successful.

As one can see from Figs. 2(a) and 2(b), the agreement between calculated and observed lineshapes is very good. However, one anomaly resulting from fitting the lines has remained. The temperature needed to fit various transitions varied considerably, with the ^1D transition requiring $T \sim 450-500$ K which is consistent with 7^1S-9^1P measurements, but the high-L states requiring considerably lower $T \sim 250-400$ K. At present we do not know whether this represents a slight deviation from a Boltzmann distribution for the atoms or has some other cause.

In the fits in Figs. 2(a) and 2(b), the only free parameters are T and the transition field B_0 for a $v_\perp = 0$ atom. For the data illustrated in Figs. 2(a) and 2(b), we are able to find B_0 to within ± 4 G (± 10 MHz). Thus assuming that we will be able to find B_0 to within ± 4 G for all our observations at the different magnetic fields (with different laser lines), we should be able to obtain values for the zero-field energies of these states with about an order of magnitude greater precision than the values that presently exist.[1]

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Resonance Charge Transfer from a Photoexcited Donor State

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We report the experimental observations of resonance charge transfer from a photoexcited donor state in sodium to excited acceptor states in protons and other rare-gas ions. This reaction is followed by rapid decay of the excited acceptor states with the emission of intense spontaneous radiation in the vuv.

The resonant charge-transfer process [see Fig. 1(a)], $A + B^+ \rightarrow A^+ + B^*$, of an electron from an atom A to an ion B has been studied extensively in the past.[1] The corresponding charge-transfer cross section is known to decrease rapidly with the energy defect ΔE between the donor state of atom A and the acceptor state of atom B; the cross section for nonresonant charge transfer [Fig. 1(b)] is generally very small but approaches 10^{-14} cm^2 in the accidental case of resonance $\Delta E = 0$ [Fig. 1(a)]. Recently, Vitlina, Chaptik, and Entin[2] and Copeland and Tang[3] considered nonresonant charge transfer between atoms and ions in the presence of intense light via a virtual intermediate state [Fig. 1(c)]. It was shown that if the $h\nu$ of the incident light is approximately equal to the energy defect ΔE between the donor and acceptor states in the nonresonant process, the intense light can "switch" the cross section for the nonresonant process to a value approaching that of the resonant process. Numerical estimates showed,[3] however, that the required light intensity for this must be quite high and the process has so far not been observed.[4] There has been suggested recently[5] the possibility of using a near-resonant intermediate real state in the donor atom [see Fig. 1(d)] to enhance the photon-induced charge-transfer cross section in analogy with the resonance-enhancement effect in multiphoton processes such as the resonant Raman effect. In this Letter, we report the first unequivocal experimental observation of charge transfer from a photoexcited near-resonant intermediate real donor state. The effect has been observed in charge-transfer systems Na-H^+, Na-D^+, Na-Ne^+, Na-Ar^+, and Na-Kr^+ with the concomitant generation of relatively intense spontaneous vuv radiation at 1216, 584, 736, 1048,

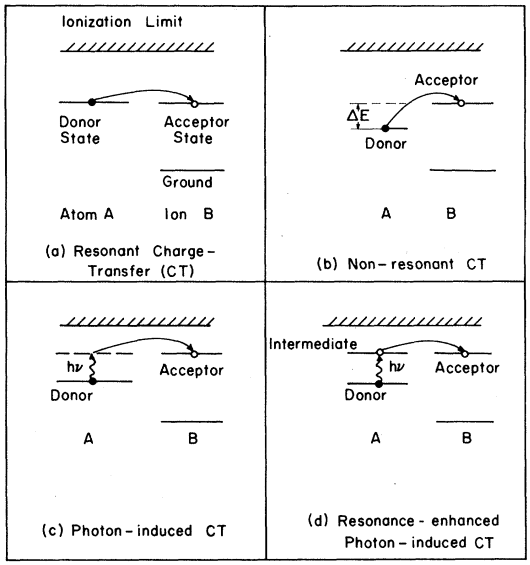

FIG. 1. Schematics for various charge-transfer processes. Note that the intermediate state in (c) is a virtual state (shown dashed) and is a real state (solid line) in the case of (d).

and 1165 Å, respectively.

We discuss first the photon-induced charge-transfer process for the case of Na-H^+ with the reaction

$$H^+ + Na(3s) + h\nu \rightarrow H(2p) + Na^+ \quad (1)$$

in which an electron is transferred from a sodium atom initially in the ground state ($3s$) to the proton forming a hydrogen atom in the $2p$ level in the presence of light. Because the radiative lifetime of H($2p$) is relatively short, Reaction (1) is immediately followed by the spontaneous decay of H($2p$) with the emission of Lyman-α radiation at 1216 Å. For charge transfer, the ground state ($3s$) of sodium is far from being resonant with the $2p$ level of hydrogen. For reaction (1) to take place without resonance enhancement, ΔE is 1.74 eV (see Table I). Experimentally, however, strong charge transfer in the presence of intense light is observed when the wavelength of the incident light was tuned near the Na D_1 and D_2 wavelengths (~ 2.1 eV). This is due to the fact that the upper states of the Na D lines, the Na $3p$ states are near resonant with the H($2p$) level, $\Delta E \cong 0.37$ eV. The intense incident light switches the charge-transfer cross section from the value for the nonresonant process Na($3s$)$\rightarrow H(2p)$ to the much larger value for the near-resonant process Na($3p$) $\rightarrow$ H($2p$). The presence of the Na $3p$ states, therefore, enhances the photon-induced charge transfer from Na($3s$) to H($2p$).

The experiment is performed by colliding a pulse of moving plasma with a stationary cloud of Na atoms. The plasma pulse is derived from a plasma gun which has been previously described.[6] The Na cloud is obtained by flash heating a W ribbon that was previously coated with Na from an oven of standard design. The interaction region of protons and Na cloud could be viewed through pin holes by a solar-blind pumped photomultiplier and through windows by an optical detection system consisting of a monochromator and a photomultiplier. The light source used was a nitrogen-laser–pumped dye laser with ~ 0.5-Å linewidth and producing a maximum energy of 350 μJ in a 7-nsec pulse.

Experimentally, following firing the gun a background of recombination Lyman-α radiation was always observed, with or without the presence of either the Na vapor or laser light. To verify Reaction (1), one must be able to distinguish the Lyman-α radiation signal that results from this particular reaction over that due to all the other possible sources. Lyman-α radiation of several

TABLE I. List of charge-transfer systems investigated with energy defects and observed signal strengths.

System	Wavelength of vuv emission (Å)	ΔE, Na($3s$) to acceptor state (eV)	ΔE, Na($3p$) to acceptor state (eV)	Charge transfer (CT) without light	Ratio of resonance-enhanced photoinduced CT to CT without light (%)
Na-H^+	1216	1.74	−0.37	Medium	80
Na-D^+	1216	1.74	−0.37	Medium	80
Na-He^+	584	1.77	−0.33	Medium	30
Na-Ne^+	736	0.42	−1.68	Large	5
Na-Ar^+	1048	1.21	−0.89	Small	< 5
Na-Kr^+	1165	1.78	−0.32	Medium	60

origins could be observed experimentally in our setup under various conditions: (1) There is always the recombination radiation that requires the presence of the plasma pulse only. (2) Under suitable conditions, an additional Lyman-α pulse is also observed in the presence of Na vapor but without any incident light. (3) Finally, with the application of a short intense pulse of sodium D light, the Lyman-α radiation that could be observed in case (2) shows a sharp jump that always coincides precisely with the incident laser pulse. This jump is associated with Reaction (1). Figure 2 shows photographs of the oscilloscope traces of various observed Lyman-α signals.

Figure 2(a) shows the observed Lyman-α signal versus time in the absence of any laser light.

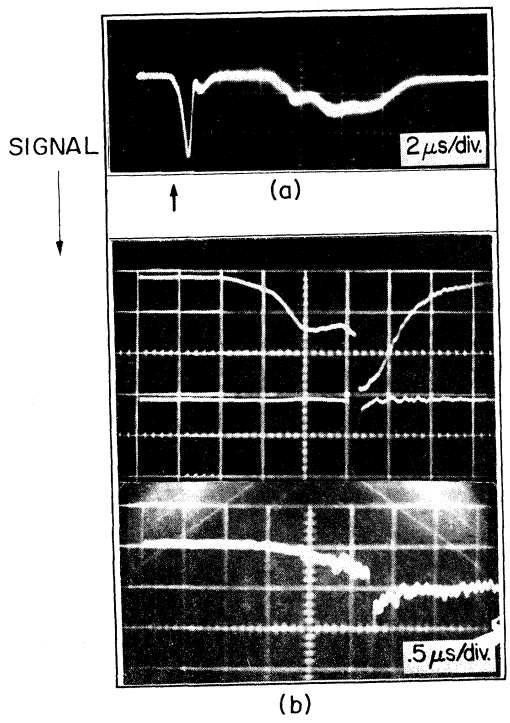

FIG. 2. (a) Charge-transfer signal without light vs time. The time scale is 2 μs/div. (b) Top trace, resonance-enhanced photon-induced charge transfer vs time; middle trace, incident laser intensity; bottom trace, Na D luminescence signal vs time. The jump in the bottom trace is due to a jump in the luminescence. The spike barely visible in the photograph at the onset of the jump on the bottom trace is due to scattered laser light. The slight shift in the upper trace with respect to the middle trace arises from the difference in the response time of the photodiode (middle) over that of the photomultiplier (top); the displacement seen in the bottom trace is due primarily to the use of a second oscilloscope with slightly different timing characteristics. The time scale is 0.5 μs/div.

The sharp pulse at the front (arrow) arises only when the plasma gun is fired in the presence of sodium vapor. This signal is attributed to charge transfer. The second rather broad signal is due to plasma recombination and is present even without any sodium. Since the ground state Na(3s) is not resonant with the H(2p) state, we believe that the charge-transfer signal shown in Fig. 2(a) arises in charge transfer from the excited state Na(3p). This is confirmed experimentally by simultaneously monitoring the sodium luminescence from the 3p state. The most-probable excitation mechanism for Na(3p) in this case is through impact excitation by plasma electrons. By appropriate settings of gun conditions and experimental manipulations, the charge-transfer pulse could be made by far the strongest signal present.

Figure 2(b) shows the effect of intense light tuned to the Na D wavelengths on the charge-transfer signal. The top trace shows the charge-transfer signal on an expanded time scale so that we look only at the first 5 μs during which the charge-transfer pulse occurs. The middle trace shows the dye laser pulse monitored by a separate photodiode. The timing of the laser pulse is adjusted such that it overlaps the long charge-transfer pulse arising from plasma electrons. As can be seen from the figure, there is a substantial jump in the charge-transfer signal (by as much as a factor of 2) which coincides in time with the laser pulse. The fall time of the observed signal is longer than that of the laser pulse because of radiation trapping due to both sodium and neutral hydrogen. The jump in the observed Lyman-α radiation was observed to scale linearly with the laser intensity, and to disappear when the laser was tuned away from the sodium D lines. We also performed a test with the plasma pulse and the laser pulse but without the sodium vapor, and there was no signal, as expected. These observations rule out such explanations as multiphoton effects in the photomultiplier and verify that the photon-induced 2p electron in the hydrogen atom originates from the ground state 3s of Na and requires the presence of the resonant intermediate state Na 3p in order to make the transfer.

The actual process of this jump in the charge-transfer signal is most probably the resonant excitation of the sodium atom from 3s to 3p followed immediately by charge transfer from Na(3p) to H(2p). Hence the laser light switches the nonresonant charge-transfer cross section of the

Na($3s$) state to the near-resonant one of the Na($3p$). A test of this conjecture was performed by observing the Na($3p$)→Na($3s$) luminescence in the sodium vapor simultaneously with the Lyman-α radiation from the hydrogen. The luminescence profile is shown in the lowest trace of Fig. 2(b). There is a long pulse in the luminescence signal corresponding to the long charge-transfer pulse. In addition, there is also a jump in the luminescence signal that coincides in time with the jump in the charge-transfer signal. The occurrence of the long pulse in the luminescence signal verifies that sodium is excited into the $3p$ states by the plasma electrons even in the absence of light; the jump verifies that additional Na is excited from the $3s$ level to the $3p$ states by the laser light which in turn leads to the jump in the H($2p$) population. Note that the luminescence signal also exhibits the radiation-trapping effect.

This experiment was also performed with other rare-gas plasmas. Our results are summarized in Table I. The Na-D^+ system is analogous to the Na-H^+ system. In the Na-He^+ system, the radiation observed is at 584 Å and is emitted by the excited $1s2p$ state of He. The signal was observed through an aluminum film window (1200 Å thick) supported on a copper grid; the window also served to separate the pumped photomultiplier from the main-chamber vacuum. In the Na-Ne^+ system, the radiation observed is at 736 Å and is emitted by the excited $2p^5 3s\,(^2P_{1/2})$ state of Ne. The signal was again observed through an aluminum film window. In the Na-Ar^+ system, the radiation is at 1048 Å and originates from the transition from the $3p^5 4s\,(^2P_{1/2})$ to the ground state. In the Na-Kr^+ system the radiation at 1165 Å is due to the transition from the $4p^5 5s\,(^2P_{1/2})$ state to the ground state of Kr. The observed relative strengths of the photon-induced charge-transfer agree qualitatively with crude estimated ratios of charge-transfer cross sections from the near-resonant excited state and the ground state of Na based upon the approximate theory of Olson.[7] In the case of Ne, the photon-induced signal is probably due to charge transfer to the second excited state of neon, Ne($3p$), which quickly decays to the Ne($3s$) level and from there to Ne($2p$). We observe the radiation from the Ne($3s$)-Ne($2p$) transition. The observed spontaneous emission at 736 Å for the Na-Ne^+ case is high; in the absence of laser light it is estimated to be 100 W for 1 cm^3, with an ion flux of 10^{21} cm^{-2} s^{-1}, ion velocity of 7×10^6 cm/s, and a sodium density of 10^{14} cm^{-3}.

In conclusion, our experiments clearly demonstrate (1) resonant charge transfer from an excited donor state of an atom to an acceptor state in ions in plasmas, and (2) the enhancement of photon-induced charge transfer from a nonresonant initial donor state through a real intermediate state in the donor atom to the acceptor state or, equivalently, switching of charge-transfer cross section by intense light.

These effects also have important consequences in the development of new laser transitions. Charge transfer of electrons from alkali atoms to protons and rare-gas ions has been considered as a possible mechanism for lasing in the vacuum uv and soft-x-ray region. A short intense pulse of light may be used to enhance the charge-transfer rate in a sharply defined interaction region, thereby creating a population inversion in the acceptor atoms.

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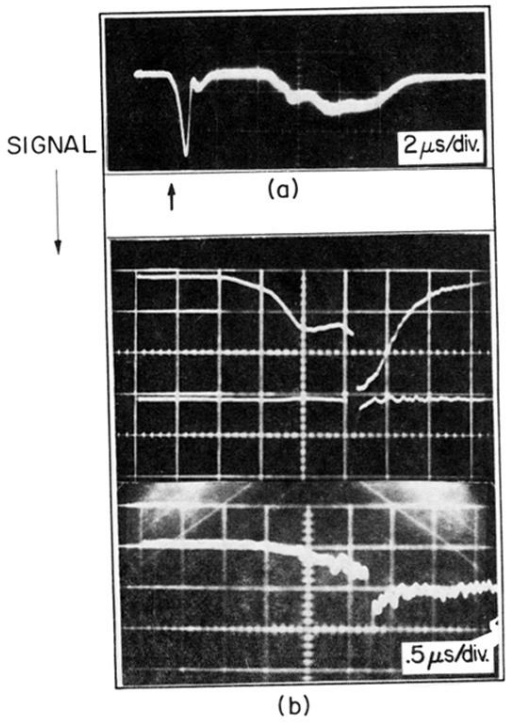

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