

7-1998

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Sukenik, C. I.; Hoffman, D.; Bali, S.; and Walker, T., "Low Saturation Intensities in Two-Photon Ultracold Collisions" (1998). *Physics Faculty Publications*. 11.

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Original Publication Citation

Sukenik, C.I., Hoffmann, D., Bali, S., & Walker, T. (1998). Low saturation intensities in two-photon ultracold collisions. *Physical Review Letters*, 81(4), 782-785. doi: 10.1103/PhysRevLett.81.782

Low Saturation Intensities in Two-Photon Ultracold Collisions

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(Received 12 June 1997; revised manuscript received 20 February 1998)

We have observed violet photon emission resulting from energy-pooling collisions between ultracold Rb atoms illuminated by two colors of near-resonant infrared laser light. We have used this emission as a probe of doubly excited state ultracold collision dynamics. We have observed the lowest saturation intensity for light-induced ultracold collisions seen to date which we identify as due to depletion of incoming ground state flux. We have also varied the detuning of the lasers which allows us to clearly identify the effect of spontaneous emission and optical shielding. [S0031-9007(98)06710-6]

PACS numbers: 34.80.Qb, 32.80.Pj, 33.80.-b

Interactions between ultracold atoms are of considerable interest because of the appearance of novel light-induced dynamics at low temperature and the onset of quantum behavior [1]. Ultracold collisions proved to be crucial in attaining Bose-Einstein condensation, and optical control of ultracold collisions appears to provide a promising technique for producing large ensembles of ultracold molecules [2]. In this Letter, we report on a study of light-induced ultracold atomic collisions which, among other striking observations, display saturation intensities for two-photon processes that are much *lower* than observed for single-photon processes.

Any realistic theoretical treatment of excited state collisions must include spontaneous emission as collision times are comparable to excited state lifetimes [3]. Furthermore, at ultracold temperatures—unlike thermal collisions—the hyperfine interaction plays an important role in the collision dynamics [4], though the analysis of virtually all experiments to date has ignored hyperfine interactions. For the two-photon experiment reported here, we analyzed the measurements using a semiclassical model incorporating all hyperfine energy levels. This allows us to elucidate the interplay between the various processes of flux depletion, spontaneous emission, optical shielding, and Franck-Condon overlap. Furthermore, we can quantitatively explain the striking observation of very low saturation intensities in the two-photon collision process.

Ultracold collisions begin when two atoms approach each other in the ground state. The ensuing dynamics depend on the number of photons absorbed and emitted by the colliding atom pair. Absorption of one photon creates a singly excited state (SES). Acceleration on the SES produces trap loss [1]. Collisions involving the absorption of two photons, producing doubly excited states (DES), allow greater control over the collision dynamics because the relative energy of the colliding atoms is set by the *difference* of the photon frequencies. Here we show that DES collisions allow insight into some processes which are masked in SES trap loss measurements. Analysis of studies of some of the important dynamical processes of DES collisions in Na [5] were hampered by the complex-

ity of the hyperfine structure in the $P_{3/2}$ fine-structure level, thus emphasizing the importance of exploring a simpler, more tractable system such as the $P_{1/2}$ manifold in Rb [6]. Further simplification results when the initial excitation is to the lowest energy molecular hyperfine manifold, i.e., from the lowest ground state hyperfine level to the lowest hyperfine levels of the $5S_{1/2} + 5P_{1/2}$ SES. This pathway avoids the complex behavior which results from entangled attractive and repulsive potential curves.

In Rb, the collisional photoassociative ionization channel is closed [7], so we employ a fluorescence detection method which takes advantage of an energy pooling reaction. Energy pooling occurs when the atom pair in the DES reaches small separation and yields a violet photon with $\lambda = 421$ nm. We have probed the dynamics of the collision by measuring the relative violet photon production rate R as a function of laser detuning and intensity when two ir laser fields (both with $\lambda \sim 795$ nm) are applied. Referring to Fig. 1, one field, of frequency ω_1 (intensity I_1), is tuned below resonance while the other, ω_2 (I_2), is tuned above resonance. We find a strong dependence of R on ω_2 , including the surprising appearance of deep modulations for ^{87}Rb , but not for ^{85}Rb . Even more striking, we observe saturation at laser intensities much lower than in any previous ultracold collisions experiment. We identify saturation associated with ω_1 as resulting from a depletion of incoming flux [8], the first such observation of this effect. Depletion results when excitation at larger interatomic separation removes ground state flux available for excitation at shorter distances. Saturation of ω_2 is also observed at small detuning, arising from an optical shielding process [9]. Our model indicates that the modulations in the ω_2 dependence originate from two sources: the onset of new collisional channels due to hyperfine structure in the DES, and Franck-Condon (FC) overlap factors. Our data also demonstrate the strong influence of spontaneous emission and optical shielding when the detuning is near the onset of the two-photon collision.

The spectrum of the spontaneously emitted photons gives important information about the collision process.

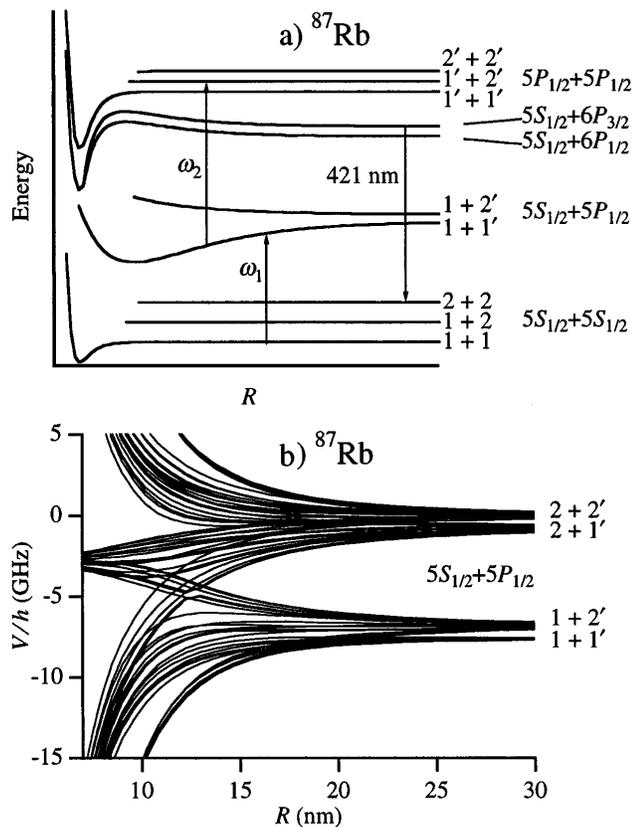


FIG. 1. (a) Schematic representation of ^{87}Rb energy levels involved in the two-photon optical collision; (b) calculated ^{87}Rb potential curves showing hyperfine structure of the singly excited $5S_{1/2}(F=1) + 5P_{1/2}(F'=2)$ manifold.

Because of the depth of the Rb molecular potentials, decay of the DES at close range could result in the emission of photons over a band of many tens of nanometers around 400 nm. Rather than observing a broad spectral distribution, however, we found that our signal was comprised exclusively of 421 nm photons arising from energy pooling: $5P + 5P \rightarrow 5S + 6P$, followed by a $6P \rightarrow 5S$ decay at 421 nm. The absolute rate coefficient for violet photon production is observed to be on the order of 10^{-14} to 10^{-13} cm^3/s for $I_1 = 70$ mW/cm^2 and $I_2 = 200$ mW/cm^2 , where I is the average laser intensity. We were able to resolve the $6P$ fine structure and found that transfer to the $6P_{1/2}$ component is favored over the $6P_{3/2}$ state by a ratio of $\sim 2:1$. This experiment is one of just two ultracold collision experiments which have successfully detected collisionally produced photons [10]. Here we measure R as a probe of the DES collision dynamics, expecting that any frequency dependence comes from light-induced dynamics at large interatomic separation, not from changes in the energy pooling probability.

In our experiment, ^{87}Rb or ^{85}Rb atoms are confined in a magneto-optical trap (MOT) similar to that described previously [11,12]. Five frequencies are derived from three external-cavity diode lasers. One laser (ω_1) is detuned $\Delta_1 = -90$ MHz from the $5S_{1/2}(F=1) \rightarrow$

$5P_{1/2}(F'=1)$ atomic transition (for ^{87}Rb), or the $(F=2) \rightarrow (F'=2)$ transition (for ^{85}Rb), and populates attractive $5S_{1/2} + 5P_{1/2}$ curves. Excitation by ω_1 typically takes place at an interatomic separation between 10 and 40 nm. The other laser (ω_2) is detuned $\Delta_2 = 0-2$ GHz above the $5S_{1/2}(F=1) \rightarrow 5P_{1/2}(F'=2)$ atomic transition (for ^{87}Rb), or the $(F=2) \rightarrow (F'=3)$ transition (for ^{85}Rb). All lasers can be switched on and off using acousto-optic modulators. The MOT lasers are switched off when ω_1 and ω_2 are present to reduce the number of collisional pathways [11]. For a typical sequence the trap laser is turned on for 80 μs , followed by 2 μs of optically pumping nearly all the atoms to the lowest ground state level, and, finally, 15 μs of ω_1 and ω_2 . Violet photons impinged on the detector at a very modest rate of only tens of photons per second, so we carefully filtered out all scattered near-ir light using a series of filters [13]. In order to obtain a violet photon spectrum, a liquid N_2 cooled charge-coupled device camera was used in conjunction with a 0.275 meter monochromator with a resolution of ~ 0.3 nm.

Our data consist of measurements of R as I_1 , I_2 , and Δ_2 are varied. In Fig. 2(a) we show R as a function of I_1 at fixed ω_2 . The curve is typical, independent of ω_2 . Notice that the saturation occurs at $I_1 \sim 30$ mW/cm^2 , much lower than previously observed in ultracold collisions. When saturation has been observed in trap-loss measurements [14] and photoassociative ionization studies [5], the saturation intensity has always been on the order of several hundred mW/cm^2 . We have identified the saturation here as arising from a population depletion mechanism similar to that suggested by Gallagher in Ref. [8]. This is the first observation of this collision mechanism. Although transitions from the ground state to a given SES state may not be saturated, after traversing several resonances the incoming ground state flux can become substantially depleted. To see this, we plot in Fig. 3 the remaining ground state flux as a function of atom-pair separation. The transition amplitudes are calculated based on the collision model discussed below.

Our data also show that ω_2 photons affect the collisions in several ways. Depending on the detuning, ω_2 can either induce excitations from the SES to one of three hyperfine levels of the $5P_{1/2} + 5P_{1/2}$ state or cause excitations from the ground state to repulsive curves of the SES. The latter excitation initiates an optical shielding process; after excitation to a repulsive curve by ω_2 the atom pair is prevented from coming close enough to be excited by ω_1 . This can occur even if $|\Delta_2| > |\Delta_1|$ because, as can be seen from Fig. 3, at a given detuning many repulsive curves have Condon points at larger R than do the attractive curves. Because only one SES is involved, depletion by ω_2 in the excitation to the DES is unimportant. The importance of shielding is demonstrated by Fig. 2(b), which shows violet photon production as a function of I_2 for two values of ω_2 in ^{87}Rb . At a detuning of 150 MHz, saturation is observed, whereas at

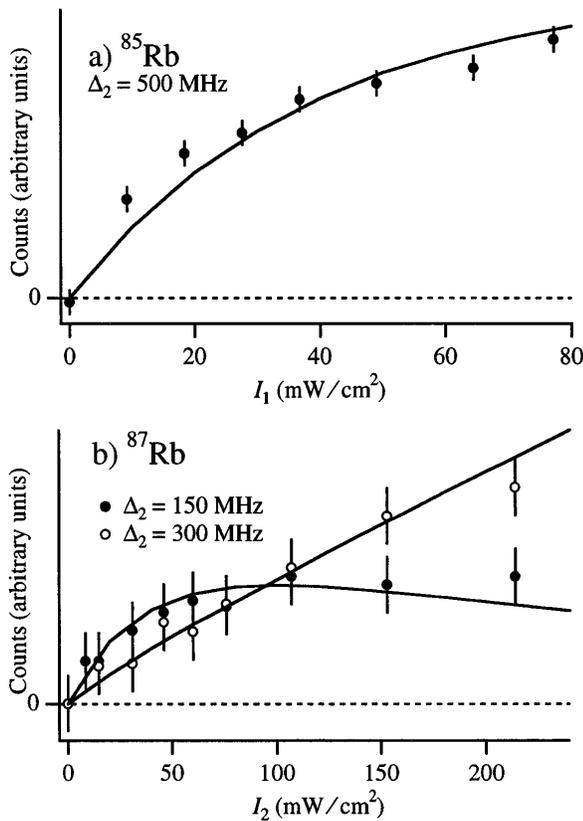


FIG. 2. Intensity dependences of violet photon production rates as a function of average intensity of (a) ω_1 for ^{85}Rb ; (b) ω_2 with $\Delta_2 = 150$ and 300 MHz for ^{87}Rb . Population depletion is responsible for the low saturation intensity in (a), while the saturation in the 150 MHz data of (b) results from optical shielding. Solid lines are results of the model described in the text.

300 MHz the rate is essentially linear with intensity. The saturation in the 150 MHz data is due to enhanced optical shielding at higher intensity. At 300 MHz, less flux is diverted by ω_2 to the repulsive SES curves resulting in little saturation of violet photon production. The shape of both curves is explained by our model and displayed with the data in Fig. 2. Thus, the saturation effects of ω_1 and ω_2 arise from very distinct physical mechanisms. For ω_1 , flux depletion produces the low saturation, while for ω_2 , optical shielding is the dominant effect.

We have also measured the violet photon production rate as a function of detuning. In Fig. 4 we show the violet photon count rate as a function of ω_2 for ^{85}Rb and ^{87}Rb . Qualitatively the two curves share many features, but in ^{87}Rb pronounced modulations of the signal are observed. We will explain these features with reference to our model which we now describe.

Our calculations begin by determining all the long-range hyperfine states [15]. We calculate the excitation probability to each SES hyperfine curve using the familiar two-level Landau-Zener (LZ) curve crossing formula, explicitly including the FC wave function overlap factor [16]. We use semiclassical WKB wave functions for

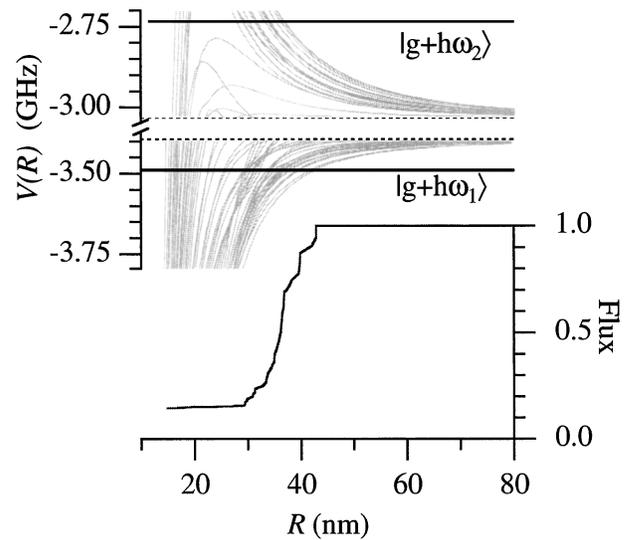


FIG. 3. Ground state flux as a function of interatomic separation for two colliding ^{85}Rb ($F = 2$) atoms. Population depletion and shielding occur as the colliding atoms traverse Condon points. A qualitatively similar curve is obtained for ^{87}Rb .

the SES and an s -wave ground state wave function with published scattering lengths [17]. The Rabi frequency used in the LZ formula is calculated using a magnetic-sublevel averaged matrix element. For all excitations, we average over the intensity of the standing waves of ω_1 and ω_2 . Optical shielding by ω_2 and flux depletion

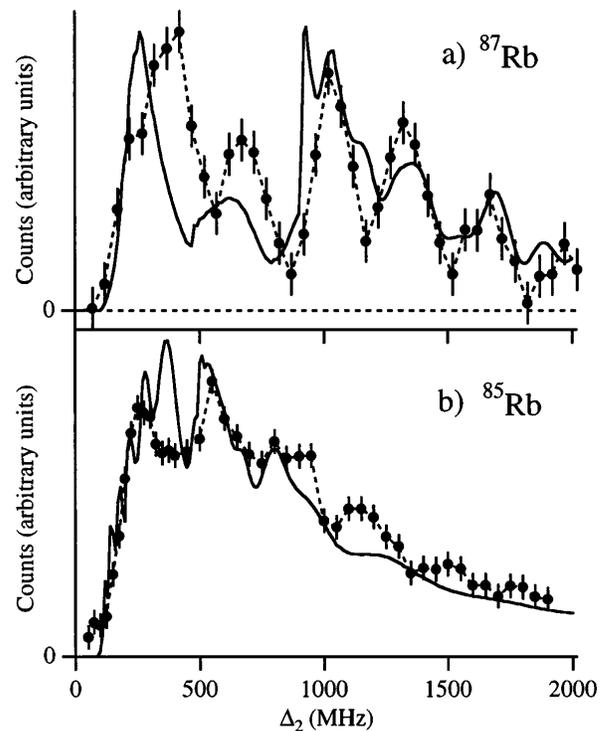


FIG. 4. Violet photon production rate as a function of Δ_2 for (a) ^{87}Rb and (b) ^{85}Rb . The solid line shows results from a calculation described in the text.

of the ground state arise naturally in the model as flux is removed from the ground state at each Condon point. Once excited by ω_1 , we allow the atoms to approach each other classically. Along their trajectory they may become resonant with ω_2 before decaying and be excited to one of the three hyperfine levels of the DES. The excitation probability to the DES is calculated according to the LZ formula. The wave functions for the three DES hyperfine levels are taken to be sinusoidal with an adjustable phase treated as a free parameter. Last, we calculate the survival probability in the DES to reach small separation (<1 nm), where we assume a violet photon is produced with a probability independent of ω_2 . We have used a single initial relative velocity of 20 cm/s.

This semiclassical model, while much more comprehensive than any others used to date in excited-state ultracold collisions, is clearly not complete. While s waves are certainly very important, higher order partial waves should also contribute to the signal. We expect that inclusion of higher partial waves in the model would result in better reproduction of the detuning dependence, whereas the effects on the intensity dependence would be minimal. By averaging over magnetic sublevels, we have neglected possible effects due to magnetic orientation. Since the excitation probability at a given Condon point is small, using a magnetically averaged oscillator strength is likely a good approximation. For similar reasons, our treatment of the Condon points as independent is probably fairly accurate. These approximations are necessitated by both tractability and an incomplete knowledge of all relevant wave functions.

Using the model, we now interpret the detuning dependence. In Fig. 4(a), the first peak, whose onset is at 90 MHz, results from excitation to the $1' + 2'$ DES [18]. Similarly, the third peak originates predominantly from excitation to the $2' + 2'$ state. The frequency difference in the onset of the peaks corresponds to the 812.5 MHz hyperfine splitting of the excited state. Other "peaks" in the data are actually modulations introduced by the FC overlap factors for excitation by ω_2 . Notice that although there is a nonvanishing oscillator strength to the lower of the three DES hyperfine levels below an ω_2 detuning of 90 MHz, violet photon production there is very strongly suppressed by optical shielding. The rather different structure for the two isotopes comes from several effects. In the case of ^{85}Rb , the smaller hyperfine splitting, the increased number of contributing hyperfine levels, and the different FC factors result in a detuning spectrum with less pronounced features than for ^{87}Rb . No single factor seems to exclusively determine the level of modulation in the data. We also note that when the ω_2 detuning is small and thus the relative momentum in the DES is low, the impact of spontaneous emission on the collision dynamics is most pronounced. The effect is manifested as a suppression of the first and third peaks in Fig. 4(a).

In conclusion, we have observed violet photon production in collisions of ultracold Rb atoms. The violet photons, which result from excitation to the $5P_{1/2} + 5P_{1/2}$ state followed by an energy pooling to the $5S + 6P$ state, are used to probe the doubly excited state dynamics. We have observed the lowest saturation intensity measured to date in an ultracold light-induced collision process. We identify this low saturation intensity as resulting from a population depletion of incoming flux. We have developed a hyperfine structure inclusive model to simulate the collision dynamics. Measurement of both the frequency and intensity dependence in conjunction with our model reveal for the first time the interplay between dynamics associated with spontaneous emission, optical shielding, oscillations in Franck-Condon overlap factors, and population depletion. Careful choice of states and laser frequencies allowed the effects of hyperfine structure to be revealed without undue complexity, thus facilitating interpretation of the experimental results.

We acknowledge support from the NSF and the Packard Foundation.

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