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Inelastic Collision Induced by Intense Optical Radiation*

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A large cross section for inelastic collision is induced by an incident laser tuned to the frequency of the interatomic energy defect. We study energy transfer from the Sr $5p\ ^1P°$ level to the Ca $4d\ ^1D$ level and measure a cross section for inelastic collision of 3×10^{-16} cm^2 at a laser power density of 8.6×10^6 W/cm^2 and a wavelength of 6409.0 Å.

The cross section for inelastic collision between atoms is infinitesimally small if the energy defect ΔE is large with respect to kT. In this Letter we report the first observation of a process where a large cross section for inelastic collision is created by applying optical radiation at a frequency $\hbar\omega = \Delta E$. Energy transfer is initiated or "switched" by the presence of the optical radiation. Inelastic collision processes of this type have recently been predicted by Gudzenko and Yakovlenko[1] and by Harris and Lidow.[2]

We have observed this process in the system (Fig. 1)

$$\mathrm{Sr}(5p\ ^1P°) + \mathrm{Ca}(4s^2\ ^1S) + \hbar\omega(6409\ \text{Å}) = \mathrm{Sr}(5s^2\ ^1S) + \mathrm{Ca}(4d\ ^1D). \qquad (1)$$

Energy was first stored in the radiatively trapped $5p\ ^1P°$ level of Sr I. This level was populated by two-photon pumping of the $5d\ ^1D$ Sr level, followed by radiative decay. Inelastic collision to the $4d\ ^1D$ level of Ca I was effected by a second laser beam at 6409 Å.

During collision of an excited Sr $5p\ ^1P°$ atom and a ground-state Ca $4s^2\ ^1S$ atom the strong dipole-dipole coupling of the $5p$-$5s$ Sr transition and the $4p$-$4s$ Ca transition causes a virtual transition of the Ca atom. Absorption of a 6409-Å

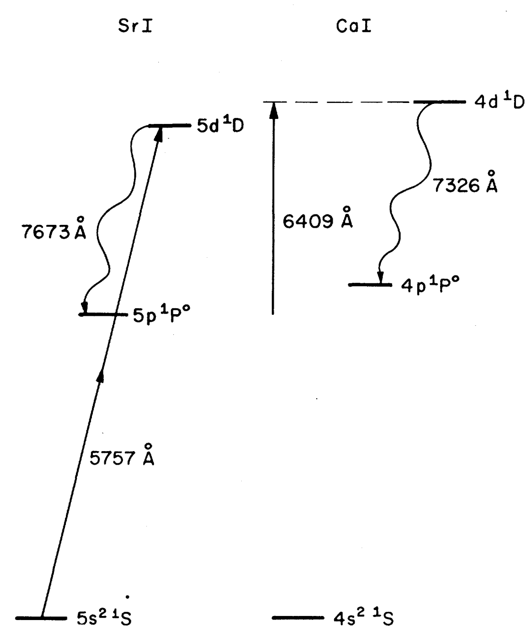

FIG. 1. Energy level diagram for Sr-Ca induced-collision experiment.

photon completes the Ca excitation. The process may thus be viewed as a virtual collisional excitation, followed by a real absorption.[2] Alternatively, and for the case described here equivalently, the process can be viewed as a free-free photon absorption of the Sr-Ca quasimolecule.[1] From either viewpoint, for dipole-dipole coupling, a maximum collision cross section is predicted when $\hbar\omega$ is equal to the interatomic transition frequency of the *infinitely separated* atoms ($R=\infty$). For incident radiation at the $R=\infty$ frequency, the predicted cross section for collision σ_c is given by[2]

$$\sigma_c = 2(\pi/\hbar^4)(\mu_{\rm Sr}\,\mu_{\rm Ca}\mu_{\rm Ca*}\,E/\overline{V}\rho_0\Delta\omega)^2, \qquad (2)$$

where $\mu_{\rm Sr}$, $\mu_{\rm Ca}$, and $\mu_{\rm Ca*}$ are the magnitudes of the dipole matrix elements of the Sr $5s$-$5p$, Ca $4s$-$4p$, and Ca $4p$-$4d$ transitions, respectively; E is the strength of the applied optical field at 6409 Å; $\hbar\Delta\omega$ is the energy difference between the Sr $5p\,{}^1P^\circ$ and Ca $4p\,{}^1P^\circ$ levels; and $\overline{V}$ is the average velocity. The quantity ρ_0 is the minimum impact parameter such that the integrated relative phase shift during transit is 1 rad, i.e.,

$$\frac{1}{\hbar}\int_{-\infty}^{+\infty}\frac{C_6}{R^6(t)}\,dt = \frac{3\pi\mu_{\rm Sr}^2\mu_{\rm Ca}^2}{8\hbar^2\overline{V}\Delta\omega\rho_0^5} = 1,$$

where C_6 is the energy-shift constant of the Sr $5p\,{}^1P^\circ$ level. We neglect the comparatively small shift of the Ca $4d\,{}^1D$ level. Equation (2) includes only the dominant path of the perturbation calculation. For our experiments, $\mu_{\rm Sr}=3.0$ a.u., $\mu_{\rm Ca}=2.8$ a.u., $\mu_{\rm Ca*}=0.95$ a.u.,[3] $\hbar\Delta\omega=1954\ {\rm cm}^{-1}$, $\overline{V}=6.6\times10^4$ cm/sec, $\rho_0=16.7$ Å, and $\sigma_c=(4.3\times10^{-23}\ {\rm cm}^4/{\rm W})P/A$.

A schematic of the experimental apparatus is shown in Fig. 2. The Sr-Ca heat-pipe–type cell was operated at a temperature of approximately 875°C and a zone length of 2 cm. The number densities of the Sr and Ca ground-state atoms were determined by measuring the linewidths of their resonance absorption lines at 4608 and 4228 Å. These were $N_{\rm Sr}=8.0\times10^{16}$ atoms/cm^3 and $N_{\rm Ca}=3.8\times10^{16}$ atoms/cm^3.

A Chromatix CMX-4 flashlamp-pumped dye laser produced the 5757-Å radiation for two-photon pumping of the Sr $5d\,{}^1D$ level. An incident power of about 1 kW was focused to a power density of 1.4×10^7 W/cm^2. Emitted radiation at the $5d$-$5p$ Sr transition frequency was measured and used to estimate the population of the radiatively trapped Sr $5p\,{}^1P^\circ$ level. Typically this density was about $N_{\rm Sr}(5p\,{}^1P^\circ)\simeq6\times10^{15}$ atoms/cm^3. Two-photon pumping and radiative decay was used as the population mechanism of the Sr $5p\,{}^1P^\circ$ storage level in order to avoid the necessity of directly applying radiation at the $5s$-$5p$ Sr frequency and thus possibly masking the desired collision process by a two-photon sum process to the Ca $4d\,{}^1D$ level.

Tunable radiation for inducing the collision process was provided by a Chromatix Model No. 1050 dye laser (Kiton Red) pumped with the second harmonic of the 1.12-μm line of a Q-switched neodymium-doped yttrium-aluminum-garnet laser. A peak power of 175 W in a 150-nsec pulse was focused to a power density of about 8.6×10^6 W/cm^2 and spatially overlapped with the 5757-Å pump beam. The relative timing of the two lasers could be adjusted over a range of several microseconds using a variable delay. Fluorescence of the Ca $4d$-$4p$ transition was measured and used to monitor the population of the Ca $4d\,{}^1D$ level.

The 5757-Å laser was first tuned to maximize the Sr 7673-Å radiation and thus the population of the Sr $5p\,{}^1P^\circ$ level. With both lasers on, a maximum signal was measured with the transfer laser tuned to 6409.0 Å, as compared to the predicted ($R=\infty$) value of 6408.6 Å. (All wavelengths are given in air.) This is within the ±0.7-Å uncertainty of our wavelength calibration. The half-

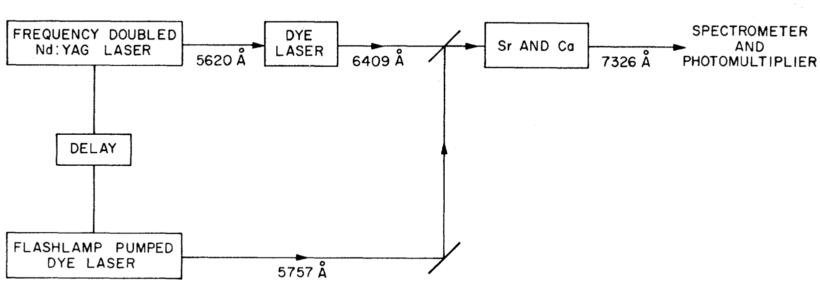

FIG. 2. Schematic of experimental apparatus.

power linewidth for transfer was 1.0 ± 0.2 Å. The linewidth of the Ca 7326-Å fluorescence signal was limited by resolution. The 5757-Å laser, by itself, produced some 7326-Å fluorescence. A signal-to-noise ratio of about 50 was obtained by integrating over five pulses.

The ratio of the fluorescence output at the 7326-Å Ca line to that at the 7673-Å Sr line was 1:36. Allowing for the somewhat tighter focusing ($\sim\times 4$) of the 6409-Å laser, as compared to the 5757-Å laser, this fluorescence ratio indicates a collisional energy transfer of 11% for the excited Sr $5p\,^1P^\circ$ atoms to the Ca $4d\,^1D$ level. This transfer takes place during the 150-nsec pulse width of the 6409-Å laser, indicating an induced cross section of 3×10^{-16} cm^2. This cross section was linear in the incident power density. We estimate an experimental uncertainty of a factor of 3. For our power density of 8.6×10^6 W/cm^2, Eq. (2) predicts a cross section of 3.7×10^{-16} cm^2. The 6409-Å transfer pulse could be delayed by about 0.5 μsec after the end of the 5757-Å pulse before the 7326-Å signal was significantly reduced. This is consistent with the estimated 0.5-μsec radiative trapping time of the Sr $5p\,^1P^\circ$ level.

We note that there are several Sr $6p\,^3P$ levels a few angstroms from coincidence with the Ca $4d\,^1D$ level. By tuning the transfer laser exactly to these levels, we ascertained that no direct collisional transfer was taking place.

Theory predicts that the optically induced collision cross section should continue to increase linearly with power density until $\sigma_c \simeq \pi\rho_0^2$ and as the square root of power density thereafter. For the Sr-Ca system studied here $\pi\rho_0^2 = 8.8\times 10^{-14}$ cm^2. Large energy transfer rates should thus be possible using this process.

The collisional process demonstrated here should be applicable to the direct measurement of interatomic potentials. Additional transfer peaks are expected at frequencies where the difference of the atomic potentials has zero slope, and at frequencies corresponding to turning points of the classical motion. Application to the construction of short-wavelength lasers is likely. Energy could be stored in a metastable atomic level, and by use of a short tunable optical pulse, collisionally switched to a radiative state of a second species. The inverse radiative process where lasing takes place between an excited state of one species and a lower state of a second species may be applicable to the construction of low-gain, high-energy lasers.

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