Measurement of Large Cross Sections for Laser-Induced Collisions

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We report measurements of cross sections greater than 10^{-13} cm^2 for laser-induced inelastic collisions between Sr and Ca. We describe an experimental system which produces such large cross sections and which greatly facilitates the study of laser-induced collisions through its ability to reduce noise from other collisional processes.

A laser-induced or "switched" collision is a process in which a laser field is used to induce selective energy transfer from a level in one atomic or molecular species to another level in a different species. This phenomenon has attracted considerable experimental and theoretical attention in recent years.[1-12] The induced collision cross section rises as the laser power density increases and it has been predicted that very large collision cross sections can be obtained.[1,2] The largest cross section reported in past experiments is about 10^{-16} cm^2.[3,4] In this Letter we report measurements of laser-induced collision cross sections exceeding 10^{-13} cm^2. These large cross sections were attained by using 40-psec laser pulses which provided high peak-power densities at reasonable pulse energies. In addition, the short-duration pulses which we employed induced the energy transfer in a time short compared to other collisional processes which also populate the final state.

The two laser-induced processes which are reported in this article are described by

$$\mathrm{Sr}(5s5p\ {}^1P_1^\circ) + \mathrm{Ca}(4s^2\ {}^1S_0) + \hbar\omega \rightarrow \mathrm{Sr}(5s^2\ {}^1S_0) + \mathrm{Ca}(4p^2\ {}^1S_0), \quad (1a)$$

$$\mathrm{Ca}(4s4p\ {}^1P_1^\circ) + \mathrm{Sr}(5s^2\ {}^1S_0) + \hbar\omega \rightarrow \mathrm{Ca}(4s^2\ {}^1S_0) + \mathrm{Sr}(5s6d\ {}^1D_2). \quad (1b)$$

Cross sections of 10^{-17} cm^2 for the first process have been previously reported[3]; an energy level diagram is shown in Fig. 1. The second switched collision process is reported here for the first time.

The experimental setup employed an actively mode-locked Nd:YAlG (neodymium:yttrium aluminum garnet) oscillator-amplifier system.[13] The output of this system was doubled and mixed in deuterated potassium dihydrogen phosphate crystals to produce a train of 3547-Å pulses. This pulse train was split and excited two synchronously pumped dye lasers. Each dye laser was cavity dumped to produce a 40-psec pulse of several megawatts peak power. In the process of Eq. (1a) the output of one laser, the pump laser, was used to populate the Sr($5s5p\ {}^1P_1^\circ$) level while in Eq. (1b) the Ca($4s4p\ {}^1P_1^\circ$) level was populated. The transfer laser, which induced the switched collision process, was delayed by 5 nsec from the pump laser. Both beams were focused into a metal vapor cell to an area of 10^{-4} cm^2. The cell was heated to 800°C providing ground-state number densities near 10^{16} atoms/cm^3.

The fluorescence from the target state was imaged into a 1-m spectrometer and photomultiplier and was recorded as a function of the wavelength of the transfer laser. The signal from the photomultiplier was sampled over four consecutive 8-nsec time intervals by a set of gated integrators. For each pulse the outputs of the gated integrators were recorded by a minicomputer, and the raw data averaged at a later time.

For the laser-induced collision in Eq. (1a) the cross section was determined experimentally from the relation

$$\sigma_{\mathrm{expt}} = (N_{\mathrm{Ca}}^*/N_{\mathrm{Sr}}^*)(1/N_{\mathrm{Ca}}\bar{V}\tau),$$

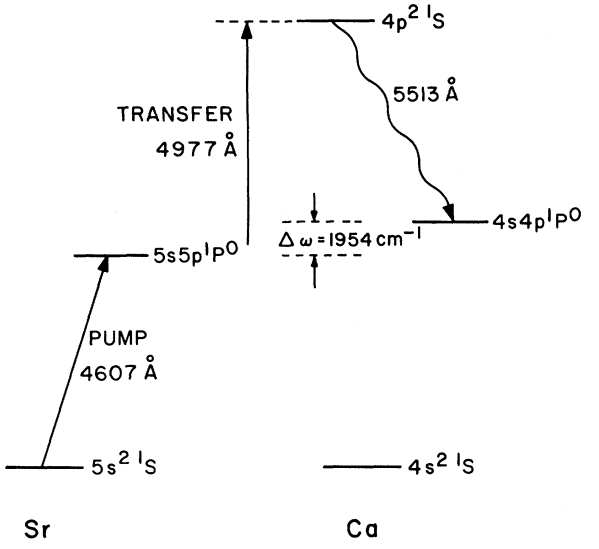

FIG. 1. Energy level diagram for $\mathrm{Sr}^* + \mathrm{Ca} + \hbar\omega \rightarrow \mathrm{Sr} + \mathrm{Ca}^*$ switched collision process.

where $\bar{V}$ is the relative velocity and τ is the pulse width. The ground-state number density of calcium N_{Ca} was determined by an absorption scan using the curve-of-growth method.[14] The initial stored population $N_{Sr}{}^*$ was determined by applying a laser with a wavelength of 4956 Å to saturate the $Sr(5s5p\ ^1P_1{}^\circ)$-$Sr(5s7d\ ^3D_2)$ transition, and subsequently observing the fluorescence at 3653 Å [$Sr(5s7d\ ^3D_2$-$Sr(5s5p\ ^3P_1{}^\circ)$]. By taking the ratio of this fluorescence to the fluorescence from the final state in calcium, the experimental determination of the cross section is made independent of the collection efficiency.

The experimental results for this system are summarized in Fig. 2 which also includes data previously taken using 1-μsec-pulse lasers.[3]

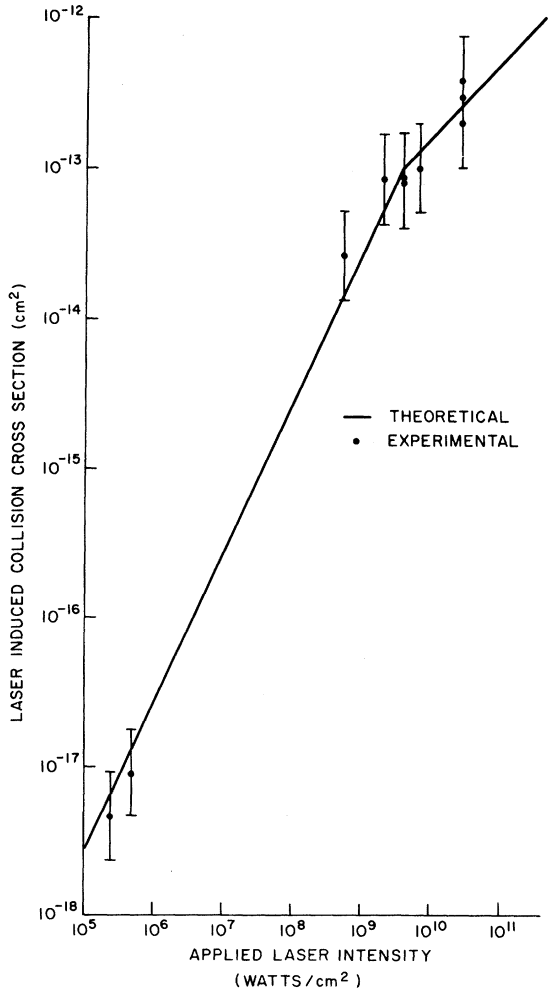

FIG. 2. Laser-induced collision cross section as a function of applied laser power density, $Sr^* + Ca + \hbar\omega \rightarrow Sr + Ca^*$.

The solid line represents the theoretical cross section as a function of power density; in the weak-field regime it is given by[1,2,5]

$$\sigma_{theor} = 1.4\,\frac{4\pi}{3}\left(\frac{\mu_{21}{}^{Sr}\mu_{12}{}^{Ca}\mu_{23}{}^{Ca}E}{\hbar^2\rho_0\bar{V}\Delta\omega}\right)^2,$$

while in the strong-field regime it is given by[1,15]

$$\sigma_{theor} = 1.4\,\frac{3\pi}{\sqrt{3}}\,\frac{\mu_{21}{}^{Sr}\mu_{12}{}^{Ca}\mu_{23}{}^{Ca}E}{\hbar^2\bar{V}\Delta\omega},$$

where ρ_0 is the effective Weisskopf radius for the system and $\Delta\omega$ is the relevant detuning. As shown in the figure, at power densities near 10^{10} W/cm^2 we observed cross sections in excess of 10^{-13} cm^2. At power densities above 10^{10} W/cm^2, significant ionization of the excited states can occur, thereby increasing the error in the measured cross section.

In Fig. 3 we show the fluorescence from the $Ca(4p^2\ ^1S_0)$ state in the process of Eq. (1a) as a function of the transfer laser wavelength. In Fig. 3(a) the 8-nsec gated integrator was coincident in time with the transfer laser pulse, while in Fig. 3(b) the gated integrator was delayed by 24 nsec from the transfer laser pulse. The peak of the laser-induced transfer is indicated by the arrow and occurs exactly at the wavelength corresponding to the energy difference of the storage and target states of the infinitely separated atoms, $\lambda_{R=\infty}$. The other lines in these traces result from transitions in one species followed by collisional transfer into the final state of Ca. In the experiments reported several years ago which used microsecond pulse lasers,[3] the signal which resulted from such collision processes exceeded that of the laser-induced collision by about 500 times. As seen, the experimental setup described here increased the relative magnitude of the laser-induced collision by more than 100.

The importance of the ability to time gate is illustrated by the scan of Fig. 3(b); in this case the 8-nsec gate was delayed 24 nsec relative to the transfer laser pulse. It can be seen that all of the lines which resulted from collisional diffusion into the final state increased dramatically relative to scan (a) while the switched collision signal decreased significantly. This is because the switched collision process transferred only during the 40-psec laser pulse while the other collisional processes continued to transfer into the final state until their respective excited states were depleted. [Since the lifetime of the $Ca(4p^2\ ^1S_0)$ state is ~ 15 nsec, some fluorescence from the switched collision process is still apparent in the

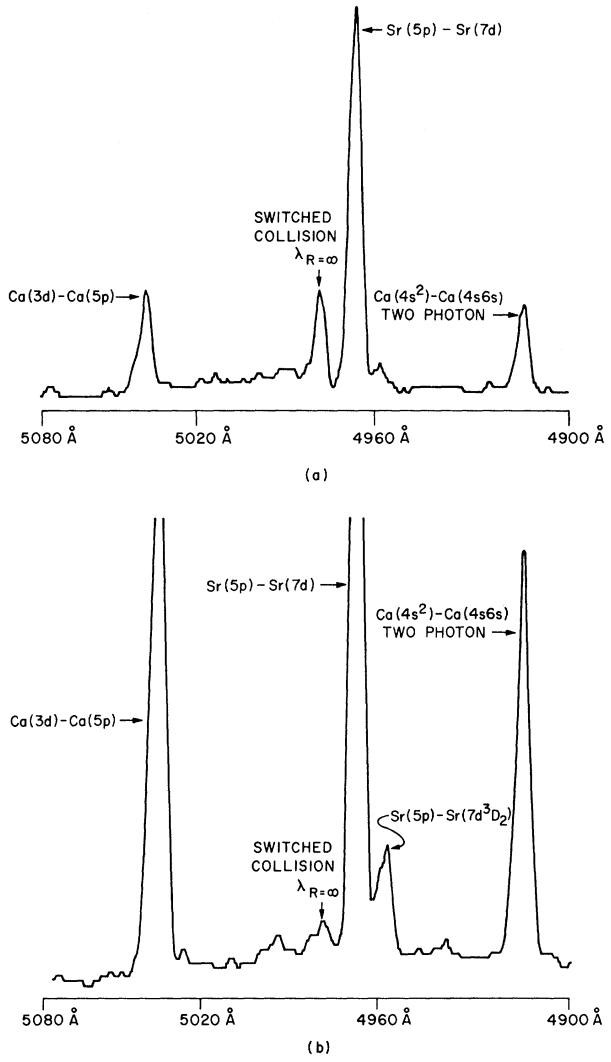

FIG. 3. Final-state [Ca($4p^2\ ^1S_0$)] fluorescence as a function of transfer laser wavelength. (a) Observation time interval coincident with transfer laser pulse. The ratio of the cross sections of the switched collision and the other collisional processes is 200 times greater than that indicated because the switched collision occurs for 40 psec while the other processes occur over the entire 8-nsec gate width. (b) Observation time interval delayed 24 nsec from transfer laser pulse.

24-nsec delayed scan.] We emphasize that these are integrated scans and that since the switched collision only occurred during a 40-psec pulse, its transfer rate is actually 200 times greater relative to the other lines shown.

A study of the process of Eq. (1b) yielded similar results. The line shape for this process again centered at the frequency difference between the storage and target state of the infinitely separated atoms, $\lambda_{R=\infty} = 6217$ Å. Since the fluorescent lifetime of the final target state in this process was about 10 times longer than that of the process of Eq. (1a), the observed signal intensity, integrated for the fixed gate time, was correspondingly smaller. The cross section was estimated to be 10^{-14} cm^2 for a power density of 3×10^9 W/cm^2, assuming that the storage-state density was about the same as in the process of Eq. (1a).

The ability to excite designated target states of selected species may be of importance for photochemical and spectroscopic studies, as well as for the construction of new types of short-wavelength lasers. We have demonstrated laser-induced cross sections about 1000 times larger than those previously reported. The application of a picosecond laser system, together with fast gated integration, dramatically enhances the ability to discriminate between laser-induced and normal collisional processes.

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