# Radiative Collisions in a Strong-Field Regime

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The observation of radiative collisions in the strong-field regime is reported, specifically the multiphoton-microwave-assisted resonant collisional energy-transfer process Na($ns$) + Na($ns$) → Na($np$) + Na(($n-1$)$p$) + $mh\nu$, where $m$ is as high as 4. By use of a "dressed-atom" approach in a low-frequency (or adiabatic) approximation, a theoretical interpretation valid for the strong-field regime is given which is in good agreement with the present experimental results.

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A collision in which an excited atom of one species transfers its excitation to a ground-state atom of a second species with the simultaneous absorption or emission of a photon of energy equal to the difference of the two excited-state energies is frequently called a radiatively assisted collision. The advent of the high-powered tunable dye laser has made it possible to study such laser-assisted processes experimentally,[1-5] and most experiments are in good agreement with a perturbation-theory approach to radiative collisions, valid for the weak-electromagnetic-field regime, which predicts a linear laser-power dependence of the cross section.[6-10] Only at power densities of ~$10^9$ W/cm$^2$ has a less than linear laser-power dependence, indicating the entrance into the "strong-field" regime, been observed.[11]

Recently an experiment analogous to the laser-induced collision experiments has been performed using Na Rydberg atoms and microwave radiation,[12] in which, because of the large dipole moments of the Rydberg atoms, it was possible to observe radiatively assisted collsional energy transfer at microwave power levels of only a few watts per square centimeter. As much higher microwave powers are easily obtained, this observation suggests that it should be possible to reach the strong-field regime for Rydberg atoms interacting with microwaves. Here we present the main features of such an experimental, and accompanying theoretical, study of the microwave-multiphoton-assisted radiative collision process Na($ns$) + Na($ns$) → Na($np$) + Na(($n-1$)$p$) + $mh\nu$, where $m$ ($0 \leq m \leq 4$) microwave photons are emitted during the collision. This process is resonant and occurs when the levels are tuned with a static electric field so that $E_{np} - E_{ns} + mh\nu = E_{ns} - E_{(n-1)p}$, where $E_{nl}$ is the energy of the $nl$ state. This is shown schematically in Fig. 1 for $n=18$. Although we have studied $17 \leq n \leq 22$ we here describe the experiments with $n=18$.

Our experiment is done using an effusive thermal beam of Na with a density of ~ $10^9$/cm$^3$ in a vacuum chamber with a background pressure of < $10^{-6}$ Torr. As shown by Fig. 2 the atomic beam passes through a microwave cavity 1 mm above the septum, where the Na atoms are excited by two 5-ns laser pulses from the ground $3s$ state to the $3p$ state and then from the $3p$ state to the $18s$ state. The laser beams are collinear with the atomic beam but counterpropagating so that a cylinder of Rydberg atoms is produced. In the

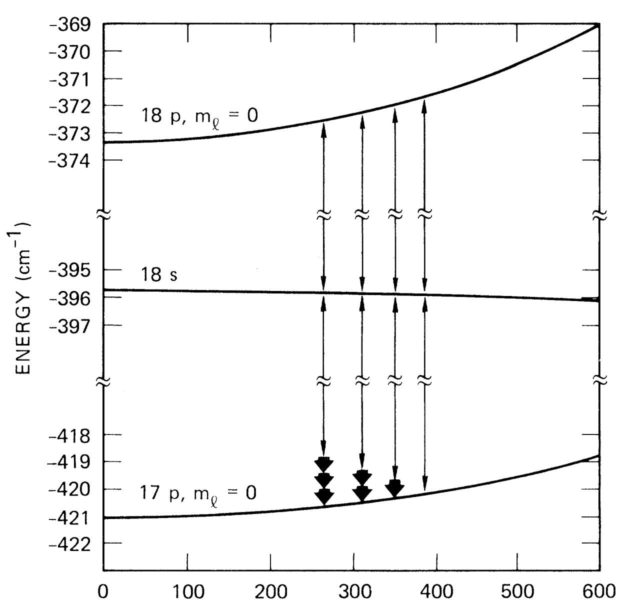


FIG. 1. Stark energy-level diagram of the $m_l = 0$ states, relevant to the multiphoton-assisted radiative collisions. The vertical lines indicate the collisional transfer and are drawn at the fields where they occur. The thick arrows correspond to the emitted photons.

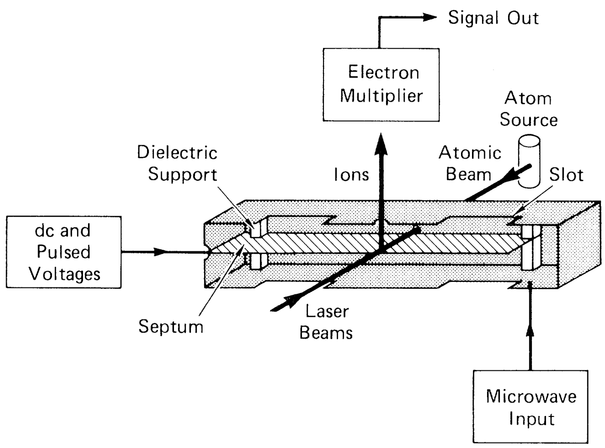


FIG. 2. Cut-away view of the microwave cavity (not to scale). The copper septum bisects the height of the cavity. Two holes of diameter 1.3 mm are drilled in the sidewalls to admit the collinear laser and Na atomic beams, and a 1-mm hole in the top cavity allows $Na^+$ resulting from the field ionization of Na to be extracted.

thermal velocity distribution of the atomic beam the spread in velocities of the Na atoms is $\sim 10^5$ cm/s. Thus the fast 18$s$ atoms overtake the slow 18$s$ atoms and collide with them. Such collisions are allowed to occur for 1.5 $\mu$s following the laser excitation when a positive voltage pulse, the amplitude of which is set to field-ionize selectively atoms in the 18$p$ state,[13] is applied to the septum inside the cavity. The ions thus produced are accelerated through a 1-mm-diam hole in the top of the cavity to a particle multiplier, and the resulting signal is recorded with a gated integrator. As shown in Fig. 1, the electric field required to tune the Na levels is provided by applying a voltage to the septum. As the static field is swept through the values at which the 18$s$ state lies midway between the 18$p$ and 17$p$ states (there are four such values due to the Stark splitting of $|m_l|=0$ and $|m_l|=1$ 17$p$ and 18$p$ states) we see four sharp increases in the ion signal, as shown in Fig. 3(a), corresponding to the resonant collisions of pairs of Na 18$s$ atoms to form one 18$p$ and one 17$p$ atom.[14] As expected, the size of the resonant collision signal varies as the square of the number of 18$s$ atoms excited (in the limit of a small fraction of the atoms undergoing collisions). Collisions also occur with background-gas molecules and ground-state Na atoms, but the cross sections for such processes are too small to be observable here.[15]

The microwave cavity is a piece of $X$-band (RG-52/U) waveguide 20.32 cm long, closed at each end, which we can excite at frequencies of 12.79, 14.07, and 15.42 GHz corresponding to the $TE_{10n}$ modes $n=15$, 17, and 19. Each of these modes has an antinode in the center of the cavity. Since the Na atoms travel only 1 mm in the 1.5-$\mu$s interval after the laser pulses, those atoms which we ultimately detect are those which have been at the antinode of the microwave field. The cavity has a $Q$ of 1400 and is excited with up to 4 W of microwave power. At 15.42 GHz, 1 W produces a microwave field amplitude of 95 V/cm ($\pm$ 10%) in the center of the cavity. The microwave field is perpendicular to the septum.

If a microwave field is present while the Na 18$s$ atoms collide, we observe the radiatively assisted collisions as the static field is swept through the values at which the $m$-photon-assisted collisions occur. In Figs. 3(b)–3(e) we show typical signals obtained by sweeping the static field for several microwave powers at a frequency of 15.42 GHz. As mentioned earlier, Fig. 3(a) corresponds to the case without microwaves ($m=0$) and shows the four zero-photon resonant collision peaks. As the microwave power is increased, Figs. 3(b), 3(c), and 3(d) each show in succession the appearance of sets of four new peaks corresponding respectively to the one-, two-, and three-microwave-photon-assisted resonant collisional energy transfers as shown in Fig. 1. For the $m$-photon-assisted collisions we label the four collisional resonances by the $|m_l|$ values of the final lower, then upper, $p$ states, respectively, with $m$ as a superscript. For instance, in order of increasing field we denote the four one-photon resonances as $(0,0)^1$, $(1,0)^1$, $(0,1)^1$, and $(1,1)^1$.

In Fig. 3 we indicate the $(0,0)^m$ resonance by the number $m$ and an arrow. Several features of these signals are worth noting. First, for all the resonances we observed shifts proportional to the microwave power $W_I$ driving the cavity. Second, note that in Fig. 3(b) the one-photon-assisted collisional resonance at 348 V/cm is weak, broad, and very asymmetric, while at the higher power of Fig. 3(c) it is narrower and stronger. This occurs because the radiative collision interaction strength depends on the microwave power and is weaker in Fig. 3(b) than 3(c) so that the collision requires a smaller impact parameter leading to the observed results.[16,17] Figure 4 shows the experimental variations of the maximum ion signal of the resonances $(0,0)^m$ versus the incident microwave power $W_I$. Note that the maximum cross sections of these different processes are of the

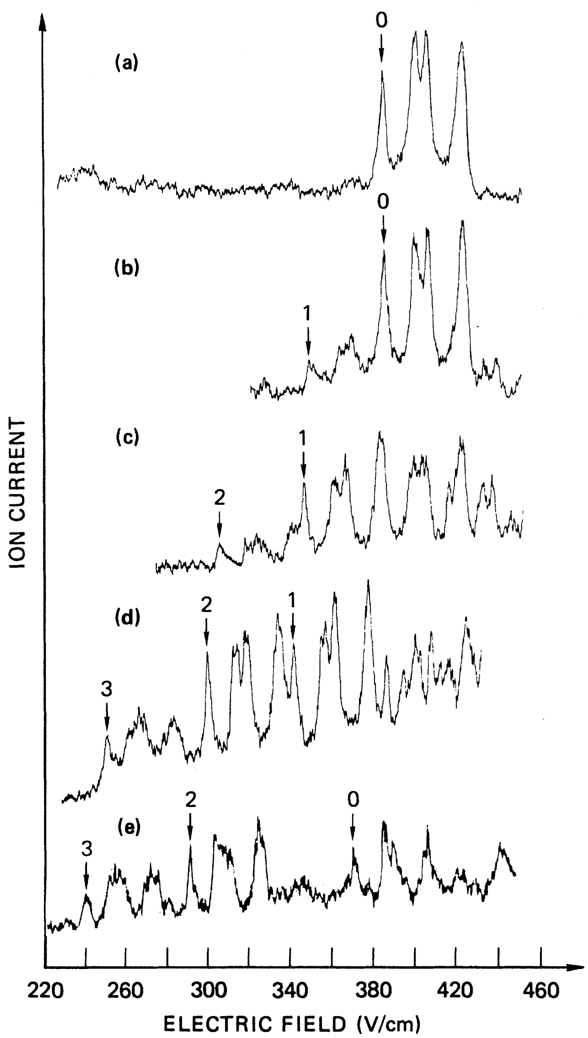


FIG. 3. Observed ion signal after the population of the 18$s$ level vs the microwave field at 15.42 GHz. Trace $a$ corresponds to no microwave power input to the cavity and shows the set of four zero-photon collisional resonances. Traces $b$, $c$, $d$, and $e$ correspond respectively to 0.02, 0.30, 1.20, 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 labeled 0, 1, 2, and 3 correspond to the lowest-field member of the set of four resonance corresponding to zero-, one-, two-, and three-photon-assisted collisions, $(0,0)^0$, $(0,0)^1$, $(0,0)^2$, and $(0,0)^3$.

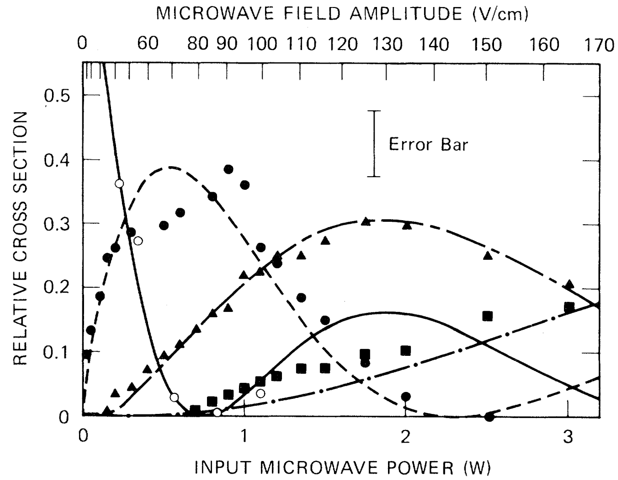


FIG. 4. Experimental cross sections for the $m$-photon-assisted collisions, as shown by the size of the $(0,0)^m$ resonance peaks, vs input microwave power; zero-photon- (open circle), one-photon- (closed circle), two-photon- (closed triangle), and three-photon- (closed square) assisted collisions. The 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.

same order of magnitude ($\sim 10^9$ Å$^2$) as the zero-photon dipole-dipole process.[14]

As shown by Figs. 3 and 4 the signal for the one- and two-photon-assisted collisions quickly becomes saturated and, in fact, as the power is increased to greater than 2 W the one-photon-assisted collision signal disappears. To our knowledge such a decrease in a radiatively assisted collision has not been previously observed.

To interpret these results we have developed a theory of radiative collisions, based on the work of Autler and Townes,[18] which is valid in a strong-field regime. First we assume that the microwave frequency is small compared to the separation between the adjacent levels, implying that the Na atom follows the microwave field adiabatically and transitions to other levels can be neglected. For frequencies of 15 GHz and $n \sim 20$ this hypothesis is valid for the $ns$ and $np$ levels of the Na as long as we are far enough from the crossings of these levels with other levels.[19] Under these conditions we can treat first the isolated atom "dressed" by the microwave field and then, as the duration of one collision ($\sim$1 ns) is long compared to a microwave period, the collisions of these dressed atoms as resonant dipole-dipole collisions.[14]

The essence of the dressed-atom approach is as follows. Since the temporal variation of the field is periodic, at frequency $\omega/2\pi$, the original level will now have sidebands spaced in energy by $\hbar\omega$.[18] For the case in which the microwave field $F_{\rm mw}$ is small compared to the static field $F_s$ the amplitude of the $i$th (including zero) sideband of state $A$ is given by[18]

$$A_i = J_i\left(\frac{dE}{dF}\bigg|_{F_s} x \frac{F_{\mathrm{mw}}}{\omega}\right),$$

where $J_i$ is the Bessel function and $dE/dF|_{F_s}$ is the derivative of the energy of state $A$ with respect to field evaluated at $F_s$. We note that this theory takes into account all other levels by means of the derivative $dE/dF|_{F_s}$. Note that only the $p$ levels will acquire significant sideband amplitudes as the $s$ states have negligible Stark shifts.

Having determined the amplitudes of each of the sidebands we calculate the cross sections for the resonant dipole-dipole collision process between the different sidebands.[20] In Fig. 4, we show the calculated variations of the cross sections of the different $m$-photon processes versus the microwave power. Recalling that $m$ equals the net number of photons emitted it is clear that processes involving several different combinations of sidebands lead to the same net number of photons emitted. The fact that the zero- and one-photon-assisted collisions' signals completely disappear as shown in Fig. 4 is an experimental indication of the coherence of the processes involving different sidebands. Our calculations support this notion and show, for example, that the disappearance of the zero-photon-assisted collision signal at $W_I = 0.8$ W is due to destructive interference between the collisional process $\mathrm{Na}(ns(0) + ns(0) \to np(0) + (n-1)p(0))$ and $\mathrm{Na}(ns(0) + ns(0) \to np(\pm 1) + (n-1)p(\mp 1))$ (0, +1, and −1 indicate the sideband considered). The cross section increases at higher power when the processes $\mathrm{Na}(ns(0) + ns(0) \to np(\pm 1) + (n-1)p(\mp 1))$ are dominant. We note that the theoretical results of Fig. 4 are in good agreement with the experimental observations.

These first observations of radiatively assisted collisions in the strong-field regime make it clear that Rydberg atoms and microwaves are in fact an ideal system for the quantitative study of such effects.

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[1]R. W. Falcone, W. R. Green, J. C. White, J. F. Young, and S. E. Harris, Phys. Rev. A 15, 1333 (1977).

[2]Ph. 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. Fiz. 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]R. Kachru, N. H. Tran, and T. F. Gallagher, Phys. Rev. Lett. 49, 191 (1982).

[13]T. F. Gallagher, L. M. Humphrey, W. E. Cooke, R. M. Hill, and S. A. Edelstein, Phys. Rev. A 16, 1098 (1977).

[14]T. F. Gallagher, K. A. Safinya, F. Gounand, J. F. Delpech, W. Sandner, and R. Kachru, Phys. Rev. A 25, 1905 (1982).

[15]T. F. Gallagher and W. E. Cooke, Phys. Rev. Lett. 42, 835 (1979).

[16]S. E. Harris and J. C. White, IEEE J. Quantum Electron. 13, 972 (1977).

[17]C. Brechignac, Ph. Cahuzac, and P. E. Toschek, Phys. Rev. A 21, 1969 (1980).

[18]S. H. Autler and C. H. Townes, Phys. Rev. 100, 703 (1955).

[19]P. Pillet, W. W. Smith, R. Kachru, N. H. Tran, and T. F. Gallagher, Phys. Rev. Lett. 50, 1042 (1983).

[20]P. Pillet, R. Kachru, N. H. Tran, and T. F. Gallagher, unpublished.