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COLLISIONAL IONIZATION AS A SENSITIVE DETECTION
SCHEME IN COLLINEAR LASER-FAST BEAM SPECTROSCOPY

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ABSTRACT

State-selective collisional ionization of fast atomic beams is used to detect optical pumping. Counting of these ions is superior in sensitivity by several orders of magnitude to the conventional fluorescence detection, provided that the energy levels involved in the optical pumping process are sufficiently well separated. A straightforward application is envisaged in the collinear laser-fast beam spectroscopy of rare-gas isotopes far from stability of which only very weak beams are available from on-line isotope separators.

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1. Introduction

The collinear laser-fast beam technique [1,2] has become a standard tool for the optical spectroscopy of radioactive nuclides. Isotope shifts and nuclear moments of about 150 isotopes have been measured during the past semi-decade, most of them at ISOLDE (CERN). This facility [3] provides the nuclides from 600-MeV proton-induced reactions in the form of mass-separated 60-keV ion beams. For laser spectroscopy it is appropriate to convert these ion beams into beams of fast neutral atoms: The charge-transfer neutralization process with alkali atoms leaves the atoms in (ground or metastable) states that can be excited by the visible light of single-mode cw dye lasers [4].

The applicability limits for isotopes far from stability are determined by the sensitivity of the technique. A high efficiency of the optical excitation is inherent in the collinear beam interaction, because the narrow longitudinal velocity spread of the ions or atoms corresponds to a Doppler width of the order of the natural line width of strong optical transitions (see e.g. [2]). This also gives the high resolution. However, the detection by counting of fluorescence photons involves losses and background from scattered laser light: The limited quantum efficiency of photomultipliers and the incompatibility of high photon-collection efficiency with low stray light background (especially for the observation over an appreciable length of the beam path) leads to an overall detection efficiency of 10^{-3} - 10^{-2} on a background of typically 10^3 - 10^4 counts per second. Moreover, optical pumping prevents the multiple excitation of atoms, except for a few cases such as the resonance lines of Ba I [2] and Ra I [5]. Therefore, the standard set-up at ISOLDE has reached practical sensitivity limits between 10^4 and 10^7 atoms per second, depending on the strengths of the optical transitions, the multiplicities of the hyperfine structures and the optical pumping processes involved.

Non-optical detection schemes are considered most promising for increasing the detection efficiency and reducing the background. Among these, the stepwise laser resonance ionization with subsequent ion counting is well developed for pulsed lasers

with high power, but low duty cycle, acting on samples of thermal atoms [6,7]. With fast beams this technique gives improved isotopic selectivity, due to the large Doppler shift and narrowing [8], but considerable duty cycle losses in sensitivity. These may be minimized by the high repetition rate of copper-vapour pumped dye lasers. Single-mode cw lasers are limited in power, and attempts are being made to achieve efficient ionization by the excitation to low Rydberg states [9,10]. They have to cope with optical pumping losses and resolved hyperfine splittings in the intermediate states.

We propose a straightforward alternative which we have shown to work efficiently and reliably for at least the rare gases. It is based on the selective ionization of metastable atoms during passage through a gas target. Similar techniques were used in early collinear-beam experiments on HD^+ by Wing et al. [11] and on Ba^+ by Gaillard et al. [12] who observed small changes in the ion beam current, due to slightly different neutralization or double-ionization cross-sections for the states involved in the optical excitation and decay. On the other hand, there are examples of strongly state-selective processes, such as the near-resonant charge transfer neutralization. Thus, Silverans has shown recently [13] that the excitation of Sr^+ in the 5s-5p resonance lines, followed by the decay to the metastable 4d levels, strongly affects the beam composition of ions and neutral atoms after passage through sodium or caesium vapour.

Our subject is a reverse scheme of ion detection in spectroscopy with neutral atoms. We first discuss the relevant features of the rare-gas spectra, then describe an experimental test, and conclude by a quantitative comparison between fluorescence and ion detection.

2. Principle of the Technique

2.1 Spectroscopy on Neutralized Rare-Gas Beams

In passing through an alkali vapour, the singly-charged rare gas ions capture their last electron predominantly in the first excited $np^5(n+1)s$ configuration,

where R and A refer to the rare gas and alkali atoms, respectively. The condition of energy resonance, $\Delta E \approx 0$, is well fulfilled for the alkali element next to the rare gas in the periodic table. Assuming roughly a statistical distribution over the four fine structure components, we estimate that the beam contains about 40 % of the atoms in the metastable $(n+1)s [3/2]_2$ state. This state lies about 10 eV above the $np^6 {}^1S_0$ ground state and 4 eV below the ionization limit. It can be excited to states of the configuration $np^5(n+1)p$ by laser light in the red.

To be more specific, we shall restrict the further discussion to the example of krypton. The relevant part of the Kr I level diagram is shown in Fig. 1. There are five allowed transitions from the metastable $5s [3/2]_2$ level to levels of the configuration $4p^5 5p$, which are all readily detected by fluorescence. The highest detection sensitivity will be obtained for the 8113 Å line to $5p [5/2]_3$, due to the absence of optical pumping and the possibility of multiple excitation (apart from hyperfine pumping in the odd isotopes). In the other transitions, the decay branches to $5s [3/2]_1$ may be used to block the stray light. An even more interesting feature could be the VUV emission in the subsequent decay to the ground state. The latter may be detected efficiently with gas counters [14] of large solid angle, but, for the application to unstable nuclides, we expect serious background problems due to radioactivity.

2.2 Detection by State-Selective Collisional Ionization

The large energy difference involved in the optical pumping process from the metastable state at -4 eV to the ground state at -14 eV entails rather different ionization cross-sections in collisions with target atoms or molecules. Although the information about these cross-sections is rather scarce in the literature [15], they can be expected to be of the order $10^{-16} - 10^{-15} \text{ cm}^2$. This is sufficiently large to preserve the beam properties and to detect the ions by deflection into a counting device (secondary electron multiplier).

We have to consider two processes:

(i) The charge transfer according to

is likely to occur from the metastable rare gas atoms R^* , if the collision partner X has a high electron affinity. It would be dominant for near-resonant cases ($\Delta E \approx 0$) under near-adiabatic conditions (low beam energy). However, the electron affinities of atomic and molecular gases are usually much less than 4 eV, and the beam energies of isotope separators are beyond the near-adiabatic regime.

(ii) The stripping process

may thus be favoured. It is not subject to a resonance condition, but can be treated as the result of a random excitation of the colliding system. A semi-classical model, proposed by Firsov [16] in 1959, can be used to estimate the cross-sections and their ratio between metastable and ground-state atoms in the beam. From the formulae given in [15,16] we find $Q^*/Q_0 \approx 2.5$ for this ratio in krypton, almost independent of the collision partner for beam energies in the 10-keV range. The absolute

values of Q^* are between 5×10^{-16} and 1×10^{-15} cm². As pointed out by Bates and Massey [17], the actual cross-sections can be enhanced by crossings of the potential energy curves of the systems RX or R^*X with those of the auto-ionized complex $(RX)^+$ and an electron of zero energy. This is more probable for the high-lying metastable than for the ground state, and therefore we can take the calculated ratio Q^*/Q_0 as a lower limit. The 5p electron is by far the most loosely bound electron in the colliding system, and quenching processes are likely to be slower than stripping.

3. Experimental

To verify these assumptions, we modified the apparatus described in [2], and the essential parts are shown in Fig. 2. A 10-keV Kr^+ beam^{*)} was neutralized by charge exchange with caesium, and the remaining ions were removed by an electrostatic deflector. The optical excitation region had a total length of 60 cm of which 20 cm were observed by the conventional fluorescence photon detector. The beam then passed through a gas-target chamber with a length of 10 cm and apertures of 5 mm, pumped at both sides. The neutral atoms and the ions created in collisions were again separated by an ion deflector, and both beams could be monitored by a combined Faraday cup and secondary electron detector. The gas flow was controlled by a needle valve, and a relative pressure scale was provided by a penning vacuum meter in the pumping zone.

^{*)} Qualitatively similar results were obtained for a 60-keV beam.

4. Results and Discussion

We have studied the effect of several target gases, and measured the pressure dependence of the ionization efficiency, as well as its change upon optical pumping into the atomic ground state. The tuning of the laser frequency to the transition $5s [3/2]_2 - 5p [3/2]_2$ (7601 Å) of ^{84}Kr was monitored by the fluorescence signal.

The results have some striking general features: (i) With very few exceptions, the ionization efficiencies of various target gases are rather similar. They reach a saturation value of the order of 10 %. (ii) In all cases an appreciable flop-out signal in the ion current of 30 - 60 % is caused by depopulation of the metastable state.

Individual ionization curves for SO_2 , Cl_2 , air, He, Ar and Kr and the corresponding relative flop-out signals are summarized in Fig. 3. The result for air also stands for H_2O and a few simple organic compounds like $\text{C}_2\text{H}_5\text{OH}$. As the pressure scale is not corrected for the sensitivity of the ionization gauge to different molecules, the similarity in the shape and position of the curves indicates cross-sections which are mainly determined by the number of electrons in the target molecule. The total ion yield seems to be limited by charge-transfer neutralization,

This is supported by the trend within the sequence of the rare gas atoms He, Ar and Kr which essentially follows the ΔE dependence of the charge-transfer cross-sections. On the other hand, the electron affinity of the target molecules or atoms seems to have little influence on the ionization efficiency. From this we conclude that the stripping process

is the dominant mechanism which discriminates well between atoms in the metastable and the ground state. From the typical flop-out signal of about 40 % and the estimated metastable population of again 40 % we obtain a lower limit for the cross-section ratio $Q^*/Q_0 \geq 2.7$, in good agreement with the Firsov estimate. The actual ratio is probably larger because of non-quantitative depopulation of the metastable state, due to imperfect overlap of the beams. This large effect also shows that quenching processes during the collision play a minor role.

For the practical application it seems that SO_2 is most convenient, because of the maximum ion yield and a rather good response to the depopulation of the metastable state.

5. Sensitivity Considerations

We finally compare the sensitivities of ion and fluorescence detection. Fig. 4 shows a typical resonance curve, taken in parallel with both detectors. The rate of photons at the centre of the resonance is more than a factor of 10^3 less than the change in the ion rate. However, this cannot stand for a direct comparison, because the experimental conditions are not optimized simultaneously for both detectors: At the laser power of 2 mW, used for efficient optical pumping, the fluorescence signal is reduced, because an appreciable fraction of the atoms have reached the ground state before entering the photon-collection chamber. For optimized conditions, the fluorescence signal would be rather 10^{-4} than 2×10^{-5} photons per atom as in Fig. 4.

The following comparison is based on this optimum number for the fluorescence signal. The strength of the ion signal is taken from the present results which may be further improved by differential pumping of the target chamber. As a measure for the sensitivities, we take the weakest neutral-beam intensities for which a resonance signal can be found under the

typical running conditions of an accelerator experiment. (With a typical 50 % transmission of the beam line and the apparatus and a 50 % neutralization the primary ion production has to be four times these values.) For this limit we assume a signal-to-noise ratio of 10 achieved within an integration time of 10 s per channel.

The optical signal of 10^{-4} photons per atom has a typical background (from stray light, radioactivity and thermal noise of the photomultiplier) of 10 kHz. This corresponds to a sensitivity limit of 10^6 atoms per second. The flop-out signal in the ion current of 4×10^{-2} , relative to the original beam, will have the same statistical quality with a beam of 60 atoms per second, if there is no additional background. This corresponds to a gain in sensitivity of four orders of magnitude. Moreover, the off-resonant ion current is proportional to the beam intensity, whereas the optical signal has a fixed background. This facilitates the compensation of even lower beam intensities by longer integration time. The optical detection would be more favourable in the transition $5s [3/2]_2 - 5p [5/2]_3$, because of the absence of optical pumping in spin $I = 0$ isotopes. Multiple excitation with increased stray-light background would increase the signal-to-noise ratio by less than a factor of 10. The corresponding gain for ion detection would still be more than three orders of magnitude.

The validity of this extrapolation from 10 - 100 nA to 10^{-17} A beam currents (corresponding to 60 atoms per second) depends crucially on all possible sources of background. Serious problems can arise from contaminations of the isotope-separated beam by atomic or molecular ions with the same or a similar charge-to-mass ratio. They may originate from the rest gas in the ion source region, the carrier gas of the discharge, the ion source and target materials, or the nuclear reactions in the target. Owing to the neutralization and re-ionization of the beam, including double separation of the charge states, the

contaminants will only reach the detector, if they undergo the same changes of charge. Therefore, one can expect a considerable background rejection from a proper choice of both reaction partners: The quasi-resonant capture of electrons in the metastable states of the rare gases will usually predominate over the neutralization of parasitic beams of which also the re-ionization may be hindered by the higher ionization energy and the reverse charge transfer to target molecules. Moreover, the dissociation will eliminate molecular contaminants in the electrostatic deflector. To some extent one can avoid parasitic beams by the design of the target-ion source assembly and a good resolution of the mass separator. Tests with ISOLDE have shown that a well-outgassed UC_2 target [18] with cold transmission line to the plasma ion source gives a background of $10^4 - 10^2$ ions per second in the heavy-mass range $A > 150$. This favours a first application of the technique to the heavy rare gases radon and xenon.

For the radioactive beams, we have to consider also the background from their radioactive decay on the ion detector. It is particularly serious for the α -activities from Rn isotopes and their daughters. As in other experiments [19], this background can be eliminated by an electron multiplier cathode in the form of a moveable metallic tape. Alternatively, the radioactivity may serve for detection discriminating against all stable contaminants of the beam, if the half-lives are sufficiently short.

We are planning to use the proposed technique for an extension of our optical experiments on radon [20] into the region of more neutron-rich isotopes $A > 222$ which fall on the border of the region of a probably stable intrinsic nuclear octupole deformation [21,22]. The neutron-rich xenon isotopes will be interesting in connection with a systematic study of the development of deformation around the neutron number $N = 90$, and in particular in comparison with the neighbouring caesium and barium isotopes (see [2] and references therein).

It may be possible to extend the technique also to other elements, such as cadmium and mercury which have two-valence-electron spectra with a considerably smaller energy difference between ground and metastable states. For this purpose, one has to study in more detail the processes involved, and in particular the conditions under which the ionization is most efficient and selective.

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Figure Captions

Fig. 1: Partial energy-level diagram of Kr I. The electrons are captured (from Cs) and stripped (by collisions) predominantly in the metastable $5s [3/2]_2$ state. Laser excitation to $5p [3/2]_2$ pumps them into the ground state.

Fig. 2: Modified set-up for collinear laser-fast atomic beam spectroscopy with detection of ions created in a gas target.

Fig. 3: Ion yield of different target gases as a function of pressure. The similarity in the pressure dependences is due to the uncorrected relative scale of an ionization gauge. On the right is shown the relative flop-out signal from optical pumping to the ground state.

Fig. 4: Optical resonance in the transition $5s [3/2]_2 - 5p [3/2]_2$ of ^{84}Kr , detected simultaneously via fluorescence (lower part) and ionization (upper part).

86 098

Fig. 1

Fig. 2

Fig. 3

KRYPTON

$(5 \times 10^{11} \text{ atoms/s})$

86 097

Fig. 4