Ion-atom charge-transfer system for a heavy-ion-beam pumped laser

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An Ar target to which Cs vapor could be added, excited by a pulsed beam of 100-MeV ^{32}S ions, was studied as a prototype ion-atom charge-transfer system for pumping short-wavelength lasers. Low-velocity Ar^{2+} ions were efficiently produced; a huge increase in the intensity of the Ar II $4d$-$4p$ spectral lines was observed when Cs vapor was added to the argon. This observation is explained by a selective charge transfer of the Cs $6s$ electron into the upper levels of the observed transitions. A rate constant of $(1.4\pm0.2)\times10^{-9}$ cm^3/s for the transfer process was determined.

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The development of accelerators that can provide intense beams of high-energy heavy ions provides an opportunity to study heavy-ion-beam pumped short-wavelength laser systems. In this Brief Report a general pumping scheme is proposed that is well suited for conditions encountered in such experiments. The scheme is based on the use of two steps for effective population of specific levels in ionized target species. The first step is ionization of target atoms into a charge state q^+ in collisions with the high-energy projectiles. In a second step, specific excited levels are efficiently populated in target ions of charge state $(q-1)^+$ by low-velocity charge-exchanging collisions.

A pulsed beam of 100-MeV ^{32}S ions from the Munich Tandem Van de Graaff accelerator was used for excitation of an argon-cesium gas target in a prototype experiment. The light emission from this target was studied with time-resolved optical spectroscopy. The results demonstrate feasibility for heavy-ion-beam pumped lasers with selective charge exchange to the upper level.

The pumping scheme makes use of the following special characteristics of the interaction of heavy ions with matter: Cross sections for single-collision multiple ionizations of target atoms by high-energy heavy ions are high [1] and the recoil velocity of such ionized target species is low [2]. With high beam flux and suitable target pressures, this can lead to a formation of substantial densities of multiply ionized target species in a cold dense environment. The recoil ions can then be used as a short-wavelength laser medium if specific upper laser levels can be effectively populated, for example by charge transfer of bound electrons from a donor atom to the ion, as will be described here. Other methods for multiple ionizations, such as heating matter by intense laser beams or multistep ionization by electron collision in high-current discharges or by intense electron-beam excitation, also lead to an overall heating of the laser medium and a less favorable situation for selective charge transfer.

Short-wavelength laser schemes using charge-transfer processes have previously been proposed by Presnyakov and Shevel'ko with ions in an ion beam as the laser species [3] and by Vinogradov and Sobelman with a plasma expanding into a neutral gas [4]. Dixon, Seely, and Elton have observed population inversion in the Balmer spectrum of C^{5+} by the latter method [5]. Selective electron transfer into excited levels of heavy-ion-beam-produced recoil ions and subsequent x-ray emission from a gas target at very low density was studied by Beyer and Mann [6]. They mention stimulated emission in the soft-x-ray region as a potential application without consideration of collisional rates and competing processes in a system at densities which would be required for lasing.

Charge transfer using the heavy-ion-beam-produced target recoil ions was proposed shortly after the first successful operation of heavy-ion-beam pumped lasers in the infrared spectral region [7]. An appropriate acceptor-donor system was necessary to demonstrate feasibility. A high density of excited ions had to be achieved in order to make the population mechanism easily detectable using optical spectroscopy, and the excited levels must not decay directly to the ground state for potential laser applications. As the ionization cross sections for the production of target species of charge state q decrease with increasing q, one must work with q as low as possible. Charge transfer into higher-lying levels, on the other hand, is easier to accomplish when q is not too low and the outer electron of the electron donor is loosely bound. A classical Coulomb model in combination with tabulated energy levels led to the selection of an argon-cesium mixture for the present experiment.

Beyer and Mann [6] describe a model that had been proposed by Olson and Salop [8] and Ryufuku and Sasaki [9] for the charge transfer which is used to predict the parameters for this experiment. In the model, the outer electron-transfer process from the neutral atom B into an excited level of the ion A,

$$A^{q+}+B\rightarrow A^{(q-1)*}+B^{+}\ ,$$

is described using the potential of the ion of charge state q and the singly ionized core of the donor atom for various internuclear distances R of A and B. This results in a lowering of the bound states E_{A0}, E_{B0} of levels localized

at the two collision partners A and B by $1/R$ and q/R, respectively. An almost resonant charge transfer may occur in this simple picture at a specific internuclear distance R_0 when the energy level E_B of the bound electron in B and the energy of a vacant level E_A in the ion coincide. Energies are measured from the ionization limits at infinite internuclear distance. As an additional requirement, the levels taking part in the transfer must lie above the energy barrier between A and B formed by the combined Coulomb potential of B^+ and A^{q+}:

$$E_A = E_{A0} - 1/R \ ,$$

$$E_B = E_{B0} - q/R \ ,$$

$$E_A = E_B \ ,$$

$$R_0 = (1-q)/(E_{A0} - E_{B0}) \ ,$$

$$E_A, E_B > -(q^{1/2}+1)^2/R_0 \ .$$

For transfer between Cs and Ar^{2+}, considered here, the values are $E_{B0} = -3.89$, $E_{A0} = -4.91$ eV, and $R_0 = 1.4 \times 10^{-7}$ cm. Despite the simplicity of this model, it turns out that the energetics of the system is described adequately for an experimental test of the scheme. The model predicts that the requirements are met by Ar II $4d$-$4p$ transitions in an argon-cesium mixture. For the detection of the process, and future laser applications, it is advantageous that these transitions emit light at wavelengths longer than the photoabsorption edge of the electron donor (Cs: 318 nm). A schematic drawing of the level scheme is shown in Fig. 1.

The model can also be used to estimate the cross section and rate constant of the charge-transfer process. If the cross section is taken as $\frac{1}{2}R_0^2 \times \pi$ then the result is 3.2×10^{-14} cm^2. The factor $\frac{1}{2}$ is the maximum probability for the electron to be transferred into one specific level in the recoil ion during the collision, as discussed by Mott and Massey [10]. A more rigorous model is given by Niehaus [11]. A typical thermal velocity of 400 m/s was used to estimate a transfer rate constant $k = v\sigma = 1.3 \times 10^{-9}$ cm^3/s.

A pulsed beam of 100-MeV $^{32}S^{8+}$ ions and an argon gas target of 100 or 250 hPa pressure were used for the experiment. Cesium could be added as an electron donor for the charge transfer by heating the target, including a cesium reservoir. The beam pulses had a repetition rate of 39 kHz, 2-ns pulse width, and consisted of 4×10^5 projectiles. The argon-cesium mixtures were irradiated by the ion beam in a glass tube and the emission of light was measured by time-resolved optical spectroscopy using a $f = 30$ cm monochromator equipped with a photomultiplier operating in the photon-counting mode. Spectroscopy of pure rare gases and data-taking techniques in a similar system were described by Krötz *et al.* [12].

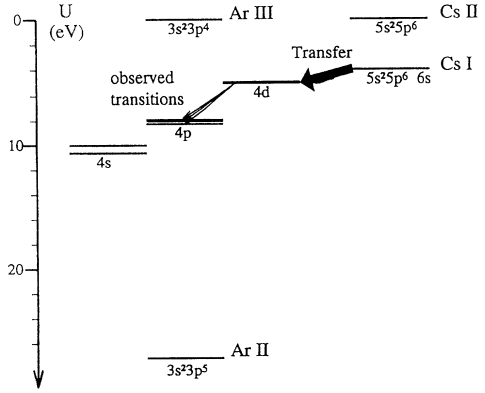

FIG. 1. Schematic drawing of the energy level diagrams of Ar and Cs.

Multiple ionization cross sections of argon in heavy-ion collisions can be obtained from studies performed by Cocke [1]. He reports cross sections of 5×10^{-15} and 2×10^{-15} cm^2 for single and double ionization of argon with 30-MeV chlorine projectiles. As a reasonable estimate, these values can be used for the experiment described here as the projectiles differ only by one atomic number. The ^{32}S ions had an energy of 30 MeV at the point which was imaged onto the entrance slit of the monochromator. Densities of Ar^{1+} and Ar^{2+} ions of the order of 3×10^{11} and 1×10^{11} cm^{-3} in the target gas result from the ionization cross sections for a typical beam diameter of 5 mm. These are approximate values immediately after termination of the beam pulse, when recombination in the ion tracks where the electron density is high can be neglected. The energy deposition of a 5-mm-diameter beam of 30-MeV ^{32}S ions in 250 hPa argon is 4×10^{-6} J per beam pulse and cm^3 which corresponds to 3.8×10^{-6} eV per argon atom. The average power deposition is only 156 mW/cm^3, which can be easily absorbed by the target system, and the target gas remains essentially at the temperature of the oven. On a microscopic scale, the recoil ion velocity encountered in heavy-ion collisions has been studied by Ullrich *et al.* [2]. They find low recoil ion energies of typically 100 meV.

A time-integrated wavelength spectrum using an argon-cesium mixture at 250 hPa Ar and 10 hPa Cs partial pressures is shown in Fig. 2, together with a spectrum of pure argon for comparison. Both spectra are shown

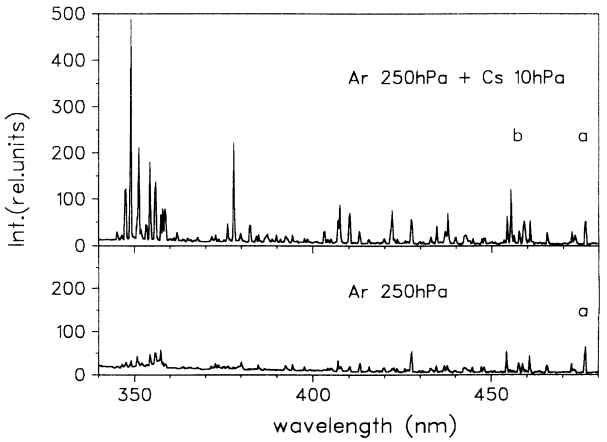

FIG. 2. Time-integrated spectra of an argon-cesium mixture (top) and pure argon (bottom). Both spectra are shown to the same intensity scale. Note the strong increase of Ar II lines between 345 and 385 nm. A strong ArII line in pure Ar spectra at 476.5 nm is indicated for comparison (a). The resonance lines of cesium are also indicated (b).

on the same intensity scale and have been corrected for the spectral response function of the detection system. The appearance of a number of strong lines at wavelengths between 345 and 385 nm is obvious. The Ar II line at 476.5 nm (a in Fig. 2), which is excited by combined ionization and excitation and has been studied before [13], appears with the same intensity in both spectra. This shows that the ion-beam-induced ion line radiation of argon is not affected by the cesium admixture. The resonance lines of cesium (b in Fig. 2) appear at 455.5 and 459.3 nm.

An expanded view of both spectra is shown in Fig. 3. The pure argon spectrum has been subtracted from the argon-cesium spectrum and only those lines remain which show an intensity change when cesium is added. The intensity scale of the pure argon spectrum [Fig. 3(b)] has been changed so that weak lines can be seen for comparison with Fig. 3(a). Note that some very weak argon lines show a strong intensity increase when cesium is added. An assignment of lines showing the intensity increase is given in Table I. It can be found from this table that the lines can be attributed to Ar II which is the first indication that an electron transfer occurs from cesium into excited levels of Ar^{+*} as described above. The upper levels of the lines almost coincide in energy, with a systematic intensity correlation where lines with upper levels of $J=\frac{7}{2}$ show a strong effect, those with $J=\frac{5}{2}$ a medium effect, and those with $J=\frac{3}{2}$ a weak effect. The groups of ^4D and ^4F levels which form the upper levels of the transitions differ in energy by about 200 meV. Energy differences within the groups are less than ± 20 and ± 100 meV, respectively. The energy difference between the groups can be compensated by the recoil ion kinetic energy and may also be caused by different Ar^{2+} precursors since there are levels only 140 and 190 meV above the ground state of these ions. In any case the small energy differences involved indicate a process with matched energy levels with a width of the order of 200 meV or less.

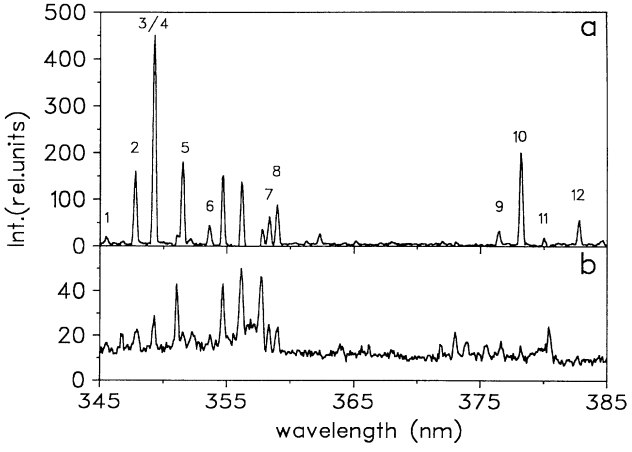

FIG. 3. Expanded and slightly modified view of the data shown in Fig. 2. The pure argon spectrum has been subtracted from the Ar-Cs spectrum to highlight the lines which show an intensity increase when cesium is added. The intensity scale of the pure argon spectrum has been changed to allow comparison with weaker lines.

The upper levels lie 4.83 eV below the ionization limit of Ar II. This value differs from the 4.91 eV predicted by the simple Coulomb model by only 80 meV. These results show that the transfer mechanism was clearly observed. It is obviously effective, as the intensity ratio of the 349.15-nm $4d\,^4D_{7/2}-4p\,^4P^o_{5/2}$ transfer transition, for example, and the 476.5-nm line, which is strong in pure argon, is 6:1.

A measurement of the transfer rate constant was performed using the 349.15-nm line. Time spectra of pure argon and argon-cesium mixtures with Cs densities between 0 and 8.5×10^{16} cm^{-3} were measured. Two examples of the time spectra are shown in Fig. 4. The short-lived component [Fig. 4(a)] is in pure argon and corresponds to the natural lifetime of the upper level. The transfer mechanism leads to an additional emission [Fig. 4(b)] starting immediately with the beam pulse, as can be expected for the two-body Cs-Ar^{2+} collisions, and decaying exponentially with a time constant τ which depends on the cesium concentration. The collisional rate constant can be determined from the slope of the linear dependence of τ versus the cesium concentration, shown as an inset in Fig. 4. The result is $k=(1.4\pm0.2)\times10^{-9}$ cm^3/s, in good agreement with the estimate given above. The value has been determined with a fixed particle density (closed cell) and various temperatures to adjust the Cs concentration. No attempt was made to study the temperature dependence of this value. The error bars shown in the inset of Fig. 4 were determined from the vapor pressure curve of cesium and an uncertainty of the target temperature of 10°C.

In summary, it has been found that *in situ* production of target ions and subsequent charge transfer can lead to selective population of excited levels in a heavy-ion-beam excited target. Low temperatures and recoil ion velocities lead to small Doppler broadening of the emission lines. Population densities of the lower levels of the transitions could be estimated from line intensities. These data show that the lower level population densities are low, so that gain estimates for a laser experiment are encouraging.

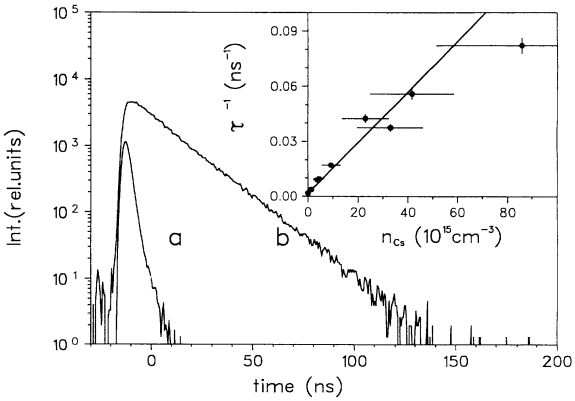

FIG. 4. Time spectra of the 349.15-nm $4d\,^4D_{7/2}-4p\,^4P^o_{5/2}$ transition following the 2-ns excitation by the pulsed ion beam. Data for pure argon (a) show essentially the natural lifetime of the transition. The Cs-Ar^{2+} transfer process leads to a significant extension of the emission time (b). Decay times for various Cs concentrations are shown in the inset.

TABLE I. List of spectral lines which show an intensity increase by adding cesium to the ion-beam-excited argon target. The numbers refer to the spectrum shown in Fig. 3. Energies are listed with respect to the ionization limit of Ar^+. The Einstein A coefficients for the transitions are listed in the last column.

No.	Wavelength (nm)	Lower level (LL)	Upper level (UL)	E_{LL} (eV)	E_{UL} (eV)	A_{ik} (10^8 s^{-1})
1	345.41	$4p\,^4P^o_{5/2}$	$4d\,^4D_{3/2}$	−8.41	−4.82	0.45
2	347.68	$4p\,^4P^o_{5/2}$	$4d\,^4D_{5/2}$	−8.41	−4.84	1.34
3	349.12	$4p\,^4P^o_{3/2}$	$4d\,^4D_{3/2}$	−8.37	−4.82	2.2
4	349.15	$4p\,^4P^o_{5/2}$	$4d\,^4D_{7/2}$	−8.41	−4.86	3.0
5	351.43	$4p\,^4P^o_{3/2}$	$4d\,^4D_{5/2}$	−8.37	−4.84	1.23
6	353.53	$4p\,^4P^o_{1/2}$	$4d\,^4D_{3/2}$	−8.33	−4.82	0.82
7	358.16	$4p\,^4D^o_{1/2}$	$4d\,^4F_{3/2}$	−7.99	−4.53	1.8
8	358.84	$4p\,^4D^o_{7/2}$	$4d\,^4F_{9/2}$	−8.14	−4.68	3.39
9	376.35	$4p\,^4D^o_{7/2}$	$4d\,^4D_{5/2}$	−8.14	−4.84	0.14
10	378.08	$4p\,^4D^o_{7/2}$	$4d\,^4D_{7/2}$	−8.14	−4.86	0.94
11	379.93	$4p\,^4D^o_{5/2}$	$4d\,^4D_{3/2}$	−8.08	−4.82	0.23
12	382.68	$4p\,^4D^o_{5/2}$	$4d\,^4D_{5/2}$	−8.08	−4.84	0.15

We have performed calculations using a model program which has been designed in connection with other heavy-ion-beam pumped lasers to predict the parameters for a mode locked Ar-Cs charge transfer laser on the 349-nm line using an easily available 15-MeV/u Bi beam for pumping. About 1% of the initially formed Ar^{2+} ion density will appear as upper laser level density after the <1-ns beam pulses, leading to about 5% gain per pass. A laser setup to demonstrate this system is being built.

Another important aspect of this work is that the pumping scheme should be applicable and easily predictable for other combinations of ion beams, target ions, and electron donors, in particular for higher charge state systems. The use of very heavy projectiles, which lead to high ionization cross sections and higher charge states in the target, may provide a promising method for implementing analogous pumping schemes at high ionization and shorter emission wavelengths.

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