OBSERVATION OF THE MODIFICATION OF OPTICAL COLLISION DYNAMICS IN INTENSE LASER FIELDS

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Introduction

In this paper we shall discuss an experiment [1] on the observation of atomic collision dynamics in strong laser fields. The method we have used is based on the interpretation of the spectrum emitted by an atom undergoing collisions in the presence of an intense laser field. An appropriate name for the method is dressed state collision spectroscopy. In this article we would like to give an assessment of the method along with an indication of the direction for future work. We shall start with a brief and rather qualitative discussion of the theory.

Dressed States

Dressed states have proven to be a very fruitful and natural language for the analysis of intense field scattering experiments [2,3]. This is because they focus on the best variables for the case in which an intense field drives an atomic transition.

We shall suppose that the incident laser field may be well represented by a classical time-dependent field. This is completely equivalent to a fully quantum-mechanical treatment in many cases. The state of the atom in the presence of this field may be obtained conveniently in the following manner.
The explicit time dependence of the problem is removed by transforming to a rotating frame — in the case of a two state problem this transformation has the geometric interpretation given by the Bloch representation [3].

In the rotating frame and neglecting rapidly varying terms the now near degenerate states are coupled by a time-independent term in the Hamiltonian. We can then diagonalize the Hamiltonian in the rotating frame to obtain the dressed states. The formal theory of dressed state radiative and collisional relaxation then follows in the same manner as for "bare" atomic states. For a detailed treatment we refer the reader to Rabin and Ben-Reuven [4], Burnett et al. [5] and Reynaud and Cohen-Tannoudji [6]. In figure 1 we have the relevant diagram for the dressed states for the case of a \( J=0 \) to \( J=1 \) transition driven by a linearly polarized laser. The diagram indicates the various collisional and radiative rates that have to be determined. Note that the spontaneous rates are interpreted as a coupling between two different sets of dressed states corresponding to different numbers of laser photons present — this is consistent with the use of a single laser field strength since \( \sqrt{N+1} \sim \sqrt{N} \).

![Diagram](image)

**Fig. 1.** Dressed states for \( J = 0 \leftrightarrow J = 1 \) system: \( \leftrightarrow \) collisional rates, \( \longleftrightarrow \) radiative rates.
In figure 2 we indicate the type of spectrum one observes from such a system. To calculate the detailed form of this spectrum we need to calculate the full density matrix for the system. In the binary collision and secular approximation the equation of motion for the dressed frame density matrix is Markoffian and the spectrum is related directly to the density matrix elements [4-6]. The spontaneous decay rates are obtained by direct transformation from the bare states. The calculation of the collision rates is a much more complex task involving the determination of $S$-matrix elements from the dressed-state-perturber interaction.

In order to appreciate the results discussed below we should discuss what behavior we should expect for the dependence of the dressed state collision rates on intensity. In the weak-field limit we can interpret the transition from $|1\rangle \rightarrow |2\rangle$, indicated in figure 1, as absorption of a photon in the middle of a collision. In the dressed state picture the potential curves representing the interaction with a perturber look

\[ \beta = \frac{\Omega - \Delta}{2} \]

\[ \omega_L \]

\[ \omega_0 \]

\[ \text{collisional} \]

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**Fig. 2.** Typical spectrum from Sr-Ar system in intense field: upper trace, parallel polarization (to laser); lower trace, perpendicular polarization.
roughly like those shown in figure 3. We see that as the field strength increases the potential curves are repelled (fig. 4) and in the region of the curve crossing (diabatic weak-field picture) the curves strongly avoid each other. We should expect, therefore, that for intense fields the transition $|1\rangle \rightarrow |11\rangle$ should switch off. We shall see that this is indeed what happens.

Fig. 3. Dressed state picture of absorption during a collision.

Fig. 4. Strong field limit of dressed state picture of absorption during collision.
The Experiment

A description of the experiment is given in Ref. [1] and we shall only briefly recall its salient features here.

A linearly polarized N$_2$-pumped H"{a}nsch-type dye laser with a bandwidth of 0.03 nm, and a pulse length of 7 ns was tuned 17 cm$^{-1}$ to the red of the Sr 460 nm resonance transition and tightly focused into a stainless-steel cross oven containing strontium vapor at 600 C ($\sim$2 x $10^{14}$ atoms cm$^{-2}$) and an argon buffer gas at pressures of 10 to 50 torr. The laser intensity was $\sim$500 MW/cm$^2$ at the center of the oven. The scattered light was detected at 90° through the sidearm of the oven, and focused onto the entrance slit of the monochromator. A polarizer was included in the detection path to obtain the polarized scattered spectrum. The spectrum observed was a time integrated one. The manner in which the theoretical prediction for the spectrum was obtained is described below.

Modeling of the Laser Pulse: Theoretical Predictions and Comparison With Experiment

To obtain predictions for the polarized scattered spectrum we need a model for the spatial and temporal behavior of the laser pulses. In our initial report (Ref. [1]) our theoretical predictions were based on an adiabatic square [3,7] pulse. Since then we have constructed a much more accurate model of the pulse based directly on the measured shape of the pulse in space and time.

The behavior of the adiabatic dressed state density matrix elements was then calculated as a function of space and time for the various atoms in the pulse and all the necessary averaging performed to obtain the prediction of the observed spectrum. In figure 5 we show the predicted variation of two of the components as a function of on-resonance Stark shift over the detuning.
Fig. 5. 20 Torr data (open points) and theory (solid curve) for intense field collisional depolarization.

The calculation of the dressed state collisional rates was based on the formalism of Light and Szöke [8] and the qualitative form of the rates is very similar to the results obtained by these authors. As one can see the agreement with theory based on these rates is rather good.

What does this variation mean physically? We should first note that $I_\perp$ is proportional to the population in state $|\Pi\rangle$ and depends on the rate of excitation from $|I\rangle$ to $|\Pi\rangle$. $I_\parallel$, on the other hand, depends on the population in $|m_J = \pm 1\rangle$ and hence on the rates from $|I\rangle$ to $|m_J = \pm 1\rangle$. The fact that the ratio $I_\perp/I_\parallel$ increases may thus be interpreted as being due to the transitions from $|I\rangle$ to $|\Pi\rangle$ being switched off as discussed above. As Light and Szöke first pointed out the transitions to $|m_J = \pm 1\rangle$ do not switch off and $I_\perp/I_\parallel$ becomes larger as the field strength increases.
Discussion and Future Work

We have shown that dressed state collision rates may be determined via the spectrum emitted by atoms undergoing collisions in the presence of an intense laser field. The method is limited by some practical matters that we shall discuss briefly here.

The first problem involves propagating an intense laser field through the vapor in a cell. We have shown that parametric processes can easily mask genuine collisional effects on the spectrum. In order to avoid such problems, rather low vapor pressures must be used — this limits the signal size rather severely. In the future we hope to use a crossed beam arrangement to avoid such problems — a supersonic nozzle source should provide sufficient densities in the interaction region.

The experiments cannot be performed using a cw laser — the necessary field strengths are much too large. We should like, therefore, to eliminate any possible uncertainties in the calculation of the time-dependent spectrum by determining the full spectrum of any fluctuations in the pulse amplitude. This is by no means a trivial task, but will be given a good deal of attention in the future.

To conclude, we should emphasize that in spite of these practical difficulties the technique is an important and powerful one with, in our estimation, a promising future.

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