Using Photo-Associative Ionization of Sodium to Demonstrate the Optical Control of Cold Collisions

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We present a study of cold collisions in a sample of magneto-optically trapped sodium atoms througli the technique of two-color photoassociative ionization spectroscopy. We demonstrate the inhibition of the process by adding an auxiliary "suppressor" laser beam and analyze tlie dependence of this optical shielding effect on the laser frequency and intensity. The possibility of using photons to control the atomic collision and to modify the thermodynamic properties of tlie cold gas is discussed.

I. Introduction

Tlie study of collisions involving laser cooled and trapped atorns has been the subject of intense investigations during the past few years. This interest has been motivated by tlie desire of achieving the regime of a degenerated quantum gas and also by intrinsic features it presents, inainly tliat in tlie presence of light, collisions involving excited state atoms have duration comparable to the spontaneous emission time. In this regime, the exchange of energy between the atom and the modes of the radiation field (including vacuum) provides a prototype for studying the properties of nonequilibrium open systems coupled to reservoirs. As the kinetic energy of a two-body collision approaches zero, the number of partial waves coiitributing to the elastic collision reduces to one, the s-wave, and the information about the atomic interaction is comyletely contained in the scattering length **a.** The properties of a cold gas are therefore dependent on the scattering lengtli and its sign can determine the behavior of the system. The recente achievement of the quantum gas regime iii a sample of laser cooled and magnetic trapped rubidium atoms[1] has opened new possibilities of studies involving the thermodymanics of a gas in such regime.

Controlling the atomic interaction with photons can provide the condition to stabilize tlie gas in a single quantum state of the confining potential and also the manipulation of its thermodynamical properties. We have recently demonstrated the possibility of controlling the atomic interaction by using photo-associative ionization (PAI) in a sample of cold sodium atoms held in a trap[2]. In this paper we present the studies of cold collisions using PAI as a prototype and the route we have taken towards the demonstration of optical control of cold collisions. We discuss the possibility of using photons as a tool to cliange the course of the atomic encounter, including the modification of attraction into repulsion aiid its applications.

II. Photo-associative ionization as a prototype of cold collisions

Conventional associative ionization occurring at ordinary temperatures proceeds in two distinct steps: excitation of isolated atoms followed by the collisional interaction between excited atomic states. Tlie collision event is fast compared to the radiative relaxation and the two steps are decoupled. In contrast, PAI starts with ground state partners moving sufficiently slow that they have time to absorb and spontaneously emit photons prior to the final ionizing interaction. The partners must be close enough when the initial absorption takes place such tliat a significant fraction of the excited population survives to relaxation back to the ground state. Thus, PAI starts by promoting the ground state of the colliding species, designated by $[Na_{**}Na]$, to an

intermediate excited molecular state at long range, designated by $[Na_{**}Na]^*$. As the partiiers accelerate towards eacli other along the incoming trajectory, tlie excited quasi-molecule absorbs a seconcl plioton to a doubly excitecl state $[Na_{**}Na]^{**}$, which then pioceeds to autoionize at close range. This three-step process is schematized as follows:

$$\begin{aligned} Na + Na &\rightarrow Na_{**}Na + h\nu_1 \rightarrow [Na_{**}Na]^* \\ [Na_{**}Na]^* + h\nu_2 &\rightarrow [Na_{**}Na]^{**} \\ [Na_{**}Na]^{**} &\rightarrow Na_2^+ + \mathrm{e}- \end{aligned} \quad (1)$$

The rate constant of PAI, K_{PAI} is given by $d[Na_2^+]/dt = K_{PAI}[Na]^2$, where $[Na_2^+]$ ancl [Na] are respectively the ion and atom densities. Tlie first study we have carried out on PAI was related to tlie measurement of K_{PAI} as a function of light intensity[3]. In order to do that, we usecl an experimental set up which consists of a vapor cell magneto-optical trap, already describecl in details elsewhere[4]. Briefly, sodium vapor containecl in a stainless steel chamber is captured and cooled usiiig laser light beams and a magnetic field gradient of about 20 Gauss/cm. Tlie inagnetic fielcl creates a spatial selection rule for electronic transitions such tliat counter- propagating laser beams in three orthogonal clirections create a viscous ancl restoring force for atoms which are then confined in a small volume ($\sim 10^{-4}$ cm^3), at a density of the order of 10^{10} cm^{-3}. Tlie deterinination of K_{PAI} requires accurate ineasurements of tlie ions prodiiction rate and of the atomic density. We obtain the density by imaging the bright trap fluorescence onto a calibrated photomultiplier tube, while measuriiig its dimensions with a survey telescope and a CCD camera. A channeltron particle multiplier with a negatively biased grid is placed near the trap center. It collects the ions produced which are then counted by a discriminator and a gated countei electronics. The photons tliat produce PAI are the same that hold the atoms trapped in tlie $3S_{1/2}(F = 2) \rightarrow 3P_{3/2}(F = 3)$ transition.

The dependence of K_{PAI} on tlie trapping laser intensity is shown in Fig. 1. When compared to the existing theories[3], we observe that the increase of K_{PAI} with the laser intensity is actually smaller than the prediction (solid line in Fig. 3). A possible reason for the difference coulcl be related to the depletion of tlie number of atoms in tlie ground state inside the collision volume as tlie light increases, a fact not taken into account in tlie theories. A second possibility for the discrepancy between experiment and theory could be the excitation of repulsive states which would change the conventional path towards the PAI process. In any case, the theoretical model still needs some improvement in order to describe the observations with more realism.

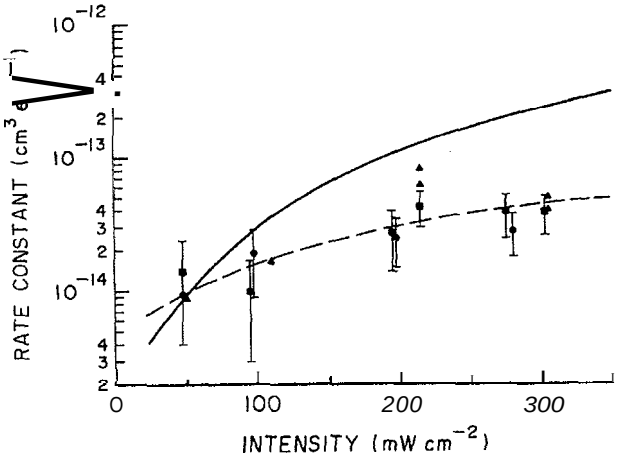

Figure 1. Dependence of the photoassociative ionization rate constant on tlie trapping laser intensity. The dashed line connecting tlie experimental points is just for visual aid, while tlie solid line represents the theory of Ref. [3], based on the optical Bloch equations.

III. Two-color PAI and the observation of long range bound states

To be able to investigate PAI in a greater detail we must have the ability to observe individual steps involved in tlie process and to reach this aim we have performed an experiment using photons with different colors for each of the steps involved in the PAI. The atomic sample used in this experiment is a conventional trapped atomic cloud (T $\sim 250\mu$K, n $\sim 10^{10}$ cm^{-3} N $\sim 10^6$ atoms) describecl elsewhere[3]. To obtain a more controlled result, the atomic sample is prepared in one of the hyperfine states of Na ground state.

The operation of tlie MOT requires a trapping transition $[3S_{1/2}(F = 2) \rightarrow 3P_{3/2}(F = 3)]$ operating with a laser of frequency ω_1 and a repumper transition $[3S_{1/2}(F = 2) \rightarrow 3P_{3/2}(F = 2)]$ at a frequency ω_2, whicli recovers population that eventualy leaks out of the $3S_{1/2}(F = 2)$ ground state. Our measurements are

carried out with ω_2 turned off such that ω_2 can optically pump all the atoms to the $3S_{1/2}(F = 2)$ ground state and thus, we investigate collisions starting with a sample 100% pure in that ground state. The trapping frequency, ω_1, is generated by a switchable electro-optical modulator (EOM) operating at 1.71 GHz while the repumper frequency is the carrier itself. A second laser, working as a probe, is introduced into the trap in such a way to copropagate with the trapping beams. Its frequency, ω_p, is scanned from the trapping transition frequency down to 3 GHz of it, while the intensity can go as high as 1 W/cm^2. The experiment measures the rate of ions produced as a function of the probe laser frequency. A schematic diagram showing the four steps involved in this experiment is presented in Fig. 2.

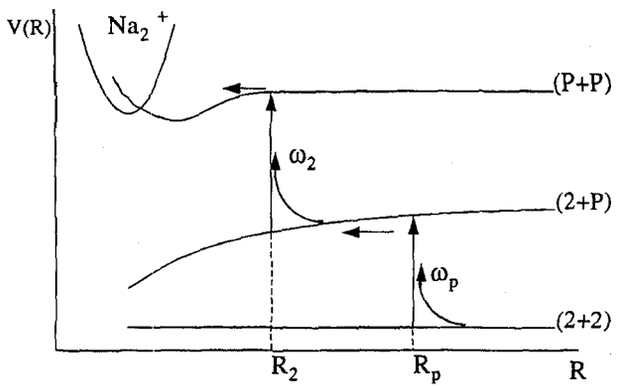

Figure 2. Schematic diagram of the photoassociative ionization process.

The colliding partners start the collision both in the hyperfine ground state F=2, represented by the energy asymptote (2+2). As they reach the nuclear separation R_p, the probe photon connects (2+2) to the attractive state (2+P). Under attraction, atoms accelerate towards each other until the point R_2, where the frequency ω_2 intercepts the incoming scattering flux and transfers it to the doubly excited level (p+p), which may have enough kinetic energy to survive without spontaneous decay until reaching short distances where the ionization takes place. For ω_p close to ω_2, the kinetic energy gained by the atoms is not large enough for the partners to survive at (p+p) until short range. The ionization process is turned off for $R_c \leq R_2$ due to energy conservation.

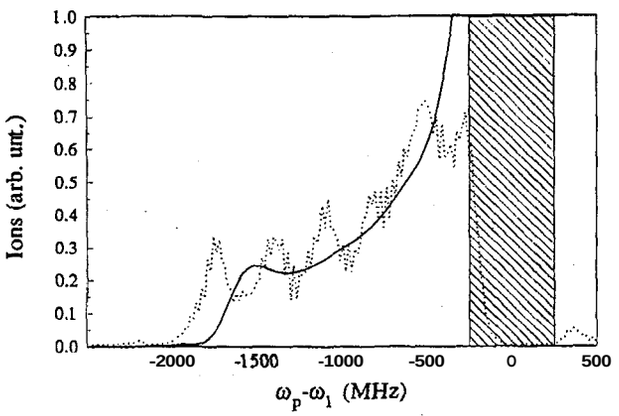

Figure 3. Two-color photoassociative ionization spectrum of magneto-optically trapped atoms (dashed line) and the curve calculated with a local equilibrium theory (solid line). Inside the dashed region, the probe laser influences the trap conditions, decreasing strongly the number and density of atoms such that the experimental results are not reliable.

Fig. 3 shows the spectrum obtained (dashed line) by scanning the probe laser ω_p about 3 GHz to the red of the trapping transition (ω_1). The solid line represents a lineshape calculation using a local equilibrium theory[5]. As can be observed, the model predicts quite well the overall lineshape, indicating a good understanding of the steps involved in the process. The structure in the measured spectrum is related to free-bound-free transitions. This corresponds to long range bound states of the (2+p) R^{-3} potential, already described elsewhere[6]. Since the model we have used to obtain the overall lineshape presented in Fig. 3 does not include the existence of bound sates in the intermediate attractive potential, it can not reproduce the structure observed in the measured spectrum. The dashed region around $\omega_p - \omega_1 = 0$ represents the frequency interval where the trap performance is affected by the probe laser. Therefore, the experimental spectrum shows a considerable decrease on the ion production in this region.

IV. Optical control of PAI

The occurrence of PAI requires the atoms to reach the short range part of the potential. The idea here is to use a laser field tuned to the blue of the atomic resonance to access repulsive long range states and to create a repulsive barrier which prevents atoms from approaching each other. This "Optical Shielding" can substantially reduce the rate of PAI. As discussed before, the incoming flux of colling atoms has to be in the

ground state potential for the occurrence of PAI. Close to the Condon point R_p, where $\hbar\omega_p$ matches the energy difference between the molecular curves, the probe laser connects the ground state to the excited attractive state. This transfer the population to a long range attractive state drives the two atoms together. The optical control of PAI can be done at this first step. Rather than transferring the population to an attractive state, an auxiliary optical field $\hbar\omega_s$, tuned to the blue of the excited state asymptote, transfers population to a repulsive curve around the Condon point R,. In this way, the collision partners are prevented from approaching close enough for the molecular autoionization to take place the collision. As the intensity of the shielding field increases, scattering flux exits in the ground state and the PAI becomes elastically shielded. A representative diagram of the shielding effect with the routes followed in the process is shown in Fig. 4: in (a) we have the conventional steps to produce PAI and in (b),

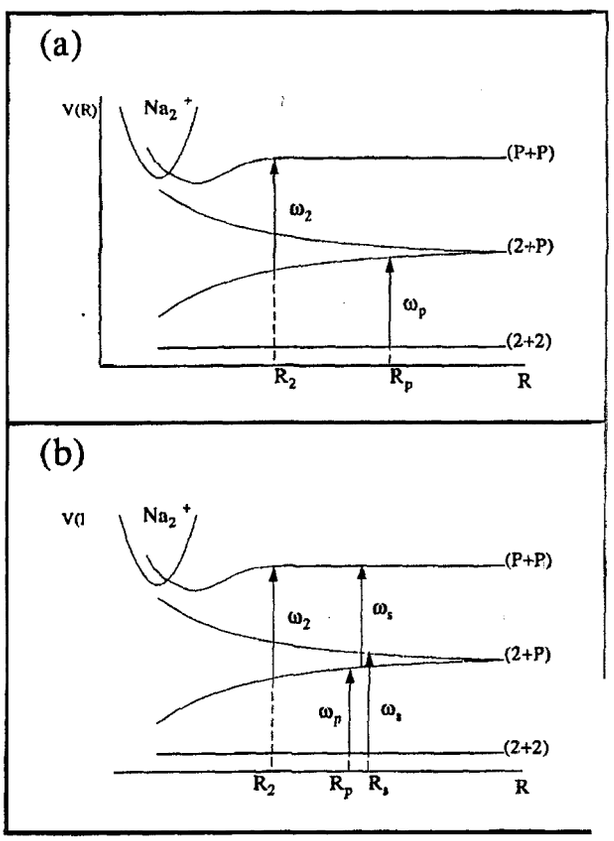

Figure 4. Diagram of the scattering flux (a) without and (b) with the suppressor light field.

the presence of the shielding field can produce different effects in the PAI. First, if w, intercepts the incoming flux before ω_p, there will be a transference to the repulsive potencial and the atoms fly apart, suppressing the PAI. On the other hand, if ω_p intercepts the incoming flux before w,, the process will not be suppressed. It is actually enhanced due to the opening of new channels of connection of w, from the attractive state to the double excited state. Fig. 5 shows the spectrum of PAI with and without the presence of the shielding field.

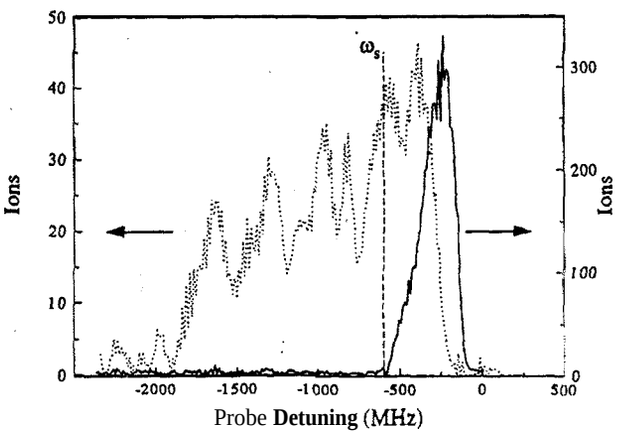

Figure 5. Two-color photoassociative ionization spectra with (full line) and without (dashed line) the presence of a suppressor laser with an intensity of 8 W/cm^2 tuned 600 MHz to the blue of the F=2 → F'=3 trapping transition. The depletion (enhancement) can be observed below (above) 600 MHz.

Increasing of the intensity of the shielding laser can promote an adiabatic transfer from (2-t-2) to (2+p) and back to (2+2)[7]. In this situation, atoms are approaching each other and, as in a hàrd sphere case, they reverse their motion and fly apart in a complete elastic process. The effective size of the sphere can be changed by tuning ω_s, which effectively changes the internuclear separation where the shielding field acts. This represents an optical control of the atomic collision which can avoid collisions, as actually done recently[8]. Optical control of cold collisions can be an useful technique to suppress undesirable collisions in order to increase trapped atom densities while enhancing ground state elastic collisions. These concepts could be explored in the context of the investigations of the thermodynamical properties of a cold gas sample. It is well known that in this regime of ultracold temperature, the atomic elastic collisions are independent of the interatomic potential details, but depends only on a single parameter obtained from the potential, the scattering length

a. Roughly speaking, a is an effective diameter for a potential acting **as** it was a liard sphere with a total scattering cross section of $4\pi a^2$. The thermodynamical properties of the quantum gas is severely dependent on the value and sign of **a**[9]. System with a negative scattering length could have this value changed by optical shielding, producing considerable moclifications on the thermodynamical behavior.

V. Conclusions

In conclusion, we have demonstrated the possibility of controlling ultracold collisions in a magneto-optical trap by using PAI **as** a prototype. Inelastic ground state collisions due to interactions in the region R < R, are greatly suppressed in favor of elastic collisions, leading the atomic pair to behave as hard-spheres. The effective diameters of the spheres can be adjusted by tunning the suppressor laser frequency and this may turn out to be an important technique for the study of the thermodynamical properties of ultracold samples.

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