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I.C.T.P., P.O. BOX 586, 34100 TRIESTE, ITALY, CABLE CENTRATOM TRIESTE

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COHERENT TRAPPING IN LASER SPECTROSCOPY

E. Arimondo

Università di Pisa
Dipartimento di Fisica
Pisa 56100
Italy

Three-level spectroscopy and coherent trapping

E. ARIMONDO (*)

Abstract:

The coherent trapping phenomenon in discrete and continuum states is briefly described and the experimental observations are discussed.

1. - Introduction

The spectroscopic investigations of atoms and molecules are based on the interaction of near-resonant radiation with the species under investigation. The use of monochromatic, intense and continuously tunable radiation sources allows to reach the maximum sensitivity and accuracy in the determination of the atomic or molecular levels. The simultaneous application of two (or more) near-resonant monochromatic radiation fields gives rise to the multiphoton absorptions and other nonlinear phenomena, whose application has been explored in the continuously expanding field of the nonlinear spectroscopy. The three-level system interacting with two monochromatic radiation fields represents a configuration where the nonlinear phenomena are greatly enhanced. The double resonance experiments, based on the modification of the three level populations by the two resonant field, have played a major role in the high resolution spectroscopy. Double resonance was started in 1949 by Brossel, Kastler and Bitter when light from a discharge lamp and a radiofrequency source were applied simultaneously to mercury atoms to investigate the atomic structure in the excited state [1]. In 1959 two-photon transitions in a three-level molecular system were observed for the first time using two microwave sources by Gozzini an coworkers [2]. The development of monochromatic and tunable dye lasers has produced an explosion of high resolution three-level investigations.

The application of two continuous wave radiation fields to a three-level system leads to a creation of a coherent superposition of states, which may be stable against the radiation field absorption, the spontaneous emission decay or

(*) Dipartimento di Fisica dell'Università di Pisa, 56100 Pisa, Italia

the molecular dissociation. This phenomenon has been designated as population trapping, to indicate the stability of the population, or coherent trapping,* to denote the presence of a coherent superposition of the state. The population or coherent trapping may be interpreted also as a pumping of the system in a non absorbing state. This phenomenon has been observed for the first time as a decrease in the fluorescent emission in an optical pumping experiment on sodium atoms performed by Gozzini and coworkers [3]. A theoretical analysis [4] showed that the fluorescent light decrease was originated by the production of a coherent superposition of the sodium ground state hyperfine levels. At the same time and independently the research group of Stoud and coworkers at the Rochester University Investigate theoretically and experimentally the pumping and trapping originated by two laser fields resonant with two transitions on sodium atoms [5, 6].

Later the coherent trapping phenomenon has been extendend theoretically to other atomic or molecular configurations, and the phenomenon has been exploited on very different applications, as high-resolution spectroscopy, metrology or optical bistability.

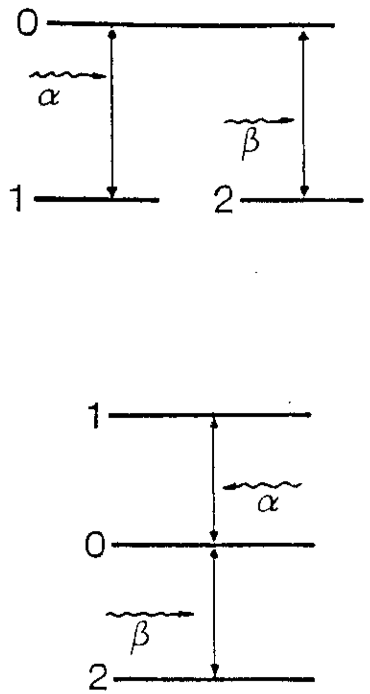
It may be noticed that the population trapping represents only an example of the richness of phenomena that may take place in the interaction of radiation fields with absorbing system. If for a long time the attention of spectroscopists was restricted to the two-level systems, the possibility of irradiating samples by several electromagnetic fields at the same time has shown that new features may be obtained in configurations with a larger number of the levels and several radiation fields. Infact the coherent trapping may be related to the SU(3) group symmetries of the Hamiltonian and to some conservation laws to be satisfied by the density matrix elements of the system under investigation during their time evolution. Conservation laws related to the observables in experiments involving a multilevel system interacting with several radiation fields have been infact examined recently by several authors [7]; however no new phenomena so remarkable as the coherent trapping have so far been identified. Coherent trapping is an interference phenomenon and as such it is closely related to other interference processes well exploited in spectroscopy, as the Fano autoionization profile, the level crossing or the Honie effect. The decay of the K mesons is another well known example of the important role played by the interference effects. However the reduction of the trapping to an interference feature is a reductive description, and in effect the experimental observations are strongly based on the role of the optical pumping as a preparation into a state suffering the interference in the absorption or radiative decay. It is well known that the optical pumping, in all its variations, represents an important method for sensitive and accurate spectroscopy.

The coherent trapping will be here briefly described, and the experiments reporting detection or exploitation of the trapping will be discussed. It turns out that the coherent superposition of states arises in quite different configurations of atomic or molecular states. Thus it is timely to discuss the features of the previous experiments in order to prepare the ground for the future ones.

2. - Three-level system with discrete states

2.1. - *Theoretical analysis*

The three-level lambda and ladder configurations under consideration are shown in Fig. 1 with the common level $|2>$ coupled to levels $|1>$ and $|3>$ by the electric dipole moments μ_{12} and μ_{23} respectively. Let denote



1. - Three-level systems in the lambda (a) and cascade (b) configurations.

the j level energy by E_j. The configurations of Fig. 1 may be treated by a single formalism if a proper definition of the energies and coupling constants is introduced. Two travelling-wave electromagnetic fields $E_1(z,t)$ and $E_2(z,t)$ at angular frequencies ω_1 and ω_2 and wavenumbers k_1 and k_2 drive the $|1> - |2>$ and $|2> - |3>$ transitions by a proper choice of the polarization vectors or the frequency detunings. The detunings for absorbers moving with velocity v along the z axis are given by:

$$\Delta_1 = (E_2 - E_1)/h - \omega_1 - k_1 v \quad (1a)$$

$$\Delta_2 = (E_3 - E_2)/h - \omega_2 - k_2 v \quad (1b)$$

Within the standard RWA the Rabi frequencies are given by:

$$\alpha = (2h)^{-1}\mu_{12}E_1; \quad \beta = (2h)^{-1}\mu_{23}E_2$$

The relaxation processes for the diagonal elements of the ensemble averaged density matrix $\rho(v,z,t)$ are accounted by the damping constants $\gamma_j (j = 1,2,3)$ composed by the decay rate to levels not included in the three-level system and the spontaneous decay or collisional transfer between the three levels. The decay rate of the out-diagonal density matrix elements, i.e. the coherences, are described by the above relaxation rates and additional rates describing the influence of finite bandwidth lasers and phase-interrupting collisions. Source rates are introduced into the equations to decribe the equilibrium population densities ρ^0_{jj} in absence of the applied fields.

The time evolution of the density matrix elements is given by

$$(\delta/\delta t + v\delta/\delta z)\rho_{ij} = (ih)^{-1}[H,p]_{ij} + \delta/\delta t\rho_{ij}| \text{ relax}$$

where the Hamiltonian H takes into account the energy separations and the coupling with the electric laser fields, and the last derivative takes into account the relaxation processes and the equilibrium populations.

The steady state solution of the density matrix equations for a three-level system, presented by several authors [4,5,6,8], cannot be expressed in a simple analytical expression and it will not be reported here. In effect a numerical representation of the density matrix elements is the best way to show the coherent trapping phenomenon.

Narrow features appeares on the response of the three-level system under investigation when the two-photon condition is satisfied, i.e. the Raman-type two-photon process in the lambda configuration and the two-photon process in the ladder configuration:

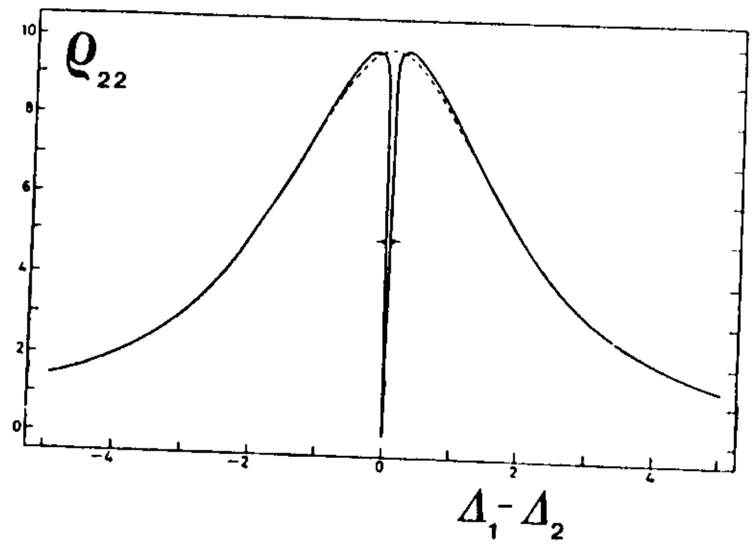
(2) $$\Delta_1 - \Delta_2 = 0$$

where the upper (lower) sign refers to the ladder (lambda) configuration. As it can be seen in Fig. 2, when the two laser fields approach to resonance on the optical transitions, i.e. $\Delta_1 = \Delta_2 = 0$, the $|2>$ level population grows following the excitation lines, and a maximum value of ρ_{22} would be expected when both laser are exactly in resonance. On the contrary a strong decrease occurs in a narrow region around the condition of Eq. (2), in which the atoms remain distributed uniformly over the $|1>$ and $|3>$ lavels, i.e. the population is trapped in those states.

For instance in the lambda configuration at the resonance conditions for the optical transitions it turns out that the state:

(3) $$|r> = \alpha|1> - \beta|3>$$

is not coupled to the excited state by the radiation fields, whence it does not absorb the radiation. Owing to the pumping created by the radiation fields and the relaxation processes, the system is finally prepared into the state $|r>$ and no absorption of radiation takes place. The state $|r>$ presents a coherence ρ_{13} between the states $|1>$ and $|3>$ so that this process been called a trapping originated by the coherence.



2. - Example of excited state population p_{22} as function of the detuning $\Delta_1 - \Delta_2$ with the tipical central dip of the coherent trapping.

The amount of coherence trapping and the dip in the ρ_{22} population presented in Fig. 2 depends on the Rabi coupling constants and the relaxation rates. The widht δ of the dip, in the low field limit, is given by:

(4) $$\delta = (\alpha^2 + \beta^2)/2(\gamma_{12} + \gamma_{13})$$

with γ_{12} and γ_{13} the relaxation rates for the optical coherences. Whence that width may be very small at reasonable values of the Rabi frequencies. The coherence trapping has been investigated in the references [4,5,6,8] for different values of relaxation and coupling rates. In a analysis of the molecular excitation and dissociation by Stettler et al [9] the coherent trapping was derived within the framework of the occupation amplitudes for the molecular states. Radmore and Knight [10] and Swain [11] have made use of a dressed state approach to investigate the influence of different parameters on the coherent trapping.

It is important to notice that coherent trapping may occur also in a cascade configuration as that shown schematically in Fig. 1b with lasers resonant on the $|1> - - |2>$ and $|2> - - |3>$ transitions. As a result of trapping when the resonance conditions for both transitions are satisfied a superposition of states $|1>$ and $|3>$ is formed and the intermediate state has a minimum in population occupation. In order to have same resonance condition for all the absorbing atoms, in the case of a Doppler broadened transition, counterpropagating laser beams should be applied to the sample under investigation. This configuration for the cascade level scheme coincides with that required for obtaining two photon transitions without Doppler effect.

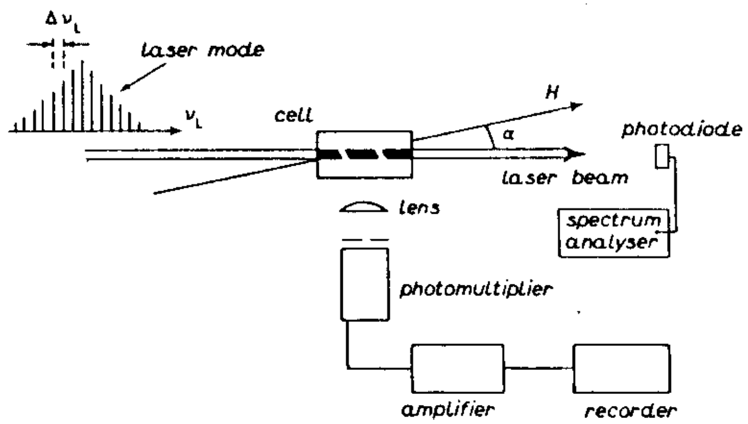
The analysis presented so far is based on two resonant monochromatic radiation fields applied on the two transitions. The two fields are coherent and the creation of a coherence in the atomic system arises from the existence of a spatial and temporal coherence in the radiation fields. A question, very relevant for the experimental detection of the coherent trapping, is that all lasers posses a finite bandwidth, whence some fluctuations, and that if two lasers are used in an experiment , their fluctuations may be independent. On the other hand if the two field are different modes of the some laser, or if the second field is produced from the first field by a splitting or frequency changing, it is possible that their fluctuations are correlated, i.e. a cross correlation of their fluctuations exist. In such a case the two photon coherences are unaffected by the laser fluctuations and the coherent trapping minimum should persist. Thus it is important to know the requirement on the cross-correlations of the two fields for the creation of a coherent trapping. This question has been investigated theoretically by few authors [12, 13]. The main question in the theoretical analyses has been the description of not completely coherent laser fields, and the creation of coherent trapping depends on the type of fluctuations introduced into the laser fields. Dalton and Knight [12] have applied a phase-diffusion model for the laser field in which the laser spectrum is considered to be lorentzian, and a cross-correlation is introduced in the relative phase diffusions of the two lasers. The influence of those fluctuations on the coherent trapping minimum was tested and of course a destruction of the coherent trapping was produced by uncorrelated phase fluctuations of the two beams, with a recover of the coherent trapping when the fluctuations are correlated. Kennedy and Swain [13] have applied a similar model to the study of the ladder configuration. It may be questioned if the phase-diffusion model is the appropriate one to describe the laser beams used in the coherent trapping experiments, so that theoretical analyses based on different laser models and experimental tests are required. In effect in ref. 14 it has been shown that in order to produce coherent trapping, particular separate conditions exist for two laser beams having simultaneous phase and amplitude variations.

An analyis of the coherent trapping on the basis of the dressed atom approach

and in the framework of the statistical properties of atoms excited by several laser fields is presented in this volume [15].

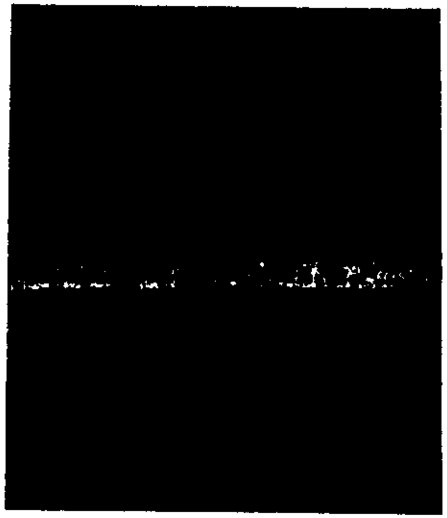
2.2. - *Experimental observations*

The first experimetnal observation by Gozzini and coworkers [3] took place, really only by a chance, in an optical pumping experiment performed on sodium atoms with radiation from a multimode dye laser tuned to the resonant transition between the $3^2S_{1/2}$ ground state and the $3^2P_{1/2}$ excited state. The frequency separation between the laser modes was around 350 MHz, so that the frequency separation between 5 adjacent modes matched the hyperfime separation of the sodium ground state. Thus the three level Λ configuration of Fig. 1 was realized with levels $|1>$ and $|3>$ corresponding to the two hyperfine levels. The experiment was performed in presence of a magnetic field H_0, whose sweep allowed a precise matching of the laser frequencies to the level separations (see Fig. 3). The experimental set-up presented a nice



3. - Sketch of the experimental apparatus used in ref 16 (reproduced with permission of the authors).

trick to make clearly visible the optical pumping phenomena: a magnetic field gradient was introduced to change the splitting of the Zeeman sublevels in the hyperfine states along the laser beam. In this way the coherent trapping, i.e. a decrease in the excited state population, appeared as a black line along the fluorescent path of the laser beam across the sodium cell. Fig. 4 from ref. [16] reports an experimental record of the decrease in the fluorescent light at the magnetic field position of the frequency matching for coherent trapping. The investigation of the phenomenon was later completed [17] through an investigation of the width of the coherent trapping feature as a function of the relaxation rate of the sodium atoms. Very narrow features were obtained if the laser beam diameter was expanded or a buffer gas was added to the cell containing the sodium atoms, i.e. if the interaction time of the sodium atoms with the radiation fields is increased. In effect the question of the width of the coherent trapping feature has been never investigated in detail in most experiments.



4. - Fluorescence of sodium vapour irradiated by a multimode laser tuned to the D_1 line with a minimum in the fluorescence produced at the magnetic field resonance condition for the coherent trapping (reproduce from ref. 16 with permission of the authors).

Accurate experiments on sodium atoms were performed at the same time of the work in Pisa by the group at Rochester [6] making use of two single mode dye lasers. Thus the decrease in the population of the upper level in the coherent trapping conditiond was observed as a function of one laser tuning. The sodium coherent trapping features were observed by the Spectroscopy group at MIT [18], and later the ultra-narrow features were considered of particular interest for the frequency standard [19]. In this context coherent trapping experiments on the hyperfine states of sodium ground state were performed on an atomic beam and an excellent cross-correlation between the two laser beams was realized by using an acousto-optic modulator to generate a second laser beam from the first one. Linewidths small as 1.3 kHz were realized.

Evidence of the trapping in the cascade configuration of Fig. 1 has been provided by an experiment on a fast beam of metastable [20] Ne atoms interacting with two counterpropagating laser fields and monitoring the

population in the intermediate level by observing the fluorescent emission [20]. At large laser intensities a dip has been observed in the population of the intermediate state; such a dip arises from the coherent trapping of atoms in the external levels of the cascade configuration, with the result that the intermediate level aquires an anomalously small population. No other experiments on the cascade configuration have been reported.

It is now appropriate to list the experimental observations where the coherent trapping phenomena resulted as a sort of nuisance and had to be avoided. On the basis of a rate equation analysis, whence neglecting the coherent interaction phenomena, it has been proposed that using two laser diodes tuned to the resonance lines from the cesium hyperfine ground states to the $P_{3/2}$ state, the optical pumping of the cesium atoms in a specific Zeeman level of the ground state should increase considerably [21]. On the contrary experimental observations based on such a pumping scheme pointed out that a large coherent trapping was present and did not allow to reach the predicted high efficiency of optical pumping. The theoretical analysis of ref. [22] has considered the possibility of avoiding the coherent trapping making use of broadband and uncorrelated laser lights. In fact it would be interesting to perform experiments where the cross-correlation between the two laser beams is modified in a controlled way and the modification of the coherent trapping is monitored.

Coherent trapping features have been also detected in laser cooled barium ions in a rf trap, when two radiation field were simultaneously applied to the ions in order to avoid the loss of the ions into the metastable $5D$ state [23]. The dip in the fluorescence from the trapped ions produced by the coherent trapping was to be avoided, because when the fluorescent stopped so did also the cooling.

The coherent trapping may be interpreted as a change in the saturation properties of an absorbing system because of the presence of the coherence created by the light between states $|1>$ and $|3>$. In effect an early analysis by Feld et al. [24] considered the problem of saturation in three-level Λ systems in presence of a low frequency coherence. At that time several mode crossing experiments, i.e. the simultaneous saturation of two transitions in a three level system, were performed for the determination of atomic or molecular structures. For instance in an experiment by Takagi et al [25], sodium atoms were irradiated with D_1 light containing a central mode and optical sidebands, and an increase in the transparency of the sodium vapour was observed when the frequency separation of the optical sidebands matched the Zeeman splitting in the ground state. However in such an experiment, as well in others performed in molecular systems, the signal was analysed on the basis of populations only and the creation of a coherence in the irradiated system was not considered. Thus an unnoticed coherent trapping in ground state may have contributed to the signal in the experiment by Takagi et al.

The theoretical analysis by Feld et al [24] considered the change in the saturation of an optical transition occuring when a state is splitted into its Zeeman components by the application of a magnetic field. When the Zeeman components are not splitted, a laser radiation field is simultaneously in resonance with all the components and a three-level configuration as that of Fig. 1 may be realized with two levels having the same energy and excited by the same laser radiation to a single excited state. In order to realize such a configuration some selection rules have to be satisfied, as for instance in the scheme of Fig. 5 from a $J = 1$ state to a $J = 0$ state with the $\Delta m = 1$ and $\Delta m = -1$

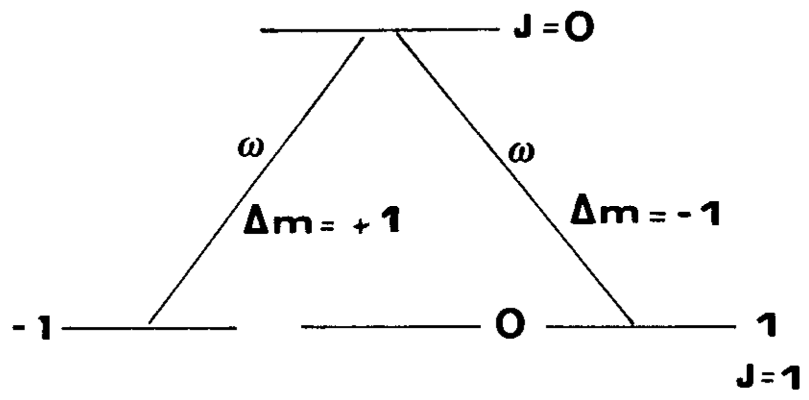
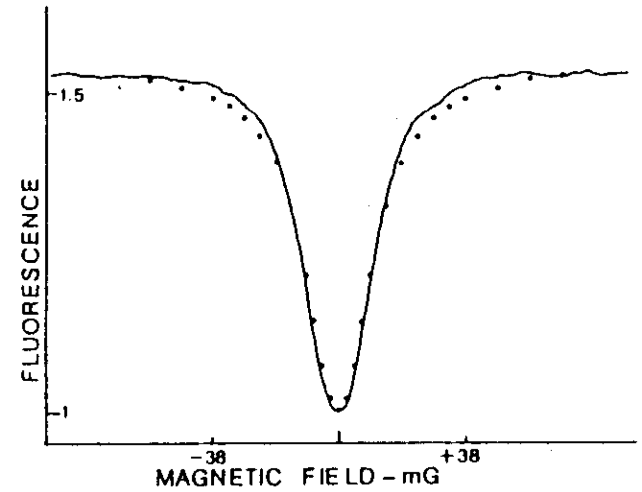


Fig. 5. - Level schemes for the transitions between $J = 1$ and $J = 0$ states irradiated by laser light on the $\Delta m = 1$ and $\Delta m = -1$ transitions. By changing the Zeeman splitting in the lower state by an external magnetic field, a coherent trapping may be tuned in or out of resonance.

transitions excited by the laser. A coherent trapping may be realized in that configuration and the trapping is destroyed by the application a magnetic field that separates the Zeeman components and bring out of resonance the laser radiation. The phenomenon has been called nonlinear Hanle effect because it resembles the ordinary Hanle effect with the monitor of the sample properties as a function of an applied magnetic field. The analysis of ref. 24 does not mention the coherent properties, but the overall saturation phenomena is a nice example of the coherent trapping. The narrow nonlinear Hanle effect resonances were observed at first in an atomic beam optical pumpping experiment by Schieder and Walther [26]. Observations of the phenomenon were reported in refs. 24 and 18, and careful investigations of the fluorescent light emitted in a $J = 0 - -J = 1$ transition of neon [27] and $J = 1, 2 - -J = 1$ transitions of a calcium beam [28] have been performed. Experiments results on the calcium beam are reproduced in Fig. 6. The nonlinear Hanle effect has been examined also with the optogalvanic detection in discharges of neon and calcium atoms [29].

It is important to notice that the coherent nonlinear response produced by an absorber in presence of a Zeeman tuned three-level structur has been used in several configurations, for instance optical bistability with the absorbing medium located inside a Fabry-Perot cavity and the cavity transmission monitored. In a sodium experiment based on the nonlinear Hanle effect, a magnetic field was applied to sodium atoms, whose nonlinear coherent response near the splitting of the Zeeman levels by the magnetic field originated the bistable response [30]. In another experiment on sodium a coherent trapping mechanism was produced by the two counterpropagating beams inside the laser cavity, detuned in resonance with the hyperfine transitions by the Doppler shifts: by the laser frequency tuning the coherent trapping was enhanced and the optical bistability produced [31]. In an experiment on samarium atoms a magnetically induced polarization-switching was produced by the Zeeman tuned coherences in the lower state [32].



6. - Typical dip in the atomic fluorescence produced by the coherent trapping resonance on the ground state of calcium atoms as a function of the externally applied magnetic field (reproduced from ref. 28 with permission of the authors).

The Zeeman coherence between the ground state degenerate levels has been used as the dominant nonlinear mechanism for the four-wave mixing [33, 34].

Phenomena closely related to the coherent trapping occur if pulsed laser are used to excite three level V or Λ configurations with a pulse laser frequency matched to the frequency separations in the $|1>$ and $|3>$ levels. In effect a Fourier analysis of the pulsed beam will present the incident radiation as equivalent to a multimode laser with frequency separation matched to the proper one for the creation of the coherent trapping. Experiments with pulse trains from mode-locked dye lasers on the Zeeman structure of sodium atoms have been performed by Lange and coworkers [35].

3. - Atomic and molecular continua

The phenomena presented in the previous paragraph for bound states have been extended to the case of states in the continuum. The coherent trapping features appear for a level scheme involving either a nonstructured continuum or an energy region where the continuum is structured, as for instance in presence of autoionization or predissociation resonances in a molecule. These preocesses are schematized in Figs. 7. Fig. 7a concerns the excitation by two lasers at frequencies ω_1 and ω_2 from the discrete states $|1>$ and $|3>$ to the continuum. Fig. 7b describes an autoionizing or dissociating state $|2>$, coupled to the continuum $\{|E>\}$ and excited by laser photons ω_1 and ω_2 starting from the $|1>$ and $|3>$ levels.

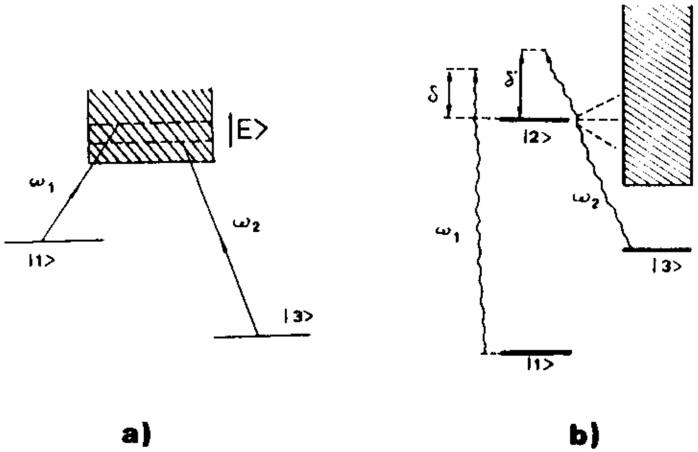


Fig. 7. - Three-level structures involving discrete and continuum states investigated for the coherent trapping. In a) two transitions from discrete states are coupled to the continuum by the laser radiations. In b) the upper states $|2>$ is coupled to the continuum by a predissociation or autoionization mechanism.

The first scheme analyzed in detail by Coleman et al [36], brings the so called induced continuum structures, which originated by the coupling between the continuum and the dressed stated corresponding to the absorption of one laser photon from either $|1>$ or $|3>$ states. Whenever a discrete state and a continuum are in resonance, a proper diagonalization of the Hamiltonian has to be introduced, and the well-know Fano profile is obtained. Also in the case of coupling between the dressed states and the continuum, a Fano lineshape is obtained. The ionization probability $P_I(t)$, i.e. the probability for being in the continuum $\{|E>\}$ is a quantity relevant in the experiments where electrons or ions are measured. The ionization probability in the scheme of Fig. 7a is

equivalent to the population in the excited state of the Λ configuration for the discrete states investigated in the previous paragraph. Thus the ionization probability may show up coherent trapping, and in effect the Fig. 8 reports the results of a theoretical analysis performed by Coleman et al showing that after the application of the laser radiations at time $t = 0$, only half of the population in the initial state $|1>$ may be ionized in the condition of coherent trapping for the two-photon Raman resonance between states $|1>$ and $|3>$. The relaxation rates introduced in the analysis are reported in the Figure caption. The ionization probability $P_I(t)$ presents a Fano asymmetric profile as a function of the laser ω_1 and ω_2 frequencies around the two-photon Ramam resonance condition.

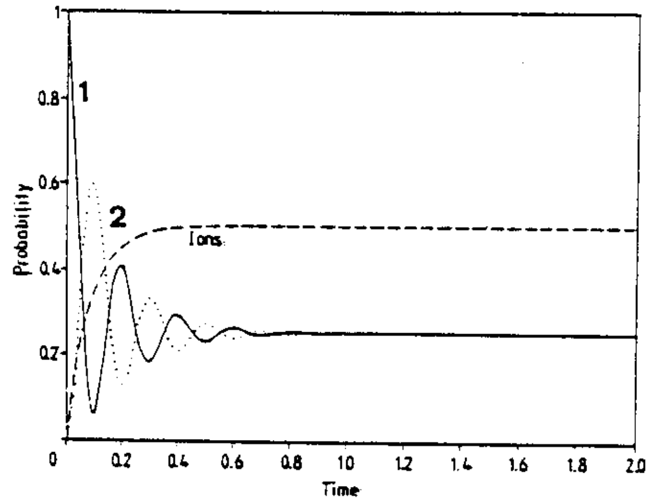


Fig. 8. - Time evolution of the probability of populations in the states $|1>$ and $|2>$ and the ions for resonance conditions $(\Delta_1 = \Delta_2 = 0)$ and relaxation rates of states $|1>$ and $|2>$ equal 1 (reproduced from ref. 36 with permissioin of the authors).

The second scheme, represented by Fig. 7b, has been investigated theoretically at first by Lami and Rahman [37] and results for the absorption of one laser radiation or for the ionization probability into the continuum states have shown up the characteristic lineshapes of the coherent trapping. The scheme of Fig. 7b shows a Fano lineshape profile produced by the coupling between the $|2>$ state and the continuum. In presence of two laser radiations and coherent trapping mechanisms a distorsion of the Fano profile by the coherent trapping is recovered as a function of the coupling parameters in the system.

Of course the phenomena presented above are modified by the presence of strong laser radiations, and unusual and interesting properties appear in those conditions. A review of most of the theoretical work is in reference [38].

New coherent features in the laser continuum structures have been presented in [39] and interference effects in the continuum-continuum transitions have been presented in [40].

If the phenomena involved with the continuum have been studied in detail theoretically, a limited amount of investigation has been performed from the experimental point of view. The laser induced structures in the continuum have been investigated experimentally in alkalis atoms with excitation from the ground state and an excited state to the continuum. Heller et al [41] have monitored the modification in the polarization of a probe ω_2 laser near the Raman resonance condition in the scheme of Fig 3.1a for the cesium atoms. Later the presence of laser induced structures in the continuum was used to enhance the third harmonic generation in sodium atoms [42]: the continuum was reached from the ground state through three photon excitation and the photons of the ω_2 laser connected the $5s$ sodium state to the three-photon laser induced structures in continuum.

A similar experiment has been performed in the case of xenon [43], where the three-photon ionization by absorption of ultraviolet excimer laser radiation was modified by the simultaneous presence of radiation at 400.73 nnm used to embed, starting from the $10p$ $J = 0$ state, an autoionizing state in the continuum at the required position to affect the laser induced structures. The continuum structures observed in the experiment on the ionization rate are in agreement with the expected theoretical profiles.

It has to be pointed that very narrow structures in the laser ionization spectra of barium and sodium atoms have been observed in two different experiments [44, 45]. Those observations are not strictly connected to the coherent trapping as presented above, but are evidence of an interference occuring in the laser simultaneous excitation to two levels with different ionization behaviours.

4. - Constants of motion

The constants of motion in the time evolution equations of the density matrix are very useful quantities in the analysis and predictions of the response in an atomic or molecular system to the externally applied electromagnetic fields. In the two level system the constants of motion are described geometrically through the introduction of the Bloch vector. The question of the constants of motion for a three level system has been considered by Elgin [46] and Hioe and Eberly [47] on the basis of the generators of the SU(3) group. Those authors have shown that a classical vector description may be used for any three level system in presence of laser fields and that appropriate constants of motion may be introduced to investigate the time evolution. Those analyses have been introduced to describe the time evolution of the density matrix in a time period short as compared to the relaxation decay times, whence for the

time evolution of the density matrix given by the Liouville equation:

(5) $$i\hbar\frac{\delta\rho}{\delta t} = [H,\rho]$$

Furthermore it has been shown by Hioe [7] that under appropriate conditions, as the two-photon resonance or equal Rabi frequencies α and β or equal detunings Δ_1 and Δ_2 additional symmetries, represented by subgroups of the SU(3) group, may be applied to provide additional insight into the density matrix properties.

If the eight SU(3) generators are introduced

(6) $$s = \{s_i, i = 1,8\}$$

with commutation relations

(7) $$\{s_i, s_k\} = 2i\, s_\ell f_{ik\ell}$$

where $f_{ik\ell}$ is the structure constant associated to the generator of the SU(3) group, the Hamiltonian and the density matrix in Eq. (5) may be expanded as a function of the generators and the identity matrix I. Thus if we define

(8) $$\Gamma_j(t) = Tr(H s_j)$$

and

(9) $$S_j(t) = Tr(\rho(t) s_j)$$

an equation of the motion results for the coherence vector $S(t)$, namely;

(12) $$\frac{d}{dt}S_j = \sum_{kj} f_{jk\ell}\Gamma_k(t) S_\ell$$

The geometric representation of the vector S is equivalent to the Bloch vector for the two-level system and may provide an useful description of the time evolution of the density matrix, as explored by Ho and Chu [48].

The explicit form of some S_j is given by:

(10a) $$S_1 = \epsilon^{-1}(\alpha u_{12} + \beta u_{23}),$$

(11b) $$S_2 = \epsilon^{-1}(\alpha v_{12} - \beta v_{23}),$$

(11c) $$S_3 = \epsilon^{-2}(\alpha^2\rho_{11} - \epsilon^2\rho_{22} + \beta^2\rho_{33} + \alpha\beta u_{13}),$$

(11d) $$S_8 = \frac{1}{\sqrt{3}\epsilon^2}[(\alpha^2 - 2\beta^2)\rho_{11} + \epsilon^2\rho_{22} + (\beta^2 - 2\alpha^2)\rho_{33} + 3\alpha\beta u_{13}],$$

(12a) where $\epsilon = (\alpha^2 + \beta^2)^{1/2}$,

(12b) and $u_{jk} = \rho_{jk} + \rho_{kj}$,

(12c) $v_{jk} = i(\rho_{jk} - \rho_{kj})$

For the two-photon resonance condition, whence the coherence trapping condition, it turns out that the following conservation laws are valid:

$$[S_1(t)]^2 + [S_2(t)]^2 + [S_3(t)]^2 = \text{const} \tag{13a}$$

$$S_8(t) = \text{const} \tag{13b}$$

in addition to the general conservation law:

$$Tr\,\{[\rho(t)]^j\} = \text{const} \quad j = 1,2,3 \tag{13c}$$

The conservation law (13b) implies coherence trapping and is equivalent to the existence of the strangeness quantity in quark physics. Furthermore this condition is equivalent to the condition expressed by Eq. (3) for the uncoupled state $|r>$ in the coherent trapping. It should be investigated if the other conservation laws may be connected to other obsevables of the system and which new information can be obtained from the experimental detection of those conservation laws.

5. - Conclusion

Quantum mechanical interference effects that involve the existance of two paths for the final process are well know in the laser spectroscopy investigations. The Hanle effect represents an important example where a coherent superposition of states radiate by spontaneous emission, and the interference in the emission leads to a modification in the spatial distribution of the emitted light. The two-photon interference originated by the Zeeman structure in the intermediate state, is another example of the interference in the preparation of the final state population through two different paths. However the coherent trapping has shown that a large variety of observations arise in different experimental configurations as consequence of the presence of nonlinear pumping and interference.

It is easy to predict that similar phenomena take place also in the interactions between multilevels systems, with discrete or continuum states, and several radiation fields, and that new faetures will be observed in high resolution spectroscopy experiments involving more than two laser beams. At the same time it may well arise that stringent experimental conditions are to be satisfied for the detection of new ultranarrow structures, so that their exploitation will be not so wide as in the case of the coherent trapping.

This paper was written in honour of Adriano Gozzini, whose interest for understanding in detail the interaction between electromagnetic radiation and absorbing media has been for me a constant inspiration and guidance.

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