total strength of $0.27\mu_0^2$ which is quite comparable to experiment. At higher energies an additional strength of $0.6\mu_0^2$ is predicted to be fragmented over a number of levels. The ground-state wave function generated in this calculation has a 17% 2p, 2h intensity.

On the other hand, the SU_3 shell-model calculations of Millener[9] predict only $\sim 0.05\mu_0^2$ of ground-state $M1$ strength between 16 and 20 MeV. The total $M1$ strength is predicted to be weak because the ground-state correlations are predicted to be mainly of maximum spatial symmetry [4^4] and hence cannot be excited by the dominant (spin) part of the $M1$ operator. The present experimental results appear to require the ^{16}O ground-state correlations to be mainly of lower spatial symmetry.

In the future it will be interesting to search experimentally for $M1$ strength at higher energies in ^{16}O, although this will probably be appreciably more difficult.

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Laser-Induced Charge Transfer to an Excited Ionic State

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We report the observation of a laser-induced charge-transfer collision. In the presence of an intense laser beam, ground-state calcium ions collide with ground-state strontium atoms, selectively producing excited strontium ions and calcium neutrals. The laser-induced collision cross section has a linewidth of about 50 cm^{-1} and peaks ~ 70 cm^{-1} from that wavelength which satisfies the energy defect of the separated atoms.

This Letter describes experimental observations which we believe demonstrate, for the first time, a laser-induced charge-transfer collision.[1-4] Energy is first stored in the form of *ground-state ions* of one species. An intense laser field is than used to transfer this energy rapidly and selectively to an excited ionic state of a second species.

The process studied is

$$Ca^+(4s\,^2S_{1/2}) + Sr(5s^2\,^1S_0) + \hbar\omega \rightarrow Ca(4s^2\,^1S_0) + Sr^+*(5p\,^2P_{3/2}{}^0) \quad (1)$$

and is shown schematically in Fig. 1. Without the laser photon, Eq. (1) is endothermic by 2.6 eV (~ 30 kT at 1000°K) and has a vanishingly small cross section. The laser photon supplies this energy and may be thought of as raising one of the two Sr valence electrons to a virtual level of approximately $Sr(5s\,5p\,^1P_1{}^0)$ character; the unexcited Sr electron can than be captured by Ca^+, leaving Sr in the $Sr^+(5p\,^2P_{3/2}{}^0)$ excited state. Theory predicts[1-4] that, in contrast to the case of the previously studied dipole-dipole processes,[5-17] the maximum cross section for the laser-induced charge-transfer process should occur when the laser is tuned to a wavelength shifted (typically) by about 100 cm^{-1} from that which exactly satis-

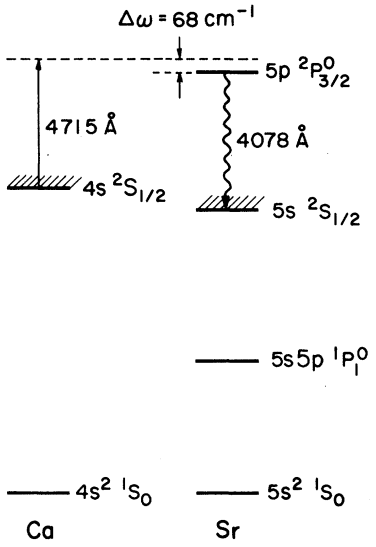

FIG. 1. Energy level diagram for the laser-induced charge-transfer process $Ca^+(4s^2S_{1/2}) + Sr(5s^2\,^1S_0) + \hbar\omega \rightarrow Ca(4s^2\,^1S_0) + Sr^+*(5p^2P_{3/2}{}^0)$.

fies the energy defect of Eq. (1).

The experimental system used to study the laser-induced charge-transfer process has been described previously.[17] Briefly, the output of an actively mode-locked Nd-doped yttrium-aluminum-garnet oscillator-amplifier system was upconverted to 3547 Å and used to synchronously pump two dye lasers. Both dye lasers were cavity dumped to produce two independently tunable 40-ps pulses of several megawatts peak power with linewidths of about 10 cm^{-1}. The output of one laser, the pump laser, was used to create Ca^+ ground-state ions. This was done by two-photon pumping the $Ca(4s^2\,^1S_0)$-$Ca(4s4d\,^1D_2)$ transition; a third photon of this same laser completed the ionization. The 40-ps-long pulse from the transfer laser was delayed by 5 ns from that of the pump laser and was scanned over the range $4510\text{ Å} < \lambda_T < 5060\text{ Å}$. Both laser beams were focused into a metal-vapor cell to an area of about 10^{-3} cm^2. The cell was operated at about 840°C providing ground-state densities of both species of about 10^{16} atoms/cm^3. To protect the windows, the cell also contained a background pressure of 15 Torr of Ar.

Population of the $Sr^+(5p\,^2P_{3/2}{}^0)$ target state was detected by imaging the resulting resonance line fluorescence at 4078 Å into a 1-m spectrometer of about 4 Å resolution equipped with an RCA 31034 photomultiplier. As the transfer laser was scanned, the signal from the photomultiplier was integrated over each each of four consecutive 10-ns time intervals by a set of gated integrators. For each pulse the outputs of the integrators were recorded by a minicomputer and smoothed at a later time.

The ability to obtain time resolution of the fluorescence output from the target state is particularly important in an experiment of this type. The transfer laser is quite intense (~ 10^9 W/cm^2) and as it is scanned it will excite various single- and multiple-photon transitions in both Sr and Ca, thus producing highly excited neutrals and ground-state ions. Collisions of these species with each other, as well as with free electrons, will populate the target state during a time which is characteristic of their lifetime (several hundred nanoseconds for ions and resonance-line neutrals). On the other hand, the laser-induced collisional process only occurs for the 40 ps during which the transfer laser is present. The end product of this process will decay at a rate corresponding to the fluorescence lifetime of the target state (~15 ns).

Figure 2 shows the integrated amplitude of the target-state fluorescence at 4078 Å as a function of the transfer-laser wavelength, during each of the sequential 10-ns gates. Laser-induced charge transfer to the excited ionic state is observed to peak at $\lambda_T = 4715$ Å. This peak occurs at a wavelength which is 15 Å shorter (68 cm^{-1}) than that wavelength ($\lambda_{R=\infty} = 4730$ Å) which exactly satisfies energy conservation for the separated atoms. The line shape of the laser-induced charge-transfer process is roughly symmetrical and has a width of about 50 cm^{-1}. One other single-photon and two multiple-photon excitations are noted in Fig. 2. These are each identified and persist almost unchanged in amplitude for the 40-ns observation period. The laser-induced charge-transfer collisional process, however, decays with a time constant of ~ 15 ns.

The target-state excitation obtained when the transfer laser is tuned to the Sr resonance line at 4607 Å (Fig. 2) is similar to the two-step process recently reported by Dutta *et al.*[18] In the first step, the $Sr(5s5p\,^1P_1{}^0)$ state is populated by the transfer laser; in the second, population is transferred to the target state by an exothermic (~ 500 cm^{-1}) charge-transfer collision. Unlike the $\lambda_T = 4715$ Å process, this two-step process persists for the lifetime of the excited Sr state and maintains a nearly constant amplitude over the full 40-ns observation period.

Figure 3 shows the relative amplitude of the

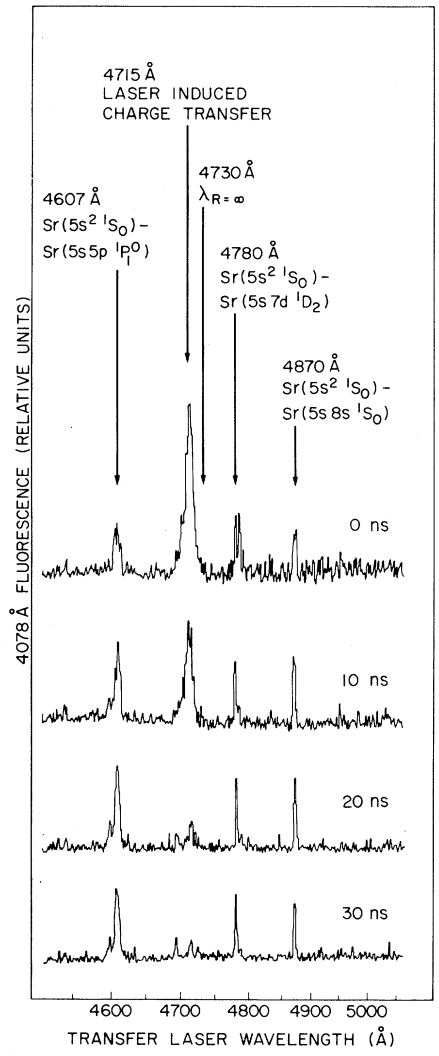

FIG. 2. Fluorescence from the $Sr^+(5p\ ^2P_{3/2}{}^0)$ target state as a function of transfer-laser wavelength integrated over each of four consecutive time gates.

$\lambda_T = 4715$ Å signal as a function of the transfer-laser power density. Some saturation is apparent at a laser power density of about 10^8 W/cm².

Since the density of ground-state Ca^+ was not measured, it is not possible to make an accurate estimate of the laser-induced collision cross section. We estimate that the population of the two-photon–pumped $Ca(4s4d\ ^1D_2)$ level was about 10^{14} atoms/cm³. An assumed cross section for photoionization from this level to the Ca^+ continuum of 10^{-18} cm² implies a Ca^+ density of about 10^{13} ions/cm³. From calibration of the target-state fluorescence we estimate a target density of 10^{10} excited ions/cm³ produced per transfer-laser

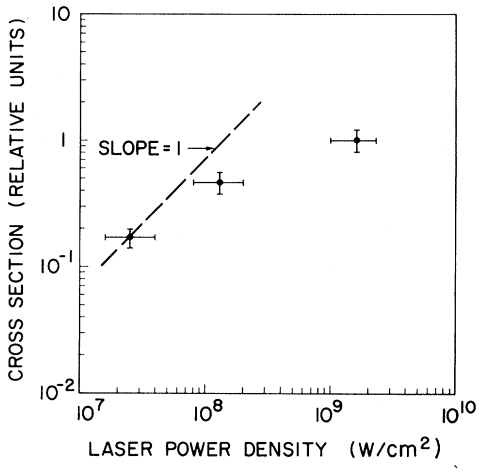

FIG. 3. Relative cross section for the laser-induced charge-transfer process as a function of laser power density.

pulse, thus implying, as a rough estimate, a laser-induced collision cross section of about $\sigma_c = 5\times10^{-15}$ cm².

To confirm our interpretation of the $\lambda_T = 4715$ Å signal, several additional measurements were made: (1) The expected selectivity of a laser-induced collision was verified by tuning the analyzing spectrometer so as to observe fluorescence from the other (800 cm^{-1} lower) member of the Sr^+ resonance line doublet. The $\lambda_T = 4715$ Å signal disappeared, while the two-photon signals remained. (2) The Ca^+ population was eliminated by detuning the pump laser by 10 Å from the two-photon state in Ca. This caused the signal at $\lambda_T = 4715$ Å and also at $\lambda_T = 4607$ Å to disappear, while the two-photon signals remained. (3) Similarly, the $\lambda_T = 4715$ Å signal was not observed in a cell containing only Sr. (4) With the pump laser tuned to give two-photon pumping of the $Sr(5p^2\ ^1S_0)$ level, thereby directly producing Sr excited-state neutrals and ions, the $\lambda_T = 4715$ Å signal was not observed. (5) The linewidth, shape, and position of the $\lambda_T = 4715$ Å peak were unaffected by increasing the Ar buffer-gas pressure by a factor of 10. (6) The analyzing spectrometer was tuned to monitor fluorescence from the $Ca^+(5p\ ^2P_{3/2}{}^0)$-$Ca^+(5s\ ^2S_{1/2})$ resonance transition. Again, no signal was present at $\lambda_T = 4715$ Å.

We note that a narrow (<3 cm^{-1}), very weak component, $J=4 \rightarrow J=4$, of the Sr autoionizing multiplet $Sr(4d5p\ ^3F^0)$-$Sr(4d6d\ ^3F)$ has been reported at 4715 Å.[19] The absence of any signal with the transfer laser tuned to the (thermalized) $J=3 \rightarrow J=3$ component at 4662 Å makes it highly

unlikely that the $J=4\rightarrow J=4$ component makes any contribution to the observed 4715-Å signal.

The experimentally observed line shape for the laser-induced charge-transfer process is narrower and more symmetric, and occurs closer to the energy defect of the infinitely separated atoms, than is expected on the basis of a Landau-Zener curve-crossing model.[1,2,4] The similarity of the polarizabilities[20] of ground-state Ca and ground-state Sr imply quite parallel potentials for the initial and final quasimolecular states, and a shallow crossing at large R. This situation probably requires a calculation along the lines of Copeland and Tang[3] or Rapp and Francis.[21]

Ground-state ions are easily created and represent a means of long-lived energy storage. Laser-induced charge-transfer collisions provide a technique for selectively and rapidly channeling this energy into a designated excited state.

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Pulsed Optical-Optical Double Resonance Spectroscopy of *Gerade* Excited Electronic States in Li_2

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Optical-optical double resonance using two nitrogen-laser–pumped dye lasers has been used to make the first spectroscopic study of the hitherto unobserved *gerade* excited electronic states of $^7Li_2$. Three $^1\Sigma_g^+$ and two $^1\Pi_g$ states are observed. Molecular constants in cm^{-1} for one of the $^1\Pi_g$ states are $T_e=31\,868.02$, $\omega_e=229.71$, $\omega_e X_e=1.654$, $B_e=0.469\,86$, $\alpha_e=0.005\,959$, $D_e=7.301$, and $r_3=3.198$ Å.

Interest in the electronic structure of Li_2 dates from the time of the earliest application of quantum mechanics to molecular structure.[1] After molecular hydrogen, Li_2 is the least complex of the stable homonuclear diatomic molecules, and its electronic structure continues to be of funda-

