Laser-Stimulated Two-Step Recombination of Highly Charged Ions and Electrons in a Storage Ring

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Two-step resonant laser-stimulated recombination of highly charged ions was performed for the first time. Nd:YAG laser pulses overlapped with an Ar^{18+} beam in the electron cooler of the ESR storage ring at GSI induced transitions from the continuum to the $n=81$ state of hydrogenlike Ar^{17+}. To avoid reionization in the bending magnet before reaching the detector, the $n=81$ population was transferred to a state well below the reionization threshold by a Ti:sapphire laser. Tuning of this laser yielded the $n=81$ to 36 and 37 transition-line profiles. The two-step method provides access to detailed Rydberg spectroscopy in an electron beam environment.

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Ions stored in a ring and electrons of a cooling beam, copropagating with good spatial overlap and equal average velocities, undergo binary collisions which give rise to spontaneous recombination [1] accompanied by photon emission. This process, being the reverse of photoionization, implies the possibility of induced radiative recombination by additionally merging ions and electrons with an intense laser beam ([2] and references therein). By tuning the laser wavelength into resonance with an optical transition from the energy band of the free cooling electrons to a bound state of the recombined ion system, ions are formed in this final state. Those ions which have experienced radiative recombination are separated from the primary stored beam by the magnetic field of the next bending magnet and are counted with nanosecond time resolution. In this way, the laser light provides a very sensitive probe of various features characterizing the energy regime around the ionization threshold of the down-charged ions in the electron beam environment.

Laser induced *one-photon recombination* was realized experimentally in the recent past for protons [2] and C^{6+} ions [3,4] at the Heidelberg TSR storage ring, and for protons also in a single-pass experiment [5]. The ratio G of the induced radiative recombination rate to the spontaneous rate in the time intervals of interaction between laser light, ions, and electrons was measured as a function of laser wavelength. This yielded the longitudinal and transversal temperatures of the electron beam [2,3]. In addition, it revealed a distortion of the hydrogenic Coulomb potential around the ionization limit, in accordance with the assumption of a radial space charge field of the electron beam, and manifested a population of states immediately below the limit.

The ions and cooling electrons constitute a completely inverted, very thin and cold plasma where effects like subionization threshold population and electrostatic screening of the Coulomb potential of down-charged hydrogenic ions are expected to be increasingly pronounced as a function of nuclear charge Z. This suggests the idea of extending laser-induced recombination to highly charged heavy ions. A long laser wavelength λ is desirable due to the $\sim\lambda^3$ dependence [2] of the induced capture rate, λ being further increased in the ion rest frame by the Doppler shift for copropagating ion and laser beams. On the other hand, the electrons existing in an excited state above a critical principal quantum number n_{cr} corresponding to a binding energy $E(n_{cr})$ are no longer bound if subjected to the motional electric field when entering the next 1 T bending dipole magnet downstream of the electron cooler. For this reason, the laser field must recombine the ions into a state well below this ionization threshold, to ensure their detection. Going to higher nuclear charge, eventually U^{92+}, which has already been stored in the Experimental Storage Ring (ESR) [6] of GSI, shifts $E(n_{cr})$ to higher values. A one-photon transition then requires shorter wavelengths, and, thus, the induced capture rate will drastically decrease. But a reasonable capture rate can be reached via a two-step process [7]. Here, a first long wavelength laser recombines ions and electrons into a state of high quantum number n_i. A second laser beam with much shorter wavelength is tuned into resonance with the transition from n_i to a lower state n_f, safe against field ionization.

Two-step recombination was for the first time realized experimentally for bare Ar^{18+} ions, stored in the ESR at a velocity of $\beta\approx 0.5$ in units of the velocity of light. The number of stored ions was kept on a constant level of 7.5×10^8 by periodically injecting additional ions from the SIS heavy-ion synchrotron. The ions were cooled with an electron beam of 0.5 A, having a length of 2 m and a diameter of 5 cm. The experimental configuration is illustrated in Fig. 1. The pulsed beam of a Nd:YAG laser (pulse energy 0.7 J, pulse length 8 ns), located near the ion injection line was overlapped parallel with the ion beam in the electron cooler. Owing to the Doppler effect at the given ion velocity, the fixed laser wavelength of $\lambda_L=1064$ nm was shifted into resonance with a transition from the electron continuum band to the $n_i=81$ state of

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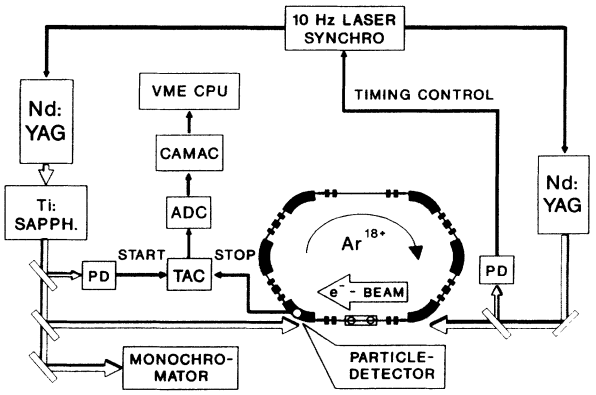

FIG. 1. Schematic view of the experimental setup for laser-induced two-step recombination at the ESR storage ring (circumference 108 m).

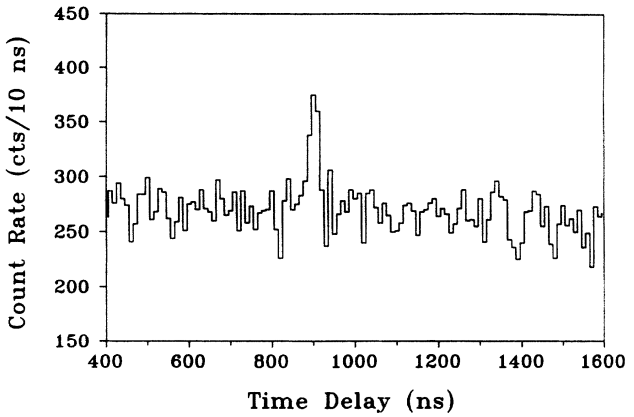

FIG. 2. Time spectrum of down-charged Ar^{17+} ions synchronized with the Ti:sapphire laser pulse, displaying the laser-induced two-step coincidence signal.

hydrogenlike Ar^{17+}, coinciding with the maximum of the capture rate calculated as a function of laser wavelength.

For the second step the beam of a Nd:YAG pumped prism-tuned Ti:sapphire laser was merged with the other beams, propagating in the opposite direction. This system was located in the laser laboratory outside the ESR concrete radiation shielding. The applied tuning range extended from about $\lambda_L = 760$ to 850 nm, corresponding to $\lambda_0 = 438.8$ to 490.7 nm in the ion rest frame.

The Ti:sapphire laser pulses had an average energy of 150 mJ, a length of 15 ns, and a linewidth of 0.45 nm. The beams of both lasers were expanded with telescopes and guided with mirrors and totally reflecting prisms, in the case of the Ti:sapphire laser over a distance of more than 60 m. Their spatial positions were monitored with removable screens, observed by video cameras, and were adjusted via motor-driven prisms. A sharp time coincidence of the laser pulses participating in the two-step process was realized in the cooler center by synchronizing both the flash lamp ignition and the Q switches of the Nd:YAG lasers. The pulse delay was monitored with a photodiode (PD) positioned immediately beside the entrance window of the Nd:YAG laser, and controlled with a fast oscilloscope. The measurements were performed with a pulse repetition rate of 10 Hz.

All down-charged Ar^{17+} ions left the Ar^{18+} storage trajectory when passing the next dipole magnet and hit a multiwire proportional counter [8]. The time spectrum of the count rate was recorded within a 2 μs time window triggered by the Ti:sapphire laser pulse. Figure 2 shows a time spectrum displaying the two-step coincidence signal. The signal peak height represents an average ratio G of induced to spontaneous rate. The signal width of ≈ 30 ns is a convolution of the time of laser interaction with ions and electrons, the propagation times of ions down charged at different sites in the cooler region, and the time resolution of the counter.

The signal disappeared when blocking either the Nd:YAG or the Ti:sapphire laser, proving the two-step nature of the recombination process under study. Tuning the Ti:sapphire laser over the resonance of the second step, detailed spectroscopy of the bound-bound transitions was possible. The line profiles of the $n_i = 81$ to $n_f = 36$ and 37 transitions as given in the laboratory rest frame are displayed in Fig. 3. The sharp on- and off-resonance behavior as a function of Ti:sapphire laser wavelength is a second proof of the two-step character of the process.

The $n_i = 81$ state possesses an l substructure, ranging from $l = 0$ to 80 and amounting to a total splitting energy (equal to the $l = 0$ lowering) of 0.14 meV. Because of the broad electron energy band (≈ 300 meV [6]), the Nd:YAG laser can populate all l sublevels, with medium sublevels favored by the transition matrix elements [9]. The $n_f = 36,37$ sublevel splittings from $l = 0$ to 35,36 are 1.6 and 1.5 meV, respectively. The $\Delta l = \pm 1$ electric dipole selection rule which would allow one to transfer only

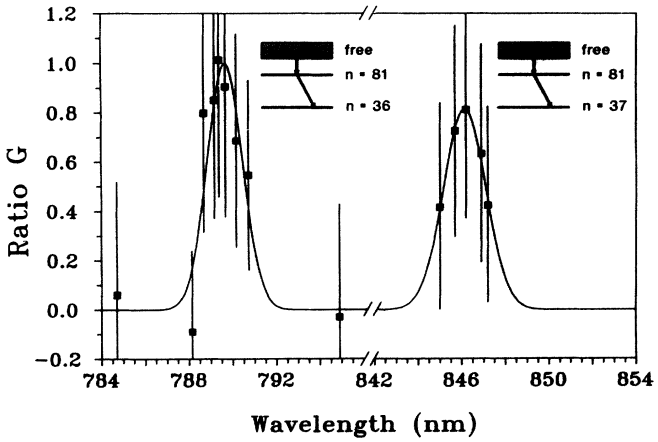

FIG. 3. Wavelength spectrum of the Ar^{17+}, $n = 81$ to $n = 36$ and 37 transitions (laboratory frame).

the $l=0$ to 36 or 37 sublevel population is weakened by mixing due to the electric and magnetic fields in the cooler.

From the signals in Fig. 3, we obtain the line centers $\lambda_{L,\mathrm{exp}}=789.8(0.5)$ nm and 846.4(0.5) nm in the laboratory rest frame. The laser wavelength tuning was measured with a monochromator whose accuracy was calibrated with tabulated atomic rubidium lines [10]. The error bars of the line centers of 0.5 nm comprise contributions of signal statistics, monochromator reading, and Ti:sapphire laser linewidth. We assume that the level energies of Ar^{17+} propagating in the cooler are not noticeably affected by the environment of the magnetized electron beam. This assumption is justified, since the influence of Zeeman splitting caused by the solenoidal magnetic field (0.11 T) and Stark splitting due to the space charge field (<100 V/cm) of the electrons is negligible compared with the experimental uncertainties. In addition, an estimate of Debye screening [11,12] of the hydrogenic Coulomb potential by the electron beam yields level shifts significantly below the size of the present error bars. Thus, identifying the experimental transition energies with the energies of an unperturbed ion, calculated by means of the fine-structure-corrected Rydberg formula, the relativistic Doppler transformation from laboratory to ion rest frame provides $\beta=0.5030(6)$. The value extracted from the acceleration voltage of the cooling electron beam, and alternatively from the ion revolution frequency and the ESR orbit length, is 0.50225(15). The deviation on the 1σ level may reflect energy shifts due to the involved static and dynamic fields and requires further investigation.

The fit curves of Fig. 3 both have a width of ≈ 2 nm, representing a convolution of several broadening mechanisms, in particular the Ti:sapphire laser linewidth, the l splittings, as well as Zeeman and Stark broadening.

In conclusion, we performed radiative recombination of Ar^{18+} ions stored in a ring with cooling electrons, applying a laser-stimulated two-step process. By tuning the Ti:sapphire laser used for the second step, the $n_i=81$ to $n_f=36,37$ transition lines of hydrogenlike Ar^{17+} were resolved. It is planned to extend the experiments from Ar^{18+} to ions with higher nuclear charge. The two-step scheme seems particularly appropriate to investigate the effect of Debye screening of ions by cooling electrons. By using a narrow-band version of the Ti:sapphire laser, even small shifts of the Rydberg levels can be resolved. Replacing the fixed-frequency Nd:YAG laser by a tunable light source, e.g., an optical parametric oscillator operating in the infrared, would allow probing the structure of the continuum-bound state transition region.

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