic influence on multiphoton absorption. Different observations concerning molecules, electrons, as well as solid surfaces illustrate the generality of these very non linear interaction between light and matter.

The existence of such processes have been foreseen for quite a long time, since 1931, when M. Göppert-Mayer presented a very interesting calculation concerning 2 photon absorption. It is only 23 years later, that the first experimental observation was made by Kusch in 1954 followed by very refined measurements performed at the Ecole Normale Supérieure by J.M. Winter. Multiphoton transitions in the radiofrequency range were observed between Zeeman sublevels of sodium.Since the event of lasers, this subject has gained a considerable importance.

We will concentrate on certain very fundamental aspects of multiphoton processes. An atom or a molecule immersed in a stream of photons will be able to absorbe "simultaneously"N photons if their density is sufficiently large. This interaction may be considered as a (N+1) body interaction involving 1 particule + N photons. The absorption is a very non linear process which is proportional to aI^N (I being

the photon flux). The coefficient a is related to the nature of the particle and to the characteristics of the light.

If the energy of the photon does not coïncide with an energy level of an atom the absorption is associated to a virtual level. In such a non resonant condition the time τ_{NR} during which the photon is kept by the atom is $\tau_{NR} = \frac{1}{\Delta E}$, where ΔE is the energy difference between the virtual level and the closest real level. τ_{NR} is currently in the range of 10^{-13} to 10^{-15} sec. Cumulative processes are allowed to build up only if other photons are absorbed in time shorter than τ_{NR} i.e. before the atom has lost his first photon, and this will only happen if I is large enough.

During the cumulative absorption of photons, if one of the virtual levels coïncide with a real level of the atom, a resonance process shows up, which have very remarkable features. The essential reason is that the corresponding critical time is nothing but the life time of the level which is usually in the 10^{-7}, 10^{-8} sec. range, considerably larger than τ_{NR}. The consequence is obvious, these resonance effects will be obtained with lower I, their probability will be higher.

If the total energy of the photons absorbed is higher than the ionization potential, the atom is ionized, we speak of Multiphoton ionization which has been a very popular field of research.

Many experiments have been performed with monomode tunable Nd- glass pulsed laser forwhich very remarkable performances have been obtained (Photon energy is 1,17 eV duration of pulse 30 nsec.)With such a laser, four-photon ionisation of cesium atoms requires a laser flux of 10^{8} w/cm^2; 22 photon ionisation of helium - 10^{15} w/cm^2.

One experimental result has received a great deal of theoretical studies, it is the four-photon ionisation of cesium atoms, via

the resonant three-photon transition 6s→6f. When the laser frequency is tuned continuously in the neighbourhood of the resonant three-photon transition 6s→6f, four resonance peaks are observed due to the hyperfine structure of the groundstate 6s of the cesium and the fine structure of the 6f doublet. The fine structure of the 6f level is no longer observed when the laser intensity is increased because of an intensity-dependent broadening of the resonance profile.

Furthermore, the resonance profile is shifted as the laser intensity increases. This leads us to consider the effect of the laser radiation on atomic levels. The exchange of photons between the laser radiation and atoms shifts and broadens atomic levels. These effects have been well described within the frame of the dressed atom theory (Cohen-Tannoudji 1967).

The experimental resonance curves shift with increasing values of the laser intensity as predicted by the theoretical model. The resonance shift ΔE is linear with respect to the laser intensity I, $\Delta E = \alpha I$, with $\alpha = 2\ cm^{-1}/GWcm^{-2}$. The linearity of the resonance shift as a function of the laser intensity, implies that one photon is absorbed by the atom and reemitted, in addition to the four-photon absorption leading to the ionisation of the atom.

One of the most important features of resonance effects is the dramatic change of the order of non-linearity $K = d\ Log\ N_i/d Log\ I$ as a function of the resonance detuning δ, where N_i is the number of ions. In the vicinity of a resonance, the observed order of non-linearity K no longer corresponds to the number of photons absorbed by the atom. However, when the resonance detuning is large enough ($\delta = 30cm^{-1}$), the value K=4 is observed again, corresponding to a non-resonant four-photon ionisation of cesium atoms. The agreement is excellent between experimental

results and the theoretical curve. It is a very sensitive test of the theoretical model.

Resonances are severely damped by the increase of of the coupling between resonant state and the continuum, for increasing values of I. In such conditions the life time of a resonant level is considerably shortened and falls in the usual range of non resonant processes. Consequences of this effect has been observed in the 12 photon ionisation of krypton for $I = 10^{13}$ w cm^{-2}. At such high fields, resonance peaks are so enlarged, that it is only the order of non-linearity which reveals the existence of these highly damped resonant processes.

In investigating resonance effects, only coherent light from a monochromatic laser radiation has been considered. However, incoherent light with statistical properties closely related to thermal light is ordinarily delivered from high power Nd-glass or dye lasers if no additional steps are taken to obtain a single frequency operation. When experiments are performed with such incoherent lights, laser temporal coherence effects are one of the most dramatic effects which have been encountered in multi-photon ionisation processes. These coherence effects are explained in terms of bunching effects. When the laser light is incoherent, photons arrive in bunches during the pulse duration, while the photons arrive uniformly in sequence when the laser operates in a single mode and gives coherent light.

This bunching effect, as will be demonstrated on a simple example, increases the probability of absorption of photons. The reason is simply related to the fact that bunching increases the density of photons, or reduces their time interval in the vicinity of atoms allowing cumulative effects within the characteristical time τ.

Coherence effects were experimentally investigated by measuring the non-resonant 11-photon ionisation of xenon by varying the mode structure of a 40 ns Nd-glass laser pulse from a single-mode to one hundred modes, that is changing the coherence time from 10^{-8}s (no bunching effect) to 10^{-11}s (important bunching effects). For a given average laser intensity, an enhancement of nearly 10^7 is observed in the number of ions when bunching effects are maximum. This enhancement factor is very close to the calculated value N! that is 4×10^7 for N = 11.

All these results strongly emphasize the considerable importance of time. This aspect has suggested to explore directly the influence of the duration of the laser pulse on multiphoton processes. Experiments have been performed with picosecond pulses in the case of 4 photon ionization of cesium. In these very stringent conditions a different behaviour was observed mainly related to the fact that the duration of interaction is smaller than the life time of the 6f resonant state reduced by its coupling with the continuum.

All the experimental contributions described so far correspond to a situation where the equivalent electric field of the light is smaller than the internal electric field of atoms. Quite recently the inverse situation has been produced, multiphoton ionization of He corresponding to 22 photons has been observed at 10^{15} w/cm^2 (E field - 10^9 V/cm).

Many other very interesting observations have been devoted to molecules, solid surfaces, electrons, some examples of the most exciting results will be given during the talk.