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RADIATIVE COLLISIONS IN A STRONG FIELD REGIME

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Résumé - Le processus de transfert d'énergie résonnant, dans les collisions entre deux atomes de Rydberg, représente un point de départ idéal pour l'étude systématique des collisions assistées par photons. En utilisant une source micro-onde courante et de puissance modeste, on atteint sans difficulté le régime de champ fort dans de tels processus.

Abstract - The process of resonant collisional energy transfer between Rydberg atoms provides an ideal starting point for systematic studies of radiatively assisted collisions. In fact using modest, readily available microwave sources it is straightforward to enter the strong field regime for such processes.

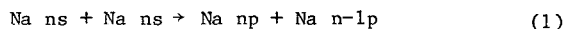
INTRODUCTION

A collision of two atoms in which a photon is absorbed or emitted is frequently called a radiatively assisted collision, although it may also be viewed as emission or absorption of the transient molecule formed in the collision of the two atoms. In either case the short duration of the collision implies that to drive such a transition, i.e. to either stimulate the emission or absorption of a photon, requires an intense radiation field. Thus it is not surprising that the study of such processes has awaited the development of the high power tunable dye laser. In recent years a variety of experimental studies has been undertaken,¹⁻⁵ but in spite of the high power of pulsed tunable dye lasers the time of a collision is so short that nearly all the experiments are in the weak field regime in which the radiation field may be adequately treated by perturbation theory.⁶⁻¹⁰

Only in one instance, in which power densities of 10^9 W/cm² were used, was a less than linear power dependence observed. This saturation marks the onset of the strong field regime in which the radiation field may not be treated by perturbation theory. As it is unrealistic to expect increases of orders of magnitude in dye laser intensities the prospect of exploring the strong field regime for such systems does not appear to be very bright.

RESONANT COLLISIONS OF RYDBERG ATOMS

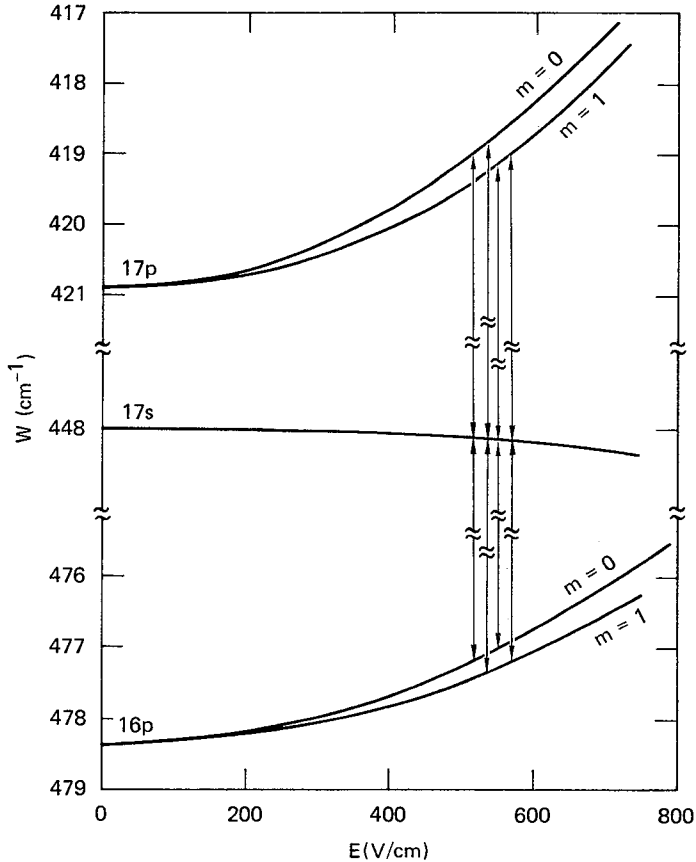
Often a phenomenon which is difficult or impossible to study in normal atoms is easily studied using Rydberg atoms, and the study of radiative collisions is an excellent example of this. Specifically, resonant Rydberg atom-Rydberg atom collisional energy transfer is an ideal system in which to study radiative collisions. Although we have studied a variety of cases¹²⁻¹⁵, the best case for the study of radiative collisions is the resonant process



which occurs when the Na levels are tuned with an electric field so the ns level lies exactly midway between the np and n-1p levels. An example of this is shown in Figure 1, a level diagram for the Na 17s, 16p and 17p states. When the electric

field is at one of the four collisional resonances near 600 V/cm, two Na ns atoms can collide producing a 16p and a 17p atom.

This experiment is done using pulsed lasers to excite Na atoms in a thermal beam to the 17s state. Atoms of different velocities in the beam collide for 1 μ s after the laser pulses at which time an electric field pulse is applied to analyze



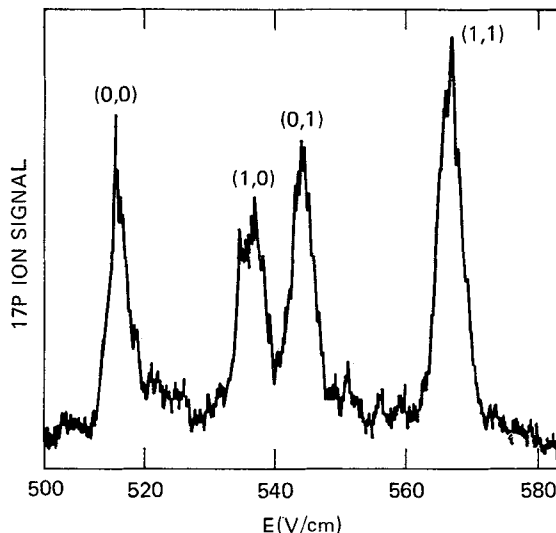
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FIG. 1. Energy-level diagram for the 16p-17s-17p states in a static electric field. The vertical lines are drawn at the four fields where the s state is midway between the two p states and the resonant collisional transfer occurs.

by selective field ionization the products of the collisions which have occurred.¹⁴ If the field ionizing pulse is set to detect the 17p states, we observe four collisional resonances corresponding to the splitting between the $|m|=0$ and 1 levels of the p state as the dc electric field is scanned. An example of such a scan is shown in Figure 2 in which the four collisional resonances are quite evident. As might be anticipated from Eq. 1, this process is a resonant dipole-dipole collision process for which the interaction matrix element H_I is given by¹³

$$H_I = \frac{\langle ns | \mu | np \rangle \langle ns | \mu | n-1p \rangle}{r^3} \quad (2)$$

where μ is the electric dipole moment and r is the internuclear separation of the two atoms. As the dipole moments scale as n^2 , n being the principal quantum number, it is straightforward to show that the cross section σ should scale as n^4 and the collision time T as n^2 (Equivalently the resonant collision linewidth scales as n^{-2}). This is in agreement with the observed n scaling. Typical values, at $n=20$ are $\sigma=10^9 \text{ \AA}^2$ and $T=10^{-9} \text{ s}$.¹³ In sum this process is a textbook example of resonant energy transfer via the long range dipole-dipole interaction. As such, it forms an excellent starting point for more elaborate experiments.



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FIG. 2. The observed 17p ion signal after population of the 17s state vs dc electric field, showing the sharp collisional resonances. The resonances are labeled by the $|m_\lambda|$ values of the lower and upper p states.

RADIATIVE COLLISIONS

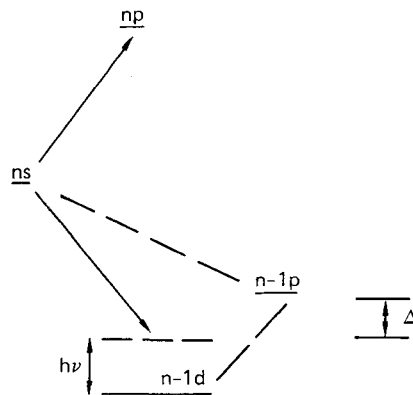
The long collision times of the resonant Rydberg atom collisions and the large dipole moments invite the consideration of radiative collisions using microwave radiation. For such collisions, the interaction matrix element has the form

$$H_I = \frac{\langle ns | \mu | np \rangle \langle ns | \mu | n-1p \rangle \langle n-1p | \mu | n-1d \rangle E}{r^3 \Delta} \quad (3)$$

where Δ is the detuning of the virtual intermediate state from the real state $n-1p$ and E is the magnitude of an applied microwave field. The levels are shown in Figure 3. The expression of Eq. 3 differs from Eq. 2 by the factor $\langle n-1p | \mu | n-1d \rangle E/\Delta$, and for such a radiatively assisted process to be observable, we require that this factor be ~ 0.3 . For typical values of $\Delta = 1 \text{ cm}^{-1}$ and $\mu = 200 e a_0$ we find that $E \sim 100 \text{ V/cm}$, a power of $\sim 1 \text{ W/cm}^2$. The initial experiments verified these estimates, as shown by Figure 4, scans of the resonant collisions of Na 22s atoms with and without microwave fields.¹⁶ As shown by Figure 4, with a microwave field present, an additional four resonances are observed at a field of $\sim 100 \text{ V/cm}$ which correspond to the emission of one photon during the collision. The field separation of the two sets of four resonances corresponds to the 15 GHz microwave frequency.

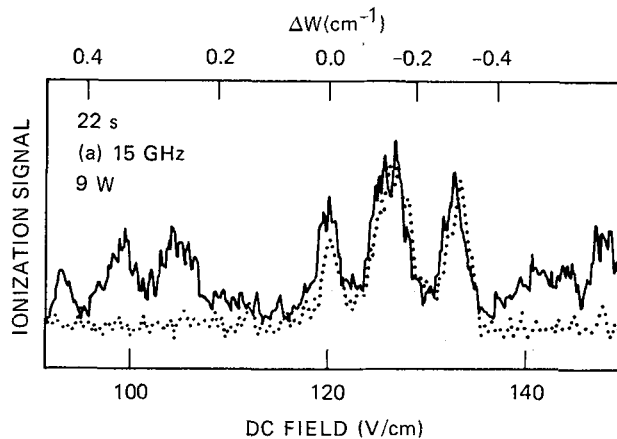
In these experiments, the radiative collision signal scaled approximately linearly with the microwave power, although there was some evidence of saturation at the highest microwave powers used, $\sim 10 \text{ W}$.

As much higher microwave powers can be readily attained it is reasonable to consider strong field experiments. The most straightforward way to do such experiments is to use a microwave cavity which multiplies the effective power by the Q of the cavity. In Figure 5, we show the cavity we have built for such experiments which allows us to apply to the Rydberg atoms a static tuning field, a high voltage



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FIG. 3. The energy levels for the radiative collision process showing the real and virtual levels.



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FIG. 4. The observed 22p ion signal after the population of 22s state vs the dc field in the presence of a microwave field (solid traces) of frequency 15 GHz. The dotted traces were observed with no microwave field, and the sharp resonances in the center (for both solid and dotted traces) result from the resonant collisions with the emission of no photons, while the displaced resonances on the sides (solid trace) are due to microwave-assisted collisions.

field ionization pulse, and a strong microwave field.^{17,18} We recall that since the Na atoms are in a thermal beam, they do not move a macroscopic distance during the experiment, and all three fields must be applied in the same place. Such an approach works quite well. In fact we can readily observe radiatively assisted collisions with 1 mW of microwave power, as shown by Figure 6. Since we have the

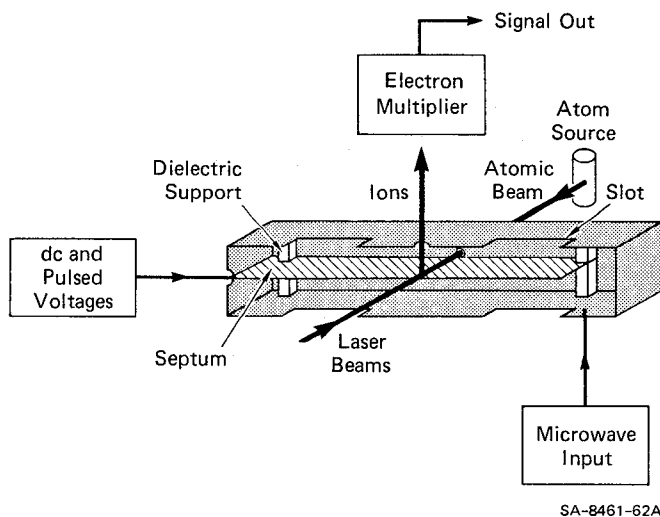


FIG. 5. Cut-away view of the microwave cavity (not to scale). The copper septum bisects the height of the cavity. Two holes of diameter 1.3 mm are drilled in the side walls to admit the collinear laser and Na atomic beams, and 1 mm hole in the top of the cavity allows Na^+ resulting from the field ionization of Na to be extracted.

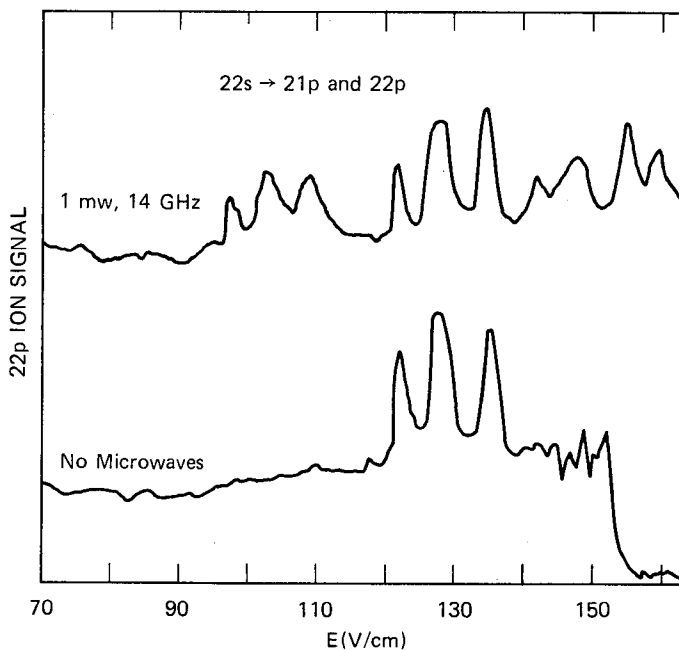
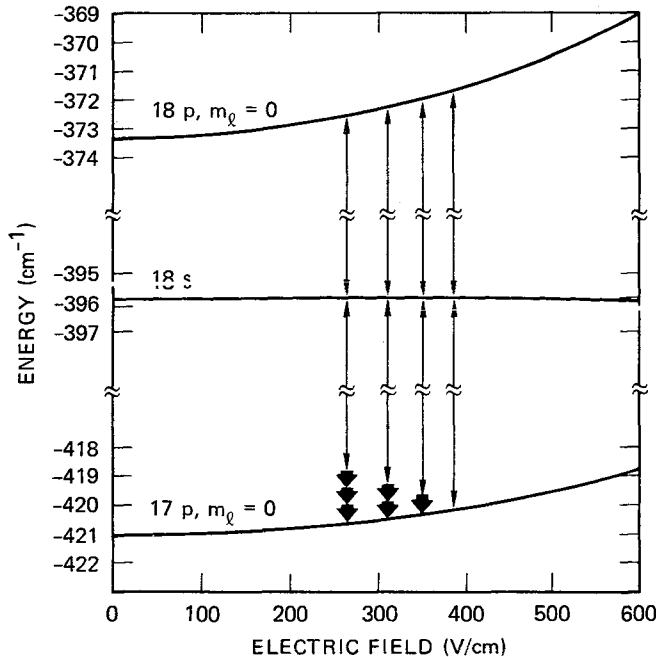


FIG. 6. The process $\text{Na } 22s + \text{Na } 22s \rightarrow 21p + 22p$ with and without 1 mw of 14 GHz microwave radiation. As shown, the cross section for the emission of one photon during the collision is as large as the cross section for the emission of no photons.

capability of putting up to 20 W into our cavity, which has a Q of ~ 2000 , it is evident that we should be able to go well into the strong field regime.

Although we have studied several states, we here concentrate on the $18s + 18s \rightarrow 17p + 18p$ system shown in Figure 7. As shown by Figure 7, it is possible for up to three microwave photons to be emitted in a collision.



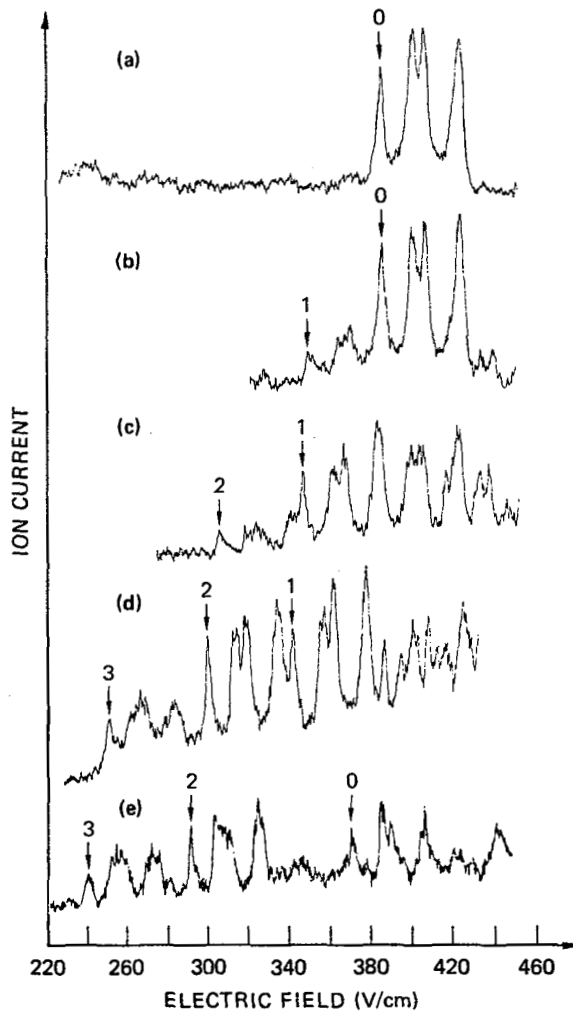
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FIG. 7. Stark energy-level diagram of the $m_l=0$ states, relevant to the multiphoton-assisted radiative collisions. The vertical lines indicate the collisional transfer and are drawn at the fields where they occur. The thick arrows correspond to the emitted photons.

In Figure 8 we show the observed collisional resonances when the Na $18s$ state is populated and the $18p$ state is detected for several microwave input powers as the tuning field is scanned. The microwave frequency is 15.4 GHz.

There are several interesting points to note regarding Figure 2. First up to three photon assisted collisions are observed, at an incident power of 1.2 W. Also at the lowest microwave power for which the n photon process is observed the collisional resonance is broader than at higher power. This presumably reflects the smaller impact parameters and shorter times required. In some sense one can adjust the strength of the coupling during the collision. Finally the zero and one photon processes saturate and actually decrease as the microwave power is increased, an observation that is obviously impossible to explain using a perturbation theory approach.

To describe these data, we have used the low frequency approach of Autler and Townes¹⁹ to describe the alteration of atoms in the strong microwave field and then we consider the resonant dipole-dipole collisions between the altered atoms. The essence of the approach is shown in Figure 9. At a static field of F_s , a microwave



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FIG. 8. The observed ion signal after the population of the $18s$ level versus the microwave field at 15.42 GHz. Trace (a) corresponds to no microwave power input to the cavity and shows the set of four zero photon collisional resonances. Traces (b), (c), (d), and (e) correspond respectively to 0.02, 0.30, 1.20, and 3.00 W of input microwave power and show additional sets of four collisional resonances corresponding to one, two and three photon radiatively assisted collisions. The peaks labelled 0, 1, 2, and 3 correspond to the lowest field member of the set of four resonance corresponding to 0, 1, 2, and 3 photon assisted collisions.

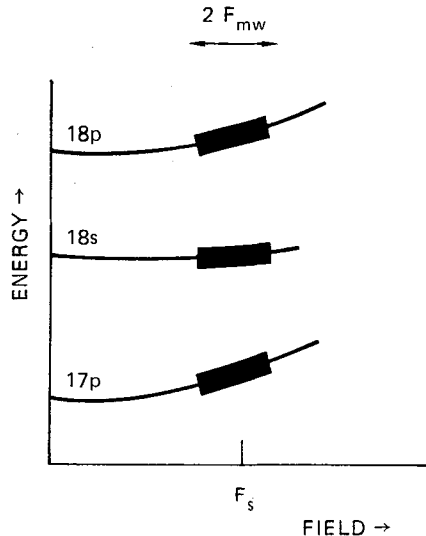
field of amplitude F_{mw} produces a sinusoidal variation in the field and hence in the energy as shown by Figure 9. Thus, the energies of the p states vary approximately as

$$E = E_0 + \alpha/2(F_S + F_{mw} \cos\omega t)^2. \quad (4)$$

Thus the energies are not stationary, but oscillate. The oscillation of the energies may be expressed in terms of a Bessel function expansion. Physically, the effect of the oscillating microwave field is to produce sidebands on each of the 17p and 18p states. The s states are unaffected as might be inferred from Figure 9. Explicitly the wave function of the 17p state $|17p_{mw}\rangle$, for example, is given by

$$|17p_{mw}\rangle = |17p\rangle \sum_i J_i \left(\frac{\partial E}{\partial F} \bigg|_{F_S} \cdot \frac{F_{mw}}{\omega} \right). \quad (5)$$

Here J_i is the Bessel function, E is the energy of the 17p state, F is the total electric field, F_S and F_{mw} are the static and microwave fields, ω is the angular frequency of the microwave field; and $|17p\rangle$ is the wavefunction in the absence of the microwave field. Note that the spatial dependence is not altered. The amplitude of the i th sideband is given by a Bessel function which is an oscillating function of the microwave field.



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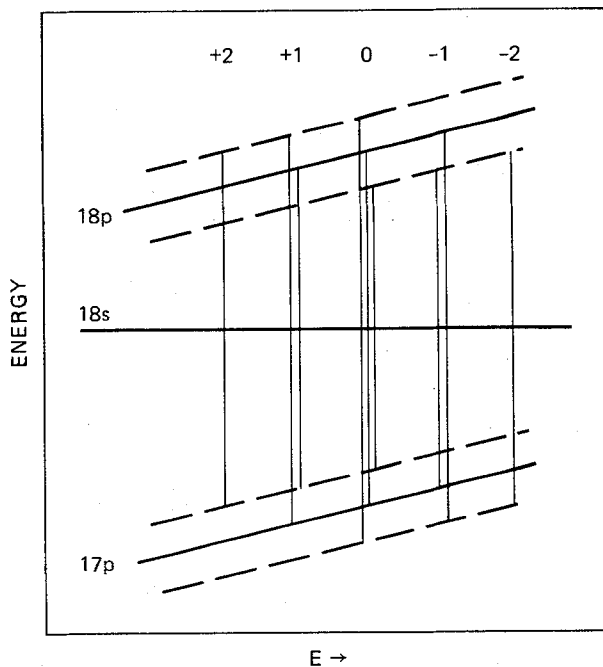
FIG. 9. Simplified energy level diagram of the 17p, 18s, and 18p states. A microwave field of amplitude F_{mw} produces a sinusoidal variation in field and a periodic variation energy as shown by the bold section of the energy level curves.

The energy levels of the states and the first sidebands of the p states as a function of the static tuning field are shown in Figure 3 where the first sidebands of the p states are shown. As shown by Figure 10, processes corresponding to the net emission or absorption of different numbers of photons occur at different fields, however several processes can lead to the emission of the same number of photons. For example, one photon is emitted when either the 17p state and the upper 18p sideband or the 18p state and upper 17p sideband are the final state.

To calculate the cross section we assume that is proportional to the square of the dipole-dipole interaction matrix element

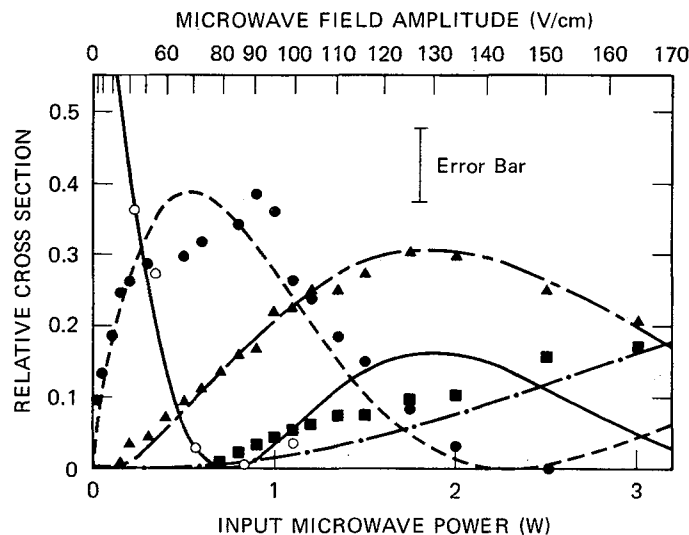
$$\sigma \sim \left| \frac{\langle 18s | \mu | 18p_{mw} \rangle \langle 18s | \mu | 17p_{mw} \rangle}{r^3} \right|^2 \quad (6)$$

From Eq. 6 it is apparent that the microwave power dependence is contained entirely in the product of the 17p and 18p Bessel function expansions given in Eq. 5. Using such a model we are able to reproduce quite accurately the experimental microwave power dependence of the cross section. This is shown in Figure 11 in which we show the observed and calculated power dependence for the various processes. One aspect of this treatment is worth noting. Consider the collisions in which one photon is emitted. It is clear from Figure 10 that three processes are possible, which are implicitly assumed to be coherent in Eq. 6. In fact if the coherence is neglected we find that the calculations do not agree with the experimental observations. For example if the coherence is neglected we would not expect to see the m photon emission collision cross section vanish.



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FIG. 10. Schematic level diagram vs tuning field. The 17p and 18p levels are shown by solid lines with positive Stark shifts and their first upper and lower sideband by broken lines. The vertical lines show the possible resonant collision processes corresponding to the net number of photons emitted indicated at the top of the figure.



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FIG. 11. Experimental cross sections for the m -photon-assisted collisions, as shown by the size of the $(0,0)^m$ resonance peaks, vs input microwave power; zero-photon- (open circle), one-photon- (closed circle), two-photon- (closed triangle), and three-photon- (closed square) assisted collisions. The cross section for a zero-photon collision at zero microwave power (not shown) is normalized to 1. Theoretical cross sections for zero-photon- (solid line), one-photon- (dashed line), two-photon- (long-dashed-short-dashed line), and three-photon- (dot-dashed line) assisted collisions with the same normalization.

These and other recent experiments leave us convinced that radiative collisions and other phenomena involving strong radiation fields may best be studied with Rydberg atoms and microwaves. It is a pleasure to acknowledge stimulating conversations with D. C. Lorents and R. M. Hill in the course of this work. This work was supported by the Air Force Office of Scientific Research under contract F49620-79-C0212.

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REFERENCES

1. R. W. Falcone, W. R. Green, J. C. White, J. F. Young, and S. E. Harris, *Phys. Rev. A* **15**, 1333 (1977).
2. P. H. Cahuzac and P. E. Toschek, *Phys. Rev. Lett.* **40**, 1087 (1978).
3. A. V. Hellfeld, J. Caddick, and J. Weiner, *Phys. Rev. Lett.* **40**, 1369 (1978).
4. W. R. Green, M. D. Wright, J. Lukasik, J. F. Young, and S. E. Harris, *Opt. Lett.* **4**, 265 (1979).

5. J. C. White, *Opt. Lett.* 6, 242, (1981).
6. S. E. Harris and D. B. Lidow, *Phys. Rev. Lett.* 33, 674 (1974).
7. L. I. Gudzenko and S. S. Yakovlenko, *Zh. Eksp. Teor. Fig.* 62, 1686 (1972) [*Sov. Phys JETP* 35, 877 (1972)].
8. S. Geltman, *J. Phys. B* 9, L569 (1976).
9. P. L. Knight, *J. Phys. B* 10, L195 (1977).
10. A. Gallagher and T. Holstein, *Phys. Rev. A* 16, 2413 (1977).
11. J. Lukasik and S. C. Wallace, *Phys. Rev. Lett.* 47, 240 (1981).
12. K. A. Safinya, J. F. Delpech, F. Gounand, W. Sandner, and T. F. Gallagher, *Phys. Rev. Lett.* 47, 405 (1981).
13. T. F. Gallagher, K. A. Safinya, F. Gounand, J. F. Delpech, W. Sandner, and R. Kachru, *Phys. Rev. A* 25, 1905 (1982).
14. T. F. Gallagher, L. M. Humphrey, W. E. Cooke, R. M. Hill, and S. A. Edelstein, *Phys. Rev. A* 25, 1098 (1977).
15. R. Kachru, T. F. Gallagher, F. Gounand, P. Pillet and N. H. Tran, *Phys. Rev. A* 28, 2676 (1983).
16. R. Kachru, N. H. Tran, and T. F. Gallagher, *Phys. Rev. Lett.* 49, 191 (1982).
17. P. Pillet, R. Kachru, N. H. Tran, W. W. Smith, and T. F. Gallagher, *Phys. Rev. Lett.* 50, 1763 (1983).
18. C. H. Townes and A. L. Schawlow, *Microwave Spectroscopy*, (McGraw-Hill, New York, 1955).
19. S. H. Autler and C. H. Townes, *Phys. Rev.* 100, 703 (1955).