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COLLISIONS IN THE PRESENCE OF A LASER FIELD AND THE LASER AS A TOOL FOR
STATE SELECTIVE PREPARATION OF MOLECULAR STATES IN COLLISIONS

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Résumé - Dans l'étude d'événements collisionnels individuels, le rayonnement
laser peut être utilisé pour modifier ou sonder le processus avant, pendant ou après l'interaction binaire entre les particules. Nous discu­
tons certains problèmes et certaines possibilités particulièrement in­
téressantes pour modifier le processus de collision dans un champ laser
intense, mais pas trop intense. Nous discutons la possibilité de pré­
parer sélectivement des états quasi-moléculaires \( \Sigma \) et \( \Pi \) dans des col­
lisions ion-atome, à partir d'états atomiques asymptotiques p pompés
optiquement par laser.

Abstract - In the study of individual collision events laser light
can be used to influence or probe the process prior to, during, or
after the binary particle interaction. We discuss some problems
and particularly challenging possibilities for modifying the colli­
sion process in a high, but not too high, laser field. We discuss
the possibilities of state selective preparation of quasimolecular
\( \Sigma \) and \( \Pi \) states in ion-atom collisions, with asymptotically laser
optical pumped atomic p-states.

I. INTRODUCTION

The main subject of this workshop, collisions in the presence of a laser field,
can be interpreted rather broadly: The laser may act on the system

a) while the colliding particles are still infinitely separated,

b) during the actual collision process, i.e., at internuclear distances where the
collisional interaction potential has become significant,

c) after the collision has occurred to probe the state of the system.

Going through the talks given at the workshop one finds that the majority of the
contributions concentrate on aspect a) while one is inclined to find aspect b) and the possibilities of modifying the interaction itself perhaps more spectacular.

Obviously this topic is much more difficult to access experimentally than theo­
retically due to the limitations on available laser power. A simple time scale
argument illuminates this: The time of passage of an atomic beam through a laser
beam may be \( 10^{-6} \) sec, the spontaneous lifetime of an excited atomic beam about
\( 10^{-8} \) sec, the time for induced transitions (i.e. the inverse Rabi frequency)
around \( t_R \sim 10^{-9} \) sec (at a conveniently available cw-laser intensity of \( 1 \ W/cm^2 \)),

however, the collisional interaction time is $t_c \sim 10^{-12}$ to $10^{-15}$ sec depending on energy and type of particle interacting with the atom. Thus, we have in most practicable cases $t_c \ll t_R$ which implies that during an individual collision act the laser cannot modify the atomic states very much. Thus, in order to observe the influence of the laser on an individual collision event we have to either observe very many collisions (typically in a bulk experiment thereby renouncing the degree of detailed information observable in a crossed beam experiment), or we have to increase the laser power drastically by a factor $(t_c/t_R)^2$, since the Rabi oscillation time scales with the square of the laser intensity. Thus this gives laser powers of the order of $10^8$ to $10^{10}$ W/cm², not impossible but difficult to use in a collision experiment. Then the observable effects are of great variety and some are genuine laser effects, i.e., effects that could not be observed with a weak photon source even if one were willing and able to accumulate data for a very long time. We will discuss one of these genuine laser effects which could open new paths for a detailed study of collisional scattering amplitudes and which so far has not achieved great attention.

II. LASER-INDUCED COHERENCE OF ELASTIC SCATTERING AMPLITUDES FOR DIFFERENT ELECTRONIC STATES

A typical differential cross section experiment involving laser excitation of an atomic resonance level usually cannot detect any interference between ground and excited state scattering amplitudes $f_g$ and $f_e$. Mostly, such studies² have concentrated on inelastic processes and their amplitudes $f_{ge}$. However, one might be interested in exploiting the coherence between ground and excited state which the laser introduces.

Let us consider a hypothetical two-level atom whose energy levels in a weak or strong near-resonant laser field are shown in Fig. 1; $|e\rangle$ and $|g\rangle$ indicate the original ground and excited states, $|e\rangle$, $|e1\rangle$, $|g\rangle$, $|e2\rangle$, $|g2\rangle$ designate these states plus the photon field with $n$ or $n+1$ or $n+2$ photons. The dressed states in the strong field are coherent superpositions of ground and excited states, the two states relating to the same energy level are split by $\hbar\Delta R$ where $\Delta R = 2\pi/t_R$ is the Rabi frequency. Transitions between these dressed states have been observed spectroscopically in cell experiments by observing the re-emitted resonance fluorescence.³ In a crossed beam experiment one would have to observe the processes 1, 2 and 3 by appropriate energy analysis of the particles if one wants to avoid coincidence experiments with high optical resolution.

The differential cross sections measurable in crossed beam experiments would be related¹ to the scattering amplitudes as shown in Table 1.

![Energy levels of a two-level atom in a laser field and some possible collisional transitions.](image)

**Fig. 1.** Energy levels of a two-level atom in a laser field and some possible collisional transitions.
Table I. Laser split level cross sections. Initial kinetic energy $E_o$, laser tuned on resonance, spontaneous decay neglected.

<table>
<thead>
<tr>
<th>Process</th>
<th>Final kinetic energy</th>
<th>Differential cross section</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$E_o$</td>
<td>$\frac{1}{4}</td>
</tr>
<tr>
<td>2</td>
<td>$E_o + \hbar\Omega_R$</td>
<td>$\frac{1}{8}</td>
</tr>
<tr>
<td>3</td>
<td>$E_o - \hbar\Omega_R$</td>
<td>$\frac{1}{8}</td>
</tr>
</tbody>
</table>

sum of 1,2,3       $\sim E_o$       $\frac{1}{2} |f_g|^2 + \frac{1}{2} |f_e|^2$

We see that interesting interference effects between ground and excited states could be observed, when the transitions among the (very closely spaced) dressed levels were observable individually in a differential scattering experiment. These effects cancel if, as usual, one sums over all processes 1,2,3 in the experiment. The splitting of the levels at easily accessible 1 W/cm² is typically $5 \times 10^{-7}$ eV and at best one would be able to resolve some meV with state-of-the-art particle spectroscopy. So, a factor of $\sim (10^4)^2 \equiv 10^8$ W/cm² is needed to do the experiment — challenging and perhaps not impossible to do in the near future. Also worthwhile considering is the possibility of using $\pi/2$ pulses for creating a coherent ground-excited state population.

III. PREPARATION OF QUASIMOLECULAR COLLISION STATES BY POLARIZED LASER LIGHT

Many of the other processes, dealing with atom-atom, ion or molecule interactions which are the focus of this workshop can be schematically summarized as shown in Fig. 2.
Figure 2(a) indicates asymptotic state selective preparation of atomic np' states by a polarized laser with subsequent superelastic collisions occurring at the curve crossing marked 'C.' Figure 2(b) represents a prototype of laser assisted collision where the photon is tuned in resonance with the molecular states accessible only during the collision. Figure 2(c) shows the collisional redistribution of radiation experiment as reviewed by Burnett, where the emphasis is (as in Fig. (a)) on the population or repopulation of the Σ and Π states in the circled area around R_L. Finally, for comparison, in Fig. 2(d) a classical line shape experiment is illustrated; the asymptotically excited atom collides with B and the quasimolecular system A*B re-emits radiation during the collision. Figure 2 illustrates the close relation of all these types of processes.

We discuss in more detail the particular aspect of whether, and how, it may be possible to prepare or detect the quasimolecular excited Σ and Π states. Take the example of the process in Fig. 2(a), which has been studied in detail, both experimentally and theoretically for the model case

\[ \text{Na}^+ + \text{Na}^*(3p) \rightarrow \text{Na}^+ + \text{Na}(3s) \]  

(1)

in a center-of-mass energy range of 15 to 50 eV. The linearly polarized laser prepares asymptotically the 3p_0 or 3p_π atomic states predominantly. The crucial question is, how do these atomic states correlate with the molecular Σ and Π states. From the potential energy diagram Fig. 2(a) we know that only preparation of the Π state can lead to the nonadiabatic process via curve crossing 'C'. Figure 3 shows the experimental result of the dependence of the differential cross section for process (1) on the laser polarization angle: For 0° one prepares asymptotically the 3p_0 orbital, for 90° the 3p_π. Obviously, the maximum is found near 90° and the asymptotic 3p_π preparation thus leads into the Π quasimolecular state. A closer look shows that the maximum of the cross section is found at an angle slightly less than 90°. The intuitive interpretation of this phenomenon is shown in the lower part of Fig. 3. At large internuclear distances, the atomic charge cloud moves space fixed, at small internuclear distances it moves body fixed, i.e., rotates with the internuclear axis. The transition between the two types of behavior occurs around R_L in the circled region of Fig. 2(a). A semiclassical calculation shows that this intuitive concept holds as long as the impact parameter \( b \ll R_L \) but loses its meaning for \( b \gg R_L \). In the above case, \( R_L \) is about 22 a.u.
At this point, we would also like to briefly mention a left-right asymmetry in the differential cross section if the atoms are prepared with circularly polarized light. In process (1) one finds a relative difference in the scattering signal of \(-30\%\) at 170 eVdegr, varying only slightly with collision energy. Here a warning is appropriate: one should not attempt to use concepts that are too simple to explain the result. The semiclassical calculation\(^6\) shows that the angular momentum transfer associated with the left-right asymmetry occurs at very large internuclear distances \(R \gtrsim R_L\) due to different phase factors for the states evolving on the long-range parts of the molecular \(\Sigma\) and \(\Pi\) states.

IV. CONCLUSIONS

We have seen that the laser radiation used to diagnose individual collision processes offers a variety of sharp tools to probe the interaction. The outcome of these experiments can now be understood for some model cases as reviewed in Ref. 6. On the other hand, the laser has, up to now, not been fully exploited in modifying collision processes during the interaction. In particular, coherent superposition of scattering amplitudes for different atomic levels could be enforced by a strong laser field, leading to a new variety of collision studies.

References

4. K. Burnett, this volume.