

# Observation of a laser-induced dipole-quadrupole collision

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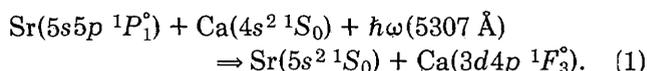
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Experimental observation of a laser-induced dipole-quadrupole collision between Sr and Ca is reported. The laser-induced collision cross section peaks at an applied photon energy equal to the energy defect of the initial and final states of the infinitely separated atoms. A cross section of  $3 \times 10^{-14} \text{ cm}^2$  was induced at a laser-power density of  $7 \times 10^9 \text{ W/cm}^2$ ; the linewidth was  $20 \text{ cm}^{-1}$ .

This is the first reported observation of a laser-induced dipole-quadrupole inelastic collision process. In general, laser-induced collisions make use of an intense laser field to transfer energy from a storage level of one atomic species to a particular target level of a second species during a collision, and they may be conveniently classified according to the nature of the collision coupling involved. In the previously studied dipole-dipole processes,<sup>1-12</sup> one species makes a parity-allowed transition while the other makes a parity-nonallowed transition. In the dipole-quadrupole process, both species make parity-allowed or -nonallowed transitions. As will be shown below, the two processes have similar line shape characteristics and can have comparable magnitude. Thus, in combination, they permit one to transfer energy selectively from a storage state of arbitrary parity to a target state of arbitrary parity.

The process studied is described by



An energy-level diagram is shown in Fig. 1. Energy is initially stored in the Sr( $5s5p \ ^1P_1^{\circ}$ ) level. As the atoms collide, the Sr atom makes a dipole transition from its excited state to ground, while the Ca atom undergoes a quadrupole transition to a virtual level of Ca( $4s3d \ ^1D_2$ ) character. The applied laser field then induces a dipole transition from the virtual level to the Ca( $3d4p \ ^1F_3^{\circ}$ ) target state. Theory predicts that this process maximizes when the applied photon energy is equal to the energy difference of the initial and final states when the atoms are fully separated (as in the case of laser-induced dipole-dipole collisions).

The experimental investigation of the laser-induced dipole-quadrupole collision employed two synchronously pumped dye lasers. The dye lasers were pumped by 3547-Å radiation, which was created by upconversion of the 1.06-μm radiation from an actively mode-locked Nd:YAG oscillator-amplifier system. Each dye laser was cavity pumped to produce a 40-psec pulse of tunable radiation of several megawatts peak power. The output of one dye laser, the pump laser, was used to prepare the Sr( $5s5p \ ^1P_1^{\circ}$ ) state, while the output of the second laser, the transfer laser, was used to induce the collision process. The transfer laser was delayed 5 nsec from the pump laser, and both beams

were focused into a metal vapor cell to an area of  $\sim 5 \times 10^{-4} \text{ cm}^2$ . The cell was heated to 900°C, providing ground-state densities of  $\sim 10^{16} \text{ atoms/cm}^3$ . The cell also contained a background pressure of 15 Torr of Ar to protect the cell windows.

The fluorescence at 5349 Å from the target state in Ca was imaged into a 1-m spectrometer equipped with a photomultiplier at the exit slit. The signal from the photomultiplier was processed by a gated integrator, which was gated on for 18 nsec, and the integrated signal was recorded as a function of the transfer-laser wavelength. Figure 2 shows this recorded signal. Within experimental accuracy, the laser-induced dipole-quadrupole collision process maximizes at an applied photon energy equal to the energy difference of the initial and final states of the separated atoms. The linewidth of the laser-induced collision process is  $20 \text{ cm}^{-1}$ . The two other lines shown in Fig. 2 result from transitions in Sr followed by collisional transfer into the target state of Ca. Note that the ratio of the rate of production of target-state Ca atoms by the laser-induced collision process to the production rate by the other collisional processes is 450 times larger than that indicated by Fig. 2 since the laser-induced collision

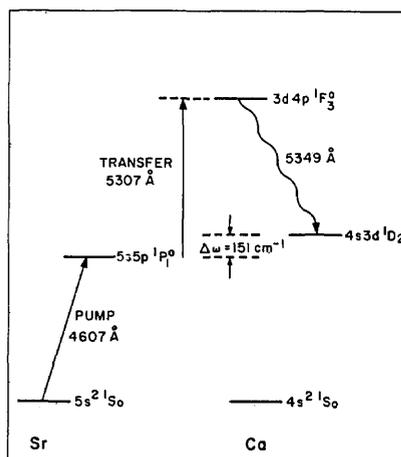


Fig. 1. Energy-level diagram for the laser-induced dipole-quadrupole collision process  $\text{Sr}(5s5p \ ^1P_1^{\circ}) + \text{Ca}(4s^2 \ ^1S_0) + \hbar\omega \rightarrow \text{Sr}(5s^2 \ ^1S_0) + \text{Ca}(3d4p \ ^1F_3^{\circ})$ .

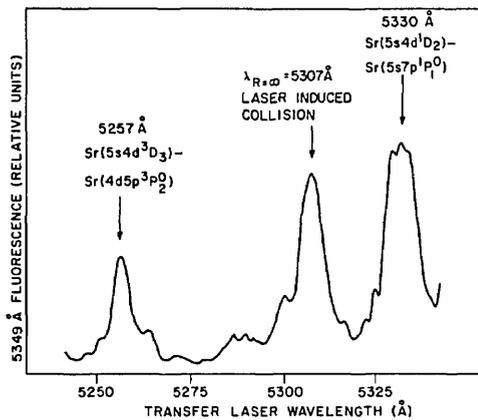


Fig. 2. Fluorescence from the  $\text{Ca}(3d4p\ ^1P_1^o)$  target state integrated over the 18-nsec time gate as a function of transfer-laser wavelength.

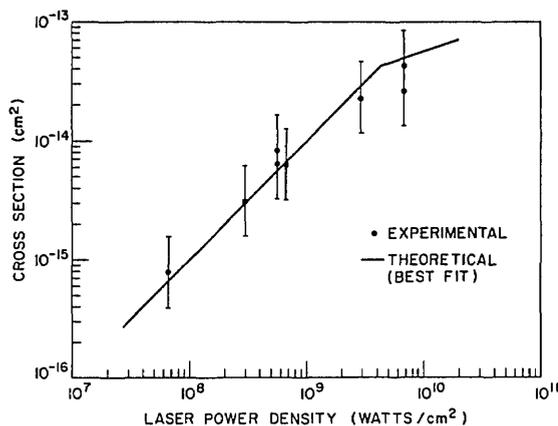


Fig. 3. Cross section for the laser-induced dipole-quadrupole collision as a function of laser-power density.

process occurs for 40 psec, whereas the collisional diffusion processes occur during the entire 18-nsec gate width.

Additional measurements that were made to confirm our interpretation of the peak at 5307 Å include the following: (1) The pump laser was detuned 5 Å from the  $\text{Sr}(5s^2\ ^1S_0)\text{-Sr}(5s5p\ ^1P_1)$  transition to eliminate the  $\text{Sr}(5s5p\ ^1P_1)$  population. Under such pumping conditions, the signal at 5307 Å did not appear. (2) The pump laser was tuned to the  $\text{Ca}(4s^2\ ^1S_0)\text{-Ca}(4s3d\ ^1D_2)$  transition, thereby creating a large  $\text{Ca}(4s3d\ ^1D_2)$  population. Again the signal at 5307 Å was absent.

Measured values of the cross section for the laser-induced collision process were determined from the following relation:

$$\sigma_{\text{expt}} = (N_{\text{Ca}^*}/N_{\text{Sr}^*})(1/N_{\text{Ca}}\bar{V}\tau), \quad (2)$$

where  $\tau$  is the pulse duration of the transfer laser. The ground-state Ca number density,  $N_{\text{Ca}}$ , was determined by a white-light absorption scan, and the excited-state  $\text{Sr}(5s5p\ ^1P_1)$  number density,  $N_{\text{Sr}^*}$ , was determined by tuning the transfer-laser wavelength to 5556 Å to saturate the  $\text{Sr}(5s5p\ ^1P_1)\text{-Sr}(5s6d\ ^3D_2)$  transition and then observing the fluorescence at 3941 Å resulting from the  $\text{Sr}(5s6d\ ^3D_1)\text{-Sr}(5s5p\ ^3P_0^o)$  transition. The experimental value of the cross section was obtained by

taking the ratio of the fluorescence at 5349 Å from the target state in Ca to the fluorescence at 3941 Å, thus eliminating a calibration of the collection efficiency of analyzing optics.

Measured values of the collision cross section as a function of applied laser-power density are shown in Fig. 3. A cross section of  $3 \times 10^{-14} \text{ cm}^2$  was obtained at a laser-power density of  $7 \times 10^9 \text{ W/cm}^2$ . The theoretical expression for the cross section of the laser-induced dipole-quadrupole collision is, in the weak field regime, given by

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

whereas in the strong field regime it is given by

$$\sigma_{\text{theor}} = \pi \left[ \left( \frac{\pi}{\hbar\bar{V}} \right) \left( \frac{3\mu_{21}^{\text{Sr}}q_{12}^{\text{Ca}}}{2} \right) \left( \frac{\mu_{23}^{\text{Ca}}E}{2\hbar\Delta\omega} \right) \right]^{2/3}. \quad (4)$$

In these formulas,  $q_{12}^{\text{Ca}}$  is the quadrupole matrix element and  $\rho_0$  is the Weisskopf radius.<sup>3</sup> The weak field formula is derived by assuming that only collisions that occur with an impact parameter greater than the Weisskopf radius contribute to the cross section. The strong field regime begins when the laser-power density is increased to the point at which the probability of transfer becomes unity for an impact parameter equal to the Weisskopf radius. Above this value of laser-power density the collision cross section only increases as the cube root of the applied laser-power density. The solid line of Fig. 3 represents the best-fit theoretical cross section obtained from Eqs. (3) and (4) by adjusting the value of the quadrupole matrix element  $q_{12}^{\text{Ca}}$ . The resulting best-fit value is  $2.7 \times 10^{-26} \text{ esu}\cdot\text{cm}^2$ . To the best of our knowledge, the matrix element has not been measured, but our inferred value is about one third as large as the quadrupole matrix element obtained from the known line strength of the same electronic transition in the Ca ion.<sup>13</sup>

These experiments have shown that laser-induced dipole-quadrupole collision cross sections can be large and thus permit one to consider practical collision systems having storage and target states of different parity from those that are accessible by using dipole-dipole interactions.

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