Resonance Ionization Spectroscopy in a Buffer Gas Cell with Radioactive Decay Detection, Demonstrated Using ^{208}Tl

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An ultrasensitive laser spectroscopic method has been developed to perform hyperfine spectroscopy of heavy-ion-induced reaction products. It is based on resonance ionization in a buffer gas cell combined with radioactive decay detection. The feasibility has been demonstrated using the β-active isotope ^{208}Tl. A sensitivity of 1.3×10^{-3} of the two-step resonance-ionization process via the $n=17$ Rydberg level has been determined, utilizing a pulsed excimer-dye-laser combination. The previously unknown nuclear magnetic moment $\mu(^{208}\mathrm{Tl})=0.292(13)\mu_N$ (μ_N is the nuclear magneton) and a rms radius difference between ^{208}Tl and ^{207}Tl of $\delta\langle r^2\rangle=0.099(15)$ fm^2 were deduced from the $6p\,^2P_{1/2}$–$7s\,^2S_{1/2}$ transition.

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Optical hyperfine spectroscopy with modern tunable narrow-band lasers has been proven to be a very effective tool for the investigation of ground-state properties of unstable nuclides far off stability. Nuclear spins, electromagnetic moments, and the change of the rms radius have been measured for long chains of isotopes (see, e.g., the recent review article by Otten [1]). Most of the results were obtained Doppler free in collinear geometry of the laser and a fast atomic beam. A flux of about 10^4–10^6 atoms/s is required for such a fluorescence-light spectroscopy experiment. The nuclides are usually produced by spallation reactions induced, e.g., by a high-energy proton beam. However, also of potential interest are short-lived isotopes far off stability produced by heavy-ion-induced nuclear reactions. We particularly mention in this respect transuranium nuclides which cannot be produced by spallation reactions, and refractory elements for which atomic beams cannot be prepared. An ultrasensitive method is required for the hyperfine spectroscopy of such isotopes since the production rate in heavy-ion reactions of interest often lies well below 10^4/s. Following this scheme, progress has been made recently by Sprouse *et al.* [2] who studied isotope shifts of ytterbium isotopes via fluorescence-light spectroscopy in an optical cell on line at the Stony Brook tandem-linac heavy-ion accelerator facility with a production rate as low as 900/s.

In this paper we report on our approach to the above-outlined problem. We will show that radioactive decay detection combined with resonance ionization spectroscopy (RADRIS) in an optical buffer gas cell also may open new prospects for on-line laser spectroscopy at low-energy heavy-ion accelerators. This method combines both very sensitive resonance ionization with pulsed lasers and low-background detection of the resonantly ionized species by means of their nuclear radiation. In addition, the utilization of a high-resolution α or γ spectrometer permits even the identification of a specific isotope or of an isomeric state. Advantage is taken of the facts that (i) about a fraction of 13% of the ions recoiling after a nuclear reaction from the target into the buffer gas neutralize while the slowing down proceeds [3] and (ii) the resulting atoms remain stored for about 50 ms in the laser illuminated domain before diffusing out. The latter offers the possibility for several chances to resonantly ionize the atom if a modern pulse laser system with a repetition rate greater than 100/s is employed. Consequently, a very high resonance-ionization efficiency of the order of unity may be achieved with laser beams of mJ/cm^2 pulse energy per unit area.

In order to prove the feasibility of such a scheme we carried out a laboratory RADRIS experiment using the β-active isotope ^{208}Tl ($T_{1/2}=183$ s), as shown in Fig. 1. Some preliminary results of this experiment have been reported [4]. The ^{212}Pb source was prepared by chemical precipitation of the lead daughter from the equilibrium activity of a 10-μCi ^{228}Th mother substance in aqueous solution. The ^{212}Pb source with an activity of typically 10^5 Bq is β active and decays with a half-life time of 10.6 h to ^{212}Bi. The ^{208}Tl recoil ions which leave the source after α decay of ^{212}Bi with an efficiency of up to 100% are slowed down in the argon buffer gas at a pressure of 28 mbar and come to rest in the domain just between the first electrode and the source. All ^{208}Tl recoils which remained positively charged after the slowing down process (approximately 85%) will be pushed back to the source electrode by applying electrical potentials on the electrodes as indicated in Fig. 1. Part of the remainder of the (neutral) ^{208}Tl atoms diffuse through the aperture of the first electrode; the fraction of diffused ^{208}Tl was estimated by solving numerically the diffusion equation. These atoms can be ionized resonantly by the laser beams

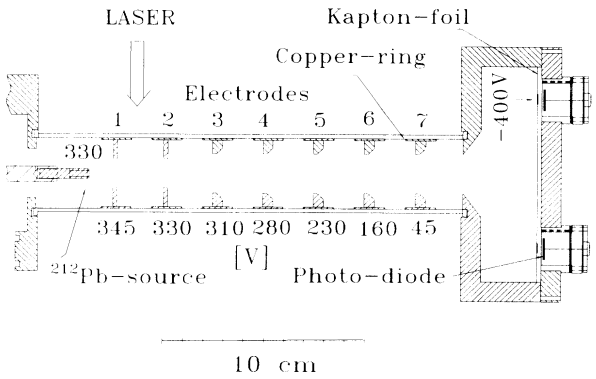

FIG. 1. Experimental setup. The ^{208}Tl ions emerging from the ^{212}Pb source are stopped in the argon buffer gas (28 mbar) and come to rest just in front of the first electrode which produces a potential maximum. Only neutral atoms diffuse into the domain between the first and the second electrode. Ions originating from the resonance-ionization process move along the electrical field lines to the metalized area on a Kapton foil in front of that one of the eight photodiode counters which was activated by a voltage of −400 V. As β counters, Hamamatsu photodiodes of the type S-3590-04 have been used with an area of 1×1 cm^2 and a thickness of 300 μm.

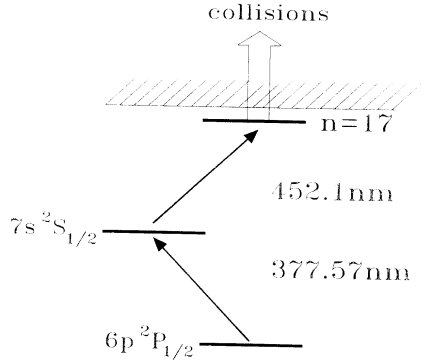

FIG. 2. Resonance-ionization scheme. Ionization of the $17p$ Rydberg state occurs by buffer gas collisions.

which illuminate a part of the region between the first and the second electrode. The created ^{208}Tl ions are transported in the electric field generated by the electrode system. They stick on the metalized area of the cover foil in front of one of the eight β counters, which are activated by high voltage.

Each of the eight counters is assigned to a certain laser wavelength, i.e., an activated counter detects only decay electrons from atoms resonantly ionized at a given wavelength setting of the dye laser. The laser wavelength and the activation voltage in front of the detector are changed simultaneously after a counting interval of typically 20-s duration. A cycle which involves all eight counters lasts 184 s. The counting is done through ten cycles, ensuring that long-term intensity changes of the laser are reduced to some mean value. The wavelength assignments of the counters were altered after the end of the ten-cycle interval, with the exception of one which was kept at the previous value for the purpose of checking the consistency of the measurements.

The resonance-ionization scheme is shown in Fig. 2. Two photons with a wavelength of 377.57 and 452.1 nm have to be absorbed to excite the ^{208}Tl atoms from the $6p\,^2P_{1/2}$ ground state via the $7s\,^2S_{1/2}$ intermediate state into the $n=17$ Rydberg state, from which the actual ionization process occurs without radiation absorption by collisions with the buffer gas atoms. The laser beams were prepared with two Lambda Physik FL 2001 lasers by using the dyes BBD ($\lambda=377.57$ nm) and Coumarin 47 ($\lambda=452.1$ nm). Both lasers were equipped with intracavity etalons (finesse $F^*=25$) to reduce the bandwidth to about $\Delta\nu=1.2$ GHz. The dye lasers were pumped with a Lambda Physik EMG 104 MSC excimer laser at a wavelength of 308 nm. The experiments were performed at a repetition rate of 200 Hz.

During the course of the experiment the frequency and bandwidth stability of the laser was checked as a matter of routine by scanning a reference cell signal. For that purpose a buffer gas quartz cell was built which was loaded with a small sample of natural thallium and kept at a temperature of about 600 K. The charge which originated from resonance ionization of the thallium vapor atoms in an individual laser pulse was collected on two plane-parallel stainless-steel electrodes (area 5 cm × 1 cm, separation 1 cm) and integrated by means of a charge sensitive preamplifier. The electrical field strength was chosen at the low value of 50 V/cm in order to avoid Stark shifts and splittings of the Rydberg levels.

The choice of the $n=17$ Rydberg state was the result of a careful investigation of the resonance-ionization signal with this buffer gas reference cell. During the course of these experiments the laser was operated without intracavity etalons at a bandwidth of about 4–6 GHz. The pulse energy flux of the $\lambda=452.1$ nm beam amounted to about 700 μJ/cm^2. Up to $n=17$ we observed a strong increase of the resonance-ionization signal with decreasing main quantum number n of the Rydberg level. At $n=17$ the signal turned out to be a factor of about 40 larger than the observed signal when the laser frequency was tuned close to the ionization limit. At still smaller n values the signal is expected to decrease since the ionization probability due to buffer gas collisions drops off steeply. However, for $n=17$, Niemax [5] reported an ionization probability of still 50% for rubidium in 16-mbar neon. In Fig. 3, scans of the $^2P_{1/2}$-$^2S_{1/2}$ transition are presented.

The sensitivity S of RADRIS, defined as the ratio of the numbers of detected β particles and of ^{208}Tl recoils implanted into the buffer gas, was determined from a high intensity scan of the $^2P_{1/2}\rightarrow{}^2S_{1/2}$ transition with a pulse energy flux of 0.9 μJ/cm^2. It turned out to be $S=1.3\times10^{-3}$. This experimental result is in accord with the product of (i) the neutralization efficiency assumed to be the same as for ^{242}Am in argon, $\epsilon_{\text{neut}}=0.13$ [3], (ii) the calculated diffusion efficiency $\epsilon_{\text{diff}}=0.12$, (iii) the

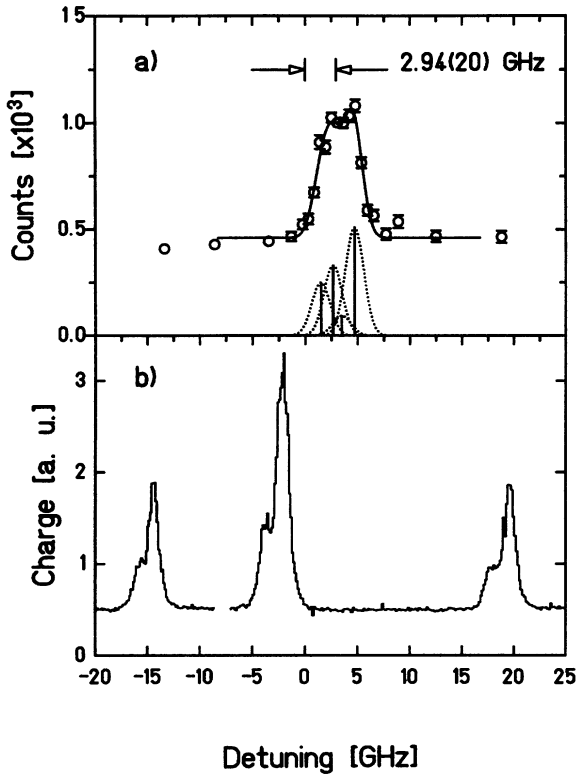

FIG. 3. Resonance-ionization signal as a function of the detuning of the first step (a) for ^{208}Tl with the experimental setup shown in Fig. 1 and (b) for ^{203,205}Tl in a reference cell. The $\lambda=452.1$ nm transition was kept on resonance with the energy flux of 1 mJ/cm^2. A low pulse energy flux of 0.08 μJ/cm^2 was chosen for the $\lambda=377.57$ nm transition in order to avoid power broadening. The quoted number indicates the isotope shift $\delta\nu^{(208,205)}$ between the isotopes ^{208}Tl and ^{205}Tl.

measured transport efficiency of the resonantly ionized ^{208}Tl atoms, $\epsilon_{\text{trans}}=0.7$, (iv) the measured detection efficiency of the β detectors, $\epsilon_{\text{det}}=0.2$, and finally (v) the resonance-ionization efficiency for which, to be consistent with the experimental result, the large value $\epsilon_{\text{RIS}}=0.6$ must be assumed. The biggest losses in the sensitivity result from the neutralization and the diffusion efficiencies which, probably, cannot be avoided in a simple manner.

The previously unknown nuclear magnetic moment of ^{208}Tl can be extracted from the broadened structure of the low intensity scan [see Fig. 3(a)]. A nuclear spin $I=5$ was assumed for the $(\pi 3s_{1/2}^{-1}\otimes\nu 2g_{9/2})$ ground-state configuration of ^{208}Tl in accord with Nordheim's rule [6]. The assumption $I=4$ gives a magnetic moment totally at variance with the extreme single-particle model. In the analysis procedure, the ratio of the hyperfine constants $A_{^2S_{1/2}}/A_{^2P_{1/2}}=0.576$ was kept constant at the experimental value of ^{205}Tl, i.e., a possible hyperfine anomaly was neglected. Four hyperfine components contribute to the observed line profile; these originate from transitions between the $F=9/2$ and 11/2 hyperfine levels of the $6p\,^2P_{1/2}$ and the $7s\,^2S_{1/2}$ terms. In principle, the intensity ratios of the four hyperfine components are given by the $6j$ coefficients of the transition matrix elements. However, the ionization probabilities of the $F=9/2$ and 11/2 hyperfine levels of the $^2S_{1/2}$ term depend also on the frequency tuning of the second laser beam. This fact was also taken into consideration in the analysis procedure. The hyperfine components were approximated by Gaussian functions as suggested by scans in the reference cell under comparable experimental conditions. The four-parameter fit yielded a hyperfine constant $A_{^2P_{1/2}}(^{208}\text{Tl})=0.377(26)$ GHz, from which the nuclear moment $\mu(^{208}\text{Tl})=0.292(13)\mu_N$ (μ_N is the nuclear magneton) was deduced, assuming $A_{^2P_{1/2}}(^{205}\text{Tl})=21.31083$ GHz [7] and $\mu(^{205}\text{Tl})=1.638\mu_N$ [8].

We discuss our experimental value of the magnetic moment in the framework of the extreme single-particle (sp) model. In the stretched configuration resulting in $I=5$ the magnetic moment turns out to be the sum of the magnetic moments $\mu(^{207}\text{Tl})=1.876(5)\mu_N$ [9] and $\mu(^{209}\text{Pb})=-1.4735(16)\mu_N$ [10] of ^{207}Tl and ^{209}Pb, respectively, which yields $\mu_{\text{sp}}(^{208}\text{Tl})=0.403(7)\mu_N$. The experimental value deviates by $-0.110(27)\mu_N$ from the sp value. The hyperfine anomaly cannot be responsible for this effect since it contributes according to the semiempirical Moskowitz-Lombardi rule only by $0.002\mu_N$. Also core polarization effects cannot be invoked as an explanation since the experimental sp values already incorporate such contributions and blocking effects are expected to be of no importance. However, it was pointed out by Vergados [11] that the dominating $(\pi 3s_{1/2}^{-1}\otimes\nu 2g_{9/2})5^+$ configuration may have admixtures of other configurations in which close-by proton orbitals are involved, in particular $\pi 2d_{3/2}^{-1}$ and $\pi 2d_{5/2}^{-1}$. The $\pi 2d_{3/2}^{-1}$ orbital, with a small magnetic moment $\mu=0.156\mu_N$ [11], can also couple with the $g_{9/2}$ neutron to yield $I=5^+$ resulting in a negative magnetic moment $\mu=-1.31\mu_N$. As a matter of fact, a 25% admixture of the $|\pi 2d_{3/2}^{-1}\otimes\nu 2g_{9/2}\rangle$ wave function to the dominating ground-state part would explain the reduction of the magnetic moment. Vergados [11] quoted a 19.5% admixture in qualitative accord with this finding.

Finally, the isotope shifts $\delta\nu^{(208,205)}=2.94(20)$ GHz between ^{208}Tl and ^{205}Tl and $\delta\nu^{(205,203)}=1.566(17)$ GHz have also been determined from the measurement shown in Fig. 3. The differences in the centers of gravity of the former measurement have been corrected for a pressure shift contribution $\delta\nu_p$ originating from the fact that the measurement on ^{208}Tl has been performed in 28-mbar argon at room temperature, while that on ^{205}Tl was done in 5-mbar Ne at 600 K. We used $\delta\nu_p=-10.8(4)$ MHz/mbar [12] and $\delta\nu_p=-2.10(56)$ MHz/mbar [13] at $T=293$ K for argon and neon, respectively. With the known difference in the rms nuclear charge radius $\delta\langle r^2\rangle^{205,203}=0.115(3)$ fm^2 [14] and the isotope shifts for the $^2P_{3/2}$-$^2S_{1/2}$ transition $\delta\nu^{(205,203)}=1.757(2)$ GHz and

$\delta\nu^{(207,205)} = 1.783(3)$ GHz [9], we obtain

$$\delta\langle r^2\rangle^{208,207} = \delta\langle r^2\rangle^{205,203}\{(\delta\nu^{208,205}/\delta\nu^{205,203})_{^2P_{1/2}\text{-}^2S_{1/2}} - (\delta\nu^{207,205}/\delta\nu^{205,203})_{^2P_{3/2}\text{-}^2S_{1/2}}\} = 0.099(15)\ \text{fm}^2.$$

Mass effects which contribute less than 0.06 GHz have been neglected in this analysis.

The rms radius change for ^{208,207}Tl agrees within the errors with the value $\delta\langle r^2\rangle^{209,208} = 0.0937(9)$ fm^2 [10] for ^{209,208}Pb. This result is at variance with the mentioned configuration mixings in ^{208}Tl [11]. Calculations of the rms charge radii in a Woods-Saxon potential [15] for $A = 15$ yield 33.546 and 32.539 fm^2 for the $\pi 3s_{1/2}$ and $\pi 2d_{3/2}$ orbitals, respectively. A 25% admixture of the $(\pi 2d_{3/2}^{-1} \otimes \nu 2g_{9/2})5^+$ configuration to the $(\pi 3s_{1/2}^{-1} \otimes \nu 2g_{9/2})5^+$ ground-state configuration would therefore change the rms radius by -0.063 fm^2 in contradiction to the experimental observation. Although the $\pi 2d_{3/2}^{-1}$ proton may polarize the lead core differently from the $\pi 3s_{1/2}^{-1}$ proton, resulting in deformation, we feel that the configuration mixing model fails to explain both the reduction of the magnetic moment of ^{208}Tl and the rms radius change between the ^{208,207}Tl isotopes.

Prospects are quite good to study with this RADRIS method nuclear moments and isotope shifts of short-lived nuclides on line at heavy-ion accelerator facilities. In principle, only the radioactive ^{212}Pb source has to be replaced by the heavy-ion target. We do not expect major complications associated with the passage of the heavy-ion beam through the buffer gas. However, it could be advantageous to separate the reaction products from the primary beam prior to the implantation into the buffer gas cell. The feasibility of such a possibility has been demonstrated again in a laboratory experiment with ^{208}Tl ions from a ^{212}Pb source. After post acceleration under high vacuum conditions with the aid of a suitable electrostatic field ($\Delta U = 90$ kV) [3], the ^{208}Tl recoil ions were implanted into the buffer gas cell. A clear resonance ionization signal has been observed also in this experiment. The sensitivity amounted to $S = 5 \times 10^{-4}$.

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