Observation of Laser-Induced Recombination in Merged Electron and Proton Beams

U. Schramm, J. Berger, M. Grieser, D. Habs, E. Jaeschke, G. Kilgus, D. Schwalm, and A. Wolf
Physikalisches Institut der Universität Heidelberg and Max-Planck Institut für Kernphysik, D-6900 Heidelberg, Federal Republic of Germany

R. Neumann
Gesellschaft für Schwerionenforschung, D-6100 Darmstadt, Federal Republic of Germany

R. Schuch
Manne Siegbahn Institute, S-10405 Stockholm, Sweden
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The first observation of laser-induced recombination, performed with merged beams of protons, electrons, and laser photons in an ion storage ring, is reported. The process was used to study the photorecombination spectrum with high resolution. Deviations from earlier theoretical predictions turned out to be due to a weak external electric field and could be well described by a simple extension of the field-free quantum-mechanical theory, based on properties of the classical electron trajectories in the combined external and Coulomb fields.

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The recombination of a positive ion with an electron in a binary collision is made possible by the spontaneous emission of a photon. This radiative recombination (RR) process is usually treated [1] as the inverse to photoionization. The basic theory of radiative transitions predicts that the recombination rate can be enhanced in an external resonant light field by stimulated photon emission. The concept of stimulated radiative recombination in a strong laser field was proposed for positronium formation [2], treated theoretically for the recombination of electrons and ions [3,4], and discussed with regard to antihydrogen formation [5] and to short-wavelength lasers [6]. However, a direct observation of the enhancement of the recombination rate by external radiation has not yet been reported.

In the experimental investigation of RR great progress was made by using merged electron and ion beams. Thus, the formation of hydrogen atoms was observed [7–9] during electron cooling [7] of a proton beam in a storage ring, where the neutral atoms leave the ring tangentially. Up to now, however, only the total recombination rate was measured, which represents the average over the center-of-mass (c.m.) energy distribution of the electrons and the sum over a large number of final states. Information about the recombination rate into specific final states and about the electron energy distribution is contained in the spectrum of the photons emitted in the RR process; however, owing to its low intensity, this radiation could not yet be investigated using merged beams.

A merged-beam arrangement with an electron cooler in a storage ring offers favorable conditions for the detection of laser-induced recombination (LIR) [5]. The induced recombination rate as a function of the laser wavelength represents the photorecombination spectrum which by this method can be measured with high resolution (owing to the narrow bandwidth of the laser) and high sensitivity (owing to the complete detection of the recombination products). Thus, the c.m. energy distribution of the electrons can be determined if, as in many cases, it is narrow compared to the energy spacing of the final states. Moreover, the influence of external perturbations on the recombination process should manifest itself at energies close to the ionization limit of the recombined atom. In particular, external fields can lower the ionization threshold; or the population of weakly bound states, caused by ternary collisions, might show up in the recombination signal if transitions to lower states are induced by the laser. Besides using it for studying the photorecombination spectrum, one can also consider application of LIR for enhancing weak recombination processes and for producing the recombined system in a well-defined final state.

Here we describe the observation of laser-induced radiative recombination in merged beams of protons, electrons, and laser photons, performed at the ion storage ring TSR in Heidelberg [10]. LIR was observed by sending intense laser light pulses at the Balmer-series limit into the overlap region of electrons and protons, and measuring the increase of the hydrogen-formation rate due to stimulated free-bound transitions to the level with principal quantum number $n=2$. Up to 5×10^9 protons of 21.0 MeV were stored in the ring of 55.4-m circumference for several hours. In the electron cooler [11] the protons were merged over a length of 1.5 m with a velocity-matched electron beam (energy of 11.4 keV) of a current of 0.917 A and a diameter of 5.1 cm. The electron beam was immersed in a longitudinal magnetic field of 30.2 mT in order to counteract the electron space charge. At an electron density of 4.53×10^7 cm^{-3}, the space charge gave rise to a radial electric field vanishing on the beam axis and increasing linearly towards the electron-beam edge up to a strength of 100 V/cm. By

Coulomb collisions in the overlapping beams, the protons acquired the same average velocity as the electrons and, moreover, a beam diameter of only ≈ 2 mm and a longitudinal velocity spread below 10^{-4}. The precise proton velocity in the interaction region was 0.2081(2) in units of the velocity of light and varied during the experiment by less than $\pm 2\times 10^{-4}$, as verified by observing the proton revolution frequency at a Schottky noise pickup.

In the steady state, the relative velocity between protons and electrons is essentially determined by the electron velocity in the beam rest frame. Mainly based on earlier dielectronic-recombination measurements [12], a transverse velocity spread corresponding to the cathode temperature of 1220 K (0.105 eV) and a longitudinal velocity spread corresponding to an energy of $\approx 1\times 10^{-3}$ eV was expected. The recombined atoms were separated from the proton beam in a bending magnet about 5 m downstream of the electron cooler. Behind this magnet they had to pass through a 0.5-mm-thick quartz plate, serving as a mirror for injecting laser light antiparallel to the proton beam, and were then detected with an efficiency of more than 96% on a 40-mm-diam microchannel plate, covered by a suitable degrader foil. The observed spontaneous-recombination rate was in good agreement with a theoretical estimate [13,14] of the rate coefficient $\sum_n \langle v\sigma_n(\mathbf{v})\rangle$. In this expression σ_n denotes the recombination cross section into level n, and $\mathbf{v}$ the electron c.m. velocity; the brackets indicate the average over the c.m. velocity distribution, and the sum extends over those final states in which the hydrogen atoms are not field ionized in the storage-ring dipole ($n \leq 5$) and can thus reach the detector region. The formation rate of hydrogen atoms by electron capture of protons in the residual gas was negligible.

Light pulses from a tunable dye laser (coumarin 2), pumped by a XeCl excimer laser, were focused to a cross section of ≈ 0.1 cm^2 in the interaction region. From the proton-beam velocity, one determines the laboratory wavelength corresponding to the (field-free) binding energy of the hydrogenic $n=2$ state to be 450.46(7) nm. The laser pulses were ≈ 20 ns long and had a peak power of 1–3 MW at the dye-laser exit. The laser power was continuously monitored via the pulse height from a fast photodiode, hit by light reflected from one of the optical elements, and calibrated versus the average laser output. A value of 0.7 is estimated for the transmission of the optical elements between the laser and the interaction region. The laser beam was aligned with the neutral hydrogen beam formed by RR, whose position was measured by scrapers outside of the interaction region. This procedure ensured the overlap of the laser beam with the proton beam in the interaction region itself since the neutral beam, unlike an ion beam, is not deflected in the magnetic field of storage-ring elements.

The hydrogen atoms formed by LIR were detected in coincidence with the laser pulses (repetition rate of ≈ 10 s^{-1}) in a window of 1 μs. The time spectrum (Fig. 1) shows a flat background due to spontaneous recombination and a large enhancement in a time interval corresponding to the passage of the laser pulse through the overlap region. The width of this LIR peak approximately agrees with the sum of the traveling time of a hydrogen atom through the overlap region (25 ns) and the laser pulse length. A gain factor G, representing the induced-recombination rate normalized to the spontaneous rate, was obtained by taking the ratio of the number of counts in the LIR peak to the number of background counts in a time interval equal to the laser pulse duration. Making use of the observed linear dependence of the gain factor on the laser power, G is normalized to an intensity of 17.5 MW/cm^2. It is presented in Fig. 2 as a function of the difference ϵ between the photon energy E_γ in the c.m. system and the field-free ionization threshold of the hydro-

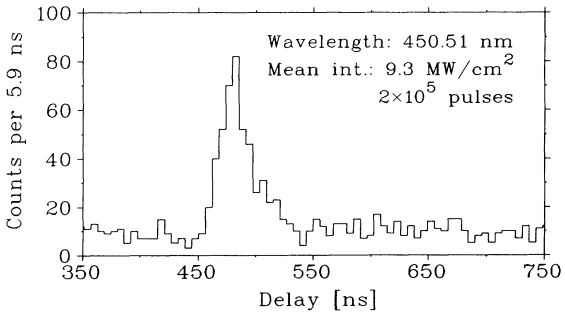

FIG. 1. Time spectrum of recombined hydrogen atoms observed in coincidence with the laser pulses for 3×10^9 stored protons.

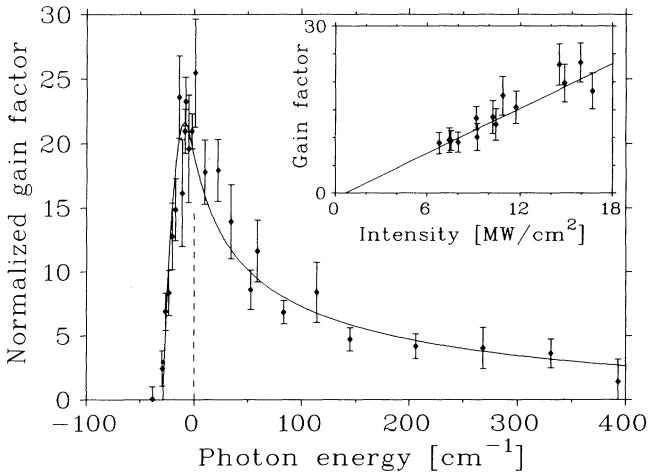

FIG. 2. Normalized gain factor for laser-induced recombination as a function of the c.m. photon energy. Solid line: Fit for external electric field, yielding $\epsilon_{sp} = -25.1 \pm 3$ cm^{-1}. Inset: Maximum gain factor (fixed photon energy) as a function of the laser intensity with fitted straight line.

genic $n=2$ level ($E_0=27420$ cm^{-1}). Via the Doppler effect, the uncertainty in the absolute beam velocity was the principal contribution (± 4.3 cm^{-1}) to the systematic error of ϵ. The uncertainty of the laser calibration corresponded to less than ± 1.5 cm^{-1}, whereas the spectral resolution was limited by the spread and the fluctuation of the proton velocity, corresponding to a bandwidth below ± 1 cm^{-1}. The gain factor $G(\epsilon)$ starts to rise already at -25.5 cm^{-1}, clearly below the expected field-free threshold. It reaches a maximum of 23 ± 2 cm^{-1} close to this threshold, falls off to half maximum within ≈ 52 cm^{-1}, and then further decreases with ϵ at a smaller rate.

The gain factor as a function of the laser intensity (see inset of Fig. 2) should saturate if the probability for recombining atoms to become reionized by the same laser pulse approaches unity. Atoms formed in the $2p$ state decay to the ground state with a mean lifetime of 1.6 ns; their ionization rate reaches [5] the inverse of this lifetime at an intensity of ≈ 20 MW/cm^2. For atoms formed in the $2s$ state the interaction time is given by the laser pulse length of 20 ns; accordingly, LIR into this subshell saturates at ≈ 2 MW/cm^2. Since the laser intensity was varied above this value, the $2s$ contribution was always saturated and the observed linear increase of the gain factor is due to the $2p$ contribution only.

In order to understand the measured photorecombination spectrum we first consider LIR in a pure Coulomb field [5], denoting by E_e the asymptotic kinetic energy of an incident electron. The spectral shape is given by the energy distribution of the electrons, multiplied by the recombination cross section [$\sigma_n(E_e)\propto 1/E_e$] and by v ($\propto E_e^{1/2}$). Putting $E_e=E_\gamma-E_0\equiv\epsilon$, one obtains for $\epsilon\geq 0$,

$$G_{nl}^0(\epsilon)=G_{nl}^0(0)\frac{\sqrt{\pi}}{2}\left(\frac{k_BT_\parallel}{\epsilon}\right)^{1/2}\times\exp\left(-\frac{\epsilon}{k_BT_\perp}\right)\mathrm{erf}\left[\left(\frac{\epsilon}{k_BT_\parallel}\right)^{1/2}\right], \quad (1)$$

and $G_{nl}^0(\epsilon)=0$ for $\epsilon<0$, where the last two factors in Eq. (1) are proportional to the electron energy distribution for an anisotropic Gaussian velocity distribution with $T_\parallel\ll T_\perp$. Taking into account the specific transition probability into the subshell nl and the effect of field ionization on the RR rate into levels n', the maximum gain is given by [5]

$$G_{nl}^0(0)=\frac{Ic^2}{8\pi\nu^3}\frac{2}{\pi k_B(T_\perp T_\parallel)^{1/2}}\frac{g_0(nl)}{n}\left[\sum_{n'=1}^{5}\frac{g_0(n')}{n'}\right]^{-1}, \quad (2)$$

where ν is the laser frequency, and $g_0(nl)$,$g_0(n')$ are the Gaunt factors for a given (sub)shell at $E_e=0$ [Eqs. (20) and (21) and Table 1 of Ref. [14]].

The function $G_{nl}^0(\epsilon)$ was fitted to the data points at $\epsilon\geq 0$, yielding a longitudinal thermal energy of $k_BT_\parallel=2.0(6)$ meV and a transverse thermal energy of $k_BT_\perp=0.08(5)$ eV in reasonable agreement with the expectations. Using these temperatures a gain factor of $G_{2p}^0(0)=59$ was calculated for the intensity of $I=17.5$ MW/cm^2. The $2s$ contribution, obtained at the saturation intensity of 2 MW/cm^2, only amounted to $G_{2s}^0(0)=2.5$; this shows that its influence on the normalization of the data in Fig. 2 could be neglected. The observed maximum gain (see Fig. 2) is, however, much below the expected sum of $G_{2p}^0+G_{2s}^0$. Moreover, according to Eq. (1) the recombination spectrum should not extend below $\epsilon=0$.

We explain these deviations by the influence of external static fields on the hydrogenic ionization limit. From measurements of the proton revolution frequency and of the hydrogen beam position, a small displacement of the proton beam from the electron-beam axis by 5 ± 1 mm can be inferred. Thus, an electric field of $F=20\pm 4$ V/cm, due to the electron space charge and directed radially towards the electron-beam axis, was present in the interaction region. The linear potential of this external field exceeds the Coulomb potential at a distance $\gtrsim 1$ μm from a proton. It leads to a saddle point of the potential at $\epsilon_{sp}=-27.4\pm 3$ cm^{-1} and reduces the ionization threshold of the hydrogenic $n=2$ level by $|\epsilon_{sp}|$, in good agreement with Fig. 2. The perturbation due to the magnetic field can be neglected since for the present field strength, the diamagnetic potential, rising quadratically, reaches the size of the electric potential only at a much larger distance from the proton ($\gtrsim 80$ μm).

We are not aware of theoretical calculations concerning photorecombination in an electric field. However, photoionization of low-lying states of hydrogen in an electric field has been investigated both theoretically [15,16] and experimentally [17,18]. In the theoretical models a complete quantum description is used only for the radiative transition occurring close to the nucleus, within the atomic radius of the low-lying state. Once the electron has been excited, a semiclassical description of its motion in the superimposed Coulomb and linear potentials is adequate. In particular, the calculation of classical trajectories [16] shows that the threshold excitation energy E_t at which an electron can just escape from the Coulomb potential is a function of the angle θ between the electron trajectory close to the nucleus and the electric field:

$$E_t(\theta)=E_0-\epsilon_{sp}\sin(\theta/2). \quad (3)$$

Along the equipotential surface for this threshold energy, the kinetic energy of electrons is given in terms of the photon energy by $E_e(\theta)=E_\gamma-E_t(\theta)$ instead of by $E_\gamma-E_0$, valid in the field-free case. Thus, we proceed by using the modified kinetic energy $E_e(\theta)$ in Eq. (1). The average over the solid angle covered by the variation of θ

then yields the spectral shape

$$G^F_{nl}(\epsilon) = \tfrac{1}{2}\int_0^\pi d\theta \sin\theta G^0_{nl}[\epsilon - \epsilon_{sp}\sin(\theta/2)]\,. \quad (4)$$

Using this simple approach, a surprisingly good fit to the data is obtained (solid line in Fig. 2). Moreover, the field-free maximum gain of 60 ± 3 obtained in the fit agrees much better with the predicted value [Eq. (2)] of $G^0_{2p}(0) + G^0_{2s}(0)$. The longitudinal temperature $k_B T_\parallel = 1.5(5)$ meV is slightly lower than the value given above and thus closer to the expectations.

In conclusion, clear evidence of laser-induced electron-proton recombination was observed and the process was used to study the photorecombination spectrum with high resolution. The observed spectral shape reflects the energy distribution of the electrons in the c.m. frame of the protons and reveals the effect of a weak electric space-charge field. It is planned to extend the LIR studies of photorecombination spectra to highly charged ions in order to investigate the influence of external perturbations, such as the deformation of the Coulomb potential by Debye screening of the ions in the electron beam, or the population of high Rydberg states by ternary recombination mentioned above.

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