PHYSICAL REVIEW A VOLUME 15, NUMBER 3 MARCH 1977

Observation of laser-induced inelastic collisions*

R. W. Falcone, W. R. Green,† J. C. White, J. F. Young, and S. E. Harris
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 16 September 1976)

We describe two new experiments which demonstrate laser-induced inelastic collisions. Energy was stored in the Sr $5p\,^1P^o$ state and selectively transferred both to the Ca $4p^2\,^1S$ and $5d\,^1D$ states. The collision cross sections maximized at the interatomic wavelengths of 4977 and 4711 Å, respectively, and had a half-power width of about 14 cm^{-1}.

In a recent Letter[1] we reported the demonstration of a laser-induced inelastic collision; further investigations, however, have shown that an artifact, rather than the reported effect, was observed.[2,3] We report two new experiments which avoid this problem.

The signals we observed previously were apparently produced by the following sequence: population of the Sr $4d\,^3D_3$ level, probably by quenching of the pumped Sr $5p\,^1P^o$ state; laser excitation at 6408.5 Å to the Sr $5p'\,^3F_4^o$ state; and finally normal inelastic collisional transfer to the Ca $4d\,^1D$ level. The resolution of our experiment was not sufficient to distinguish between this process and the laser-induced transfer predicted to occur at the interatomic ($R=\infty$) wavelength of 6408.6 Å. (All wavelengths are given in air.) When we tuned the transfer laser to other lines in the Sr $4d\,^3D$-$5p'\,^3F^o$ series, we observed signals of comparable magnitude and line shape, thus confirming that such an artifact, rather than the reported effect, had been observed. We note that these processes exhibited narrow linewidths which pressure broadened as expected for atomic transitions.

Recently we have performed two new experiments in the Sr-Ca system which eliminate the above artifact by avoiding a wavelength coincidence between the interatomic transfer wavelength and any known Sr or Ca transitions.[4-6] Laser-induced energy transfer to both the Ca $4p^2\,^1S$ and $5d\,^1D$ states has been observed[7]:

$$\mathrm{Sr}(5p\,^1P^o)+\mathrm{Ca}(4s^2\,^1S)+\hbar\omega(4977\text{ Å}) = \mathrm{Sr}(5s^2\,^1S)+\mathrm{Ca}(4p^2\,^1S), \quad (1)$$

$$\mathrm{Sr}(5p\,^1P^o)+\mathrm{Ca}(4s^2\,^1S)+\hbar\omega(4711\text{ Å}) = \mathrm{Sr}(5s^2\,^1S)+\mathrm{Ca}(5d\,^1D). \quad (2)$$

The experimental details were similar to those described in Ref. 1. A heat-pipe-type cell provided a ~3-cm-long zone of Sr and Ca vapor at atomic densities of $N_{\mathrm{Sr}}=4\times10^{16}$ and $N_{\mathrm{Ca}}=6\times10^{15}$ cm^{-3}, as determined by the resonance line curve of growth method.[8] The Sr $5p\,^1P^o$ level was populated by direct single-photon absorption of a flash-lamp-pumped dye laser tuned about 50 cm^{-1} to the red side of the resonance line, focussed to an area of 10^{-3} cm^2 and a power density of 1×10^4 W/cm^2. The radiatively trapped, excited state density N^*_{Sr} was determined using a 6357-Å probe laser to saturate the Sr $5p\,^1P^o$-$7s\,^3S$ transition and measuring the resulting $7s\,^3S$-$5p\,^3P^o$ fluorescence. The measured density was typically $N^*_{\mathrm{Sr}}=3\times10^{13}$ cm^{-3}, and should be considered an upper bound, since collisional quenching could reduce the Sr $5p\,^1P^o$ population in the absence of the strong probe field.

The excited-state Ca population, N^*_{Ca}, produced by the laser-induced collisional energy transfer was determined by measuring the fluorescence of the Ca $4p^2\,^1S$-$4p\,^1P^o$ [Eq. (1)] or Ca $5d\,^1D$-$4p\,^1P^o$ [Eq. (2)] transitions. An energy-level diagram for transfer into the Ca $4p^2\,^1S$ level is shown in Fig. 1. The ratio of the Ca to Sr fluorescence signals was used to determine the transfer ratio $N^*_{\mathrm{Ca}}/N^*_{\mathrm{Sr}}$, and

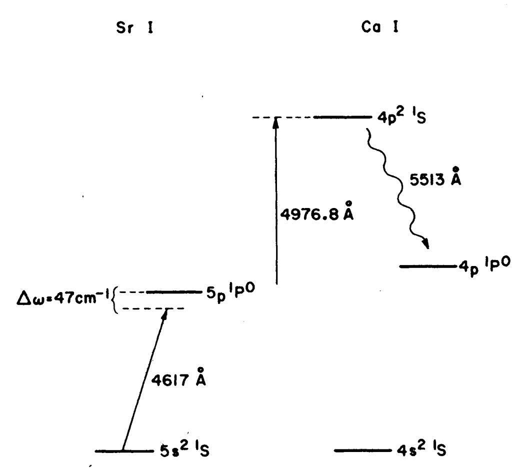

FIG. 1. Energy-level diagram for laser-induced transfer from Sr $5p\,^1P^o$ to Ca $4p^2\,^1S$.

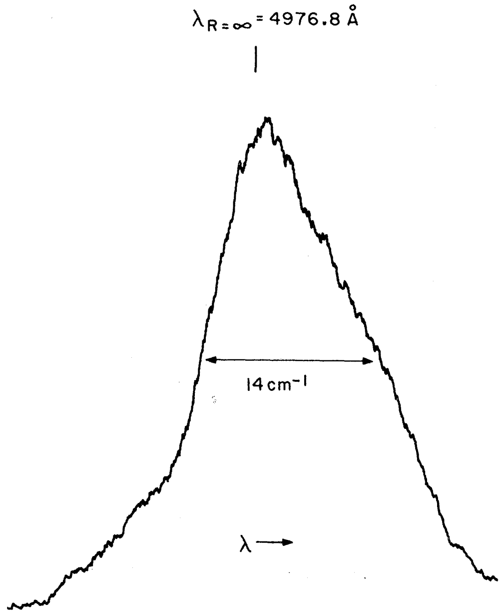

FIG. 2. Induced cross section as a function of transfer laser wavelength.

thus the cross section $\sigma_c = (N^*_{\mathrm{Ca}}/N^*_{\mathrm{Sr}})/N_{\mathrm{Ca}}\overline{V}\tau$, where $\overline{V}$ is the average velocity (9×10^4 cm/sec), and τ is the laser pulse length (1 μsec).

The relative magnitude of the cross section for transfer to the Ca $4p^2\,{}^1S$ level is shown in Fig. 2 as a function of transfer laser wavelength; the curve has not been corrected for the 2-cm^{-1} laser linewidth. The cross section has a maximum at about the $R=\infty$ wavelength of 4976.8 Å, a half-power linewidth of 14 cm^{-1}, and drops less rapidly for red detuning than for blue detuning of the transfer laser. This line shape remained constant when the argon buffer-gas density was varied from about 10^{17} to 10^{18} cm^{-3}, in contrast to "artifact" signals of the type described above. The line shape and position also remained unchanged as the pump laser wavelength was tuned both above and below the Sr $5p\,{}^1P^o$ level, thus eliminating the possibility that the signals were produced by quasimolecular two-photon absorption into the Ca $4p^2\,{}^1S$ state. We have also examined the possibility that the signals are caused by two-photon absorption, with one photon provided by the transfer laser, and a second by spontaneous fluorescence from the Sr resonance level. Calculations show that such a process would be at least 1000 times weaker than the signals observed. In addition, the observed constant linewidth and red asymmetry is inconsistent with the known asymmetric Ar broadening of the Sr resonance level.[9]

The red asymmetry in Fig. 2 indicates that the van der Waals constant C_6 for the Ca $4p^2\,{}^1S$ state exceeds that for the Sr $5p\,{}^1P^o$ storage state; this is consistent with a rough calculation based on large Ca matrix elements to nearby states.[10] The line shape for transfer into the Ca $5d\,{}^1D$ state [Eq. (2)] appears similar to Fig. 2, but precise evaluation is not possible since the wing is distorted by an adjacent peak at 4714 Å associated with the Sr $4d\,{}^3D$-$7p\,{}^3P^o$ transition.

For transfer into the Ca $4p^2\,{}^1S$ level, theory predicts a velocity averaged cross section of $\sigma_c = 4\times10^{-23}P/A(\mathrm{W/cm^2})\ \mathrm{cm}^2 = 2\times10^{-17}\ \mathrm{cm}^2$ for our transfer power density of 5×10^5 W/cm^2. Experimentally we found that $\sigma_c = 9\times10^{-18}$ cm^2, and that σ_c varied linearly for small variations of transfer laser power density. We feel the measured value of σ_c represents a good lower bound, and experimental uncertainties, especially number densities, can reasonably account for the theoretical-experimental discrepancy.

We would like to note that considerable care in the adjustment of experimental parameters is required in order to observe this effect. In particular, background noise (no transfer laser) at the observation wavelength increased quadratically with increasing pump power while the signal increased only linearly. The Sr $5p\,{}^1P^o$ density was chosen to provide both a reasonable signal-to-noise ratio (~5) and detectable signals. For our conditions, the signals are typically $\frac{1}{50}$ of that obtained by tuning the transfer laser to a frequency such that the sum of it plus the pump laser excites the Ca $4p^2\,{}^1S$ level by two-photon absorption.

These experiments have shown that laser-induced transfer provides a means of selectively directing stored atomic energy into particular states. In addition, the cross-section line shapes can be used to determine interatomic potentials. Topics for future study include laser-induced spin-exchange and charge-exchange collisions; these processes should have cross sections comparable to that reported here, but should exhibit a maximum considerably shifted from the interatomic wavelength.

The authors gratefully acknowledge helpful discussions with Dr. Alan Gallagher, Dr. Sidney Geltman, and Dr. M. G. Payne.

---

*Work jointly supported by the Office of Naval Research under contract number N00014-75-C-0576 and by the Energy Research and Development Administration.

†W. R. G. gratefully acknowledges support from the Fannie and John K. Hertz Foundation.

[1]D. B. Lidow, R. W. Falcone, J. F. Young, and S. E.

Harris, Phys. Rev. Lett. 36, 462 (1976).
[2]Erratum to Ref. 1: D. B. Lidow, R. W. Falcone, J. F. Young, and S. E. Harris, Phys. Rev. Lett. 37, 1590(E) (1976).
[3]S. E. Harris, D. B. Lidow, R. W. Falcone, and J. F. Young, in *Tunable Lasers and Applications*, edited by A. Mooradian, T. Jaeger, and P. Stokseth (Springer-Verlag, New York, 1976).
[4]C. E. Moore, *Atomic Energy Levels*, U.S. Natl. Bur. Stand. NSRDS-NBS 35 (U.S. GPO, Washington, D.C., 1971), Vols. I and II.
[5]A. R. Striganov and N. S. Sventitskii, *Tables of Spectral Lines of Neutral and Ionized Atoms* (Plenum, New York, 1968).
[6]A. N. Zaidel', V. K. Prokf'ev, S. M. Raiskii, V. A. Slavni, and E. Ya. Shreider, *Tables of Spectral Lines* (Plenum, New York, 1970).
[7]The Ca $4p^2\,{}^1S$ state, at 41786.3 cm^{-1}, is incorrectly designated in Ref. 4 as $6s\,{}^1S$. G. Risberg, Ark. Fys. 37, 231 (1968).
[8]A. P. Thorne, *Spectrophysics* (Chapman and Hall, London, 1974), p. 307.
[9]J. M. Farr and W. R. Hindmarsh, J. Phys. B 4, 568 (1971).
[10]G. McGinn, J. Chem. Phys. 51, 5090 (1969).