Collinear Laser Spectroscopy on Fast Atomic Beams

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In collinear geometry we have observed laser-excited, narrow resonances in fast beams of Na and Cs atoms obtained from ion beams by charge transfer collisions. Being very sensitive, the method is suited for measuring isotope shifts and hyperfine structure of isotopes far from stability provided by on-line mass separators. It may be used furthermore to study inelastic charge transfer from the energy-loss spectrum of the Doppler-shifted lines.

The high collimation and intensity of laser light sources have rendered possible resonance spectroscopy on fast beams of ions or atoms. The first experiment of this kind was performed by Andrä, Gaupp, and Wittman[1] on a Ba^+ beam crossed by the light beam of an Ar^+ laser under an angle such that the laser line was Doppler shifted to the resonance point. The advantages of using a collinear geometry instead (Fig. 1) have been pointed out by one of us in a proposal published while preparing this experiment.[2]

An important advantage is based on the velocity-bunching phenomenon occurring in accelerated beams. Consider a beam of thermal ions emitted by an ion source. If it is accelerated through a potential difference U, the velocity distribution along the beam direction is narrowed by a factor $R = \frac{1}{2}(kT/eU)^{1/2}$; for $T = 1500$ K and $U = 5$ kV, $R = 2.5 \times 10^{-3}$. The Doppler width is thus reduced to the order of the natural linewidth of typical resonance lines. This effect has been used in laser spectroscopy of HD^+ ions.[3] A different aspect of the collinear concept has been treated by Dufay *et al.*,[4] namely a Doppler-tuned saturation spectroscopy of a fast-ion beam.

Velocity bunching also increases the sensitivity since it brings all atoms simultaneously to resonance instead of utilizing just the resonant section of the Doppler spectrum. This may be seen in principle by comparison with another standard high-resolution technique, namely laser excitation of a collimated thermal atomic beam at right angles. Let the (fluorescence) signal (S) per particle be proportional to the ratio of the interaction length with the laser (l) to the particle velocity (v). If one assumes typical values of $l = 100$ mm for collinear and $l = 1$ mm for crossed geometry one may use in the former case a beam accelerated to $v = 100 v_{\text{thermal}}$ still achieving the same S as in the latter case but gaining a factor of about 100 in resolution. For obtaining the same resolution in crossed geometry, however, the beam has to be collimated to roughly 10 mrad, thus utilizing only 10^{-4} of the total flux from the oven (assuming molecular flow with an angular dependence $\sim \cos\theta$).

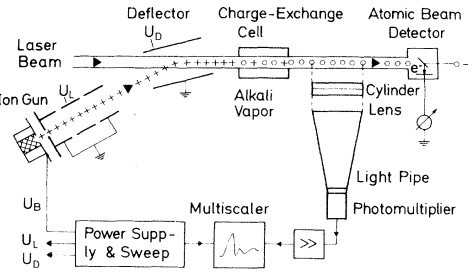

FIG. 1. Schematic diagram of experimental apparatus.

Since resonance lines of ions lie mostly in the uv which is inaccessible with present single-mode dye lasers, the range of application of fast-beam spectroscopy can be enlarged very much by converting the ion beam to an atomic beam. Effective conversion is obtained by using charge-transfer collisions.[5] Cross sections reach 10^{-14} cm^2 corresponding to impact parameters much larger than atomic radii. Consequently the momentum transfer to the target atom is negligible and the phase space of the beam is not enlarged, which has been confirmed under experimental conditions similar to ours.[6] This conclusion holds for resonant as well as nonresonant charge transfer. In the latter case the energy defect ΔE being of the order of a few eV equals almost exactly the energy loss of the fast particle because conservation laws require that in forward scattering the kinetic energy transferred to the target particle to be of the order $(\Delta E)^2/eU$, which is negligible for beam energies in the range of keV. Starting from Ref. 2, Meier *et al.* circumvented the necessity of beam neutralization realizing that substantial fractions of certain ions, e.g., noble gases, leave the source in metastable states

which are easily excited optically.[7] Similarly, charge transfer to metastalbe atomic states may also be useful; it can be achieved by choosing a suitable reaction, for example, $H^+ + Cs \rightarrow H(2s) + Cs^+$.[8]

Our experiment has been performed using beams of Na^+ and Cs^+ ions produced by ionization on a hot tungsten surface and accelerated to energies up to 10 keV. After focusing, the ion beam is deflected by a condenser and thus superimposed to the laser beam. The collinear beams cross an alkali-vapor cell in which the ions are neutralized by charge-transfer collisions. Inside the cell an alkali-vapor pressure of about 10^{-3} Torr is maintained by heating the reservoir and recycling the metal in liquid state by capillarity.[9]

The fast atoms are detected by electron emission from a metal surface. Currents ranging from 10^7 to 10^{10} atoms per second have been used. At a beam diameter of about 2 mm the maximum divergence is 5 mrad. No significant difference between ionic and atomic beam profiles is observed in accordance with the above statements.

The light source is a Coherent Radiation Model No. CR-599 dye laser, running single mode and frequency stabilized by an external temperature-controlled etalon. fm jitter as well as long-term drift of the laser frequency are considerably less than 10 MHz. Na is excited to the $3p\ ^2P_{3/2}$ state, and Cs to the $7p\ ^2P_{3/2}$, at wavelengths of $\lambda = 5890$ Å and $\lambda = 4555$ Å, respectively. Whereas the former lies in the convenient range of rhodamine 6G the latter region is still difficult to reach. Pumped by 1–2 W of uv light from an Ar^+ laser (Coherent, model No. CR18) reliable single-mode lasing at a power of some milliwatts was obtained from the dye coumarin 2. A grating monochromator controls coarse setting of the laser frequency, taking into account the large Doppler shift. Tuning is accomplished by varying the acceleration potential; for Na atoms at $U = 5$ kV the Doppler shift is 300 GHz and a tuning interval of 1 V corresponds to 34.8 MHz.

Optical resonance is detected by counting fluorescence photons. Light emitted from the beam is collected over a path length of 20 cm by a cylindrical lens focused on a light guide and transmitted to the photomultiplier cathode. During these first experiments, no special effort has been made to reduce the overall background counting rate to less than 10 kHz, resulting from laser stray light, collisional excitation of residual gas, and multiplier dark current.

The hyperfine structure (hfs) of alkalis is well known and compiled in Table I. At sufficient laser power the atoms are optically pumped from the absorbing hfs level of the ground state into the other "silent" one. Only for the transitions between the highest and the lowest F, F' values this so-called "hyperfine pumping" does not occur because of the selection rule $\Delta F \leq 1$. These are the transitions (there for Cs in brackets) $F = 2$ (4) $\rightarrow F' = 3$ (5) and $F = 1$ (3) $\rightarrow F' = 0$ (2).

In the case of Na the atoms are pumped completely into the silent state before reaching the field of view of the photomultiplier. The observable part of the hfs pattern reduces therefore to the two transitions mentioned above. Figure 2(a) shows a scan of the $F = 2 \rightarrow F' = 3$ transition for a Na beam with $U = 6.6$ kV neutralized in a K vapor. The structure observed is interpreted as the energy-loss spectrum of nonresonant charge transfer. The peak on the left is assigned to the superelastic transfer into the $3s$ state with an energy gain of 0.8 eV; the one in the middle, to the inelastic transfer into the $3p$ doublet with an energy loss of 1.3 eV. They are separated exactly by the excitation energy $E(3p) - E(3s) = 2.1$ eV. Transfer to higher excited states is present but not resolved. This interpretation is confirmed by Fig. 2(b) showing a scan obtained from a slower Na beam neutralized in a Na vapor. In this case the colliding system is approaching already an adiabatic regime favoring very much the resonant channel. Indeed the spectrum is reduced almost completely to a single line.

Figure 2(c) shows a scan of the transition group $F = 4 \rightarrow F' = 3, 4, 5$ in a 8.6-keV Cs beam neutral-

TABLE I. Compilation of hfs data for ^{23}Na and ^{133}Cs (Refs. 10 and 11).

	Ground state $^2S_{1/2}$		Excited state $^2P_{3/2}$		
	hfs states	hfs splitting (MHz)	hfs states	A factor (MHz)	B factor (MHz)
^{23}Na	$F = 1, 2$	1771.631	$F' = 0, 1, 2, 3$	18.66(14)	3.0(3)
^{133}Cs	$F = 3, 4$	9192.631 772 0	$F' = 2, 3, 4, 5$	16.609(5)	−0.16(6)

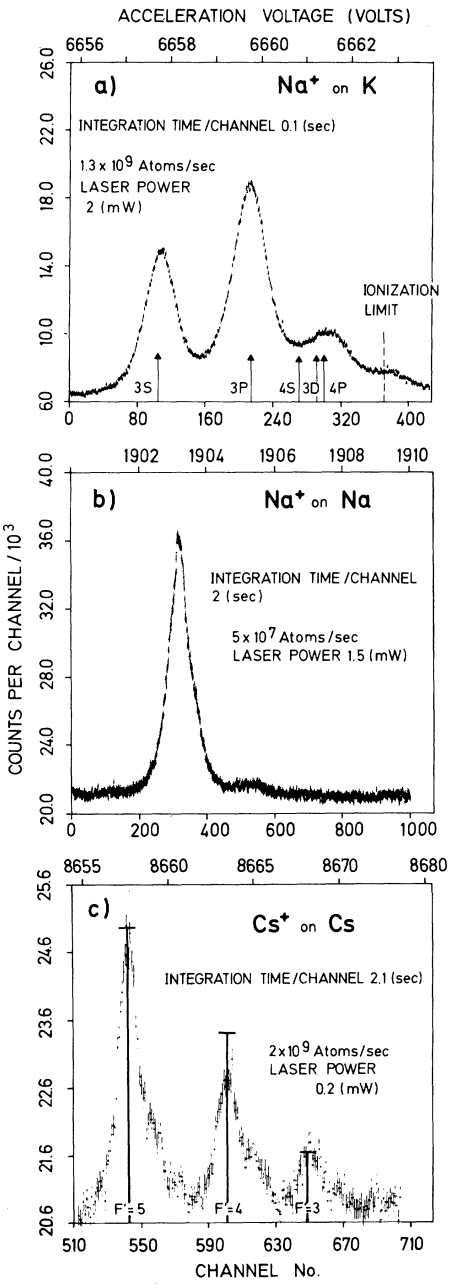

FIG. 2. Number of fluorescence photons counted as a function of the acceleration voltage.

ized in a Cs vapor predominantly through the resonant channel. Since all three components are observed, pumping is obviously weak because of the much smaller laser power and absorption strength of the transition as compared to the Na case. The relative heights and positions of the components agree with the theoretical ones drawn in. The theoretical heights have been calculated for random population of the ground-state levels and observation of the fluorescence perpendicular to the polarization axis. A preliminary evaluation yields $A = 16.6(2)$ MHz, $B = -0.4(5)$ MHz in agreement with values given in the literature. A weak but clearly visible energy-loss spectrum of charge transfer is observed at the right wing of each component.

The minimum linewidth observed is 30 MHz for Na and 16 MHz for Cs. This is about twice the value estimated in Ref. 2 for a beam of thermal energy spread and 5-mrad divergence. In most cases this will certainly be sufficient to measure isotope shifts and hfs. The resolution of the hfs may be increased further by applying additionally level crossing or double resonance. Regarding the energy-loss spectrum of charge transfer the present setup reaches a resolving power of $\Delta E = 1$ eV being sufficient for studying the dominant channels. Higher resolution requires finer collimation and energy selection of the beam. The ultimate limit is given by the natural linewidth $\Delta\nu_{\mathrm{nat}}$ and the resoance frequency ν_0 of the probing line through the relation $\Delta E = c\,\Delta\nu_{\mathrm{nat}}/\nu_0(2meU)^{1/2}$.

Finally we wish to discuss the possibility of applying this technique to unstable isotopes, especially since this was the initial purpose of this line of research. Isotope separators put on-line with a reaction target, for instance the ISOLDE at CERN,[12] can provide beams at intensities of 10^7 to 10^{10} ions per second over an isotopic range as wide as 20 mass numbers.[13] On the other hand, Fig. 2(b) shows that about 18 000 photons per second are counted from a beam as weak as 5×10^7 atoms per second, a number which obviously does not mark the lower limit. Moreover, the method is ideally adapted to the physical conditions at a mass separator since it is working directly on the separated beam (which usually covers a phase space not very much larger than quoted for this experiment). Thereby it avoids any kind of processing the isotope such as stopping it on a foil, re-evaporating it, containing the vapor in a resonance vessel, etc. That is very important since these manipulations cause serious problems in cases of chemically reactive or refractory elements. To begin with, the method will be applied to heavy radioactive Cs isotopes being produced and mass separated on line at the reactor at Mainz.[14]

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Ion-Electron Coincidence Measurements of the Azimuthal Dependence of Electrons from Autoionizing He Atoms Excited in 2000-eV He^+-He Collisions

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The azimuthal angular dependence and corresponding energy spectra have been measured for electrons ejected at 135° from the beam direction by autoionizing He atoms in the $(2p^2)^1D$ and $(2s2p)^1P$ states, which were excited by 2000-eV He^+ ions scattered through 6°. The data show a marked dependence on the azimuthal angle of the scattered projectile and are used to calculate the moduli and phases of the population amplitudes of the magnetic sublevels excited.

The excitation and decay of the doubly excited $(2s2p)^1P$ and $(2p^2)^1D$ autoionizing states of He targets has been investigated by analyzing the ejected electrons in coincidence with 2000-eV He^+ projectiles which have been scattered through 6°. The experiment samples electrons ejected at 135° with respect to the incident beam direction. By measuring the intensities and the energy spectra of coincident electrons as a function of the azimuthal angle, i.e., the angle between the scattering plane and the plane containing the beam axis and direction of electron ejection, the moduli and phases of the complex population amplitudes for the magnetic sublevels excited in the target may be deduced. The connection between excitation parameters and angular distributions of coincident electrons has recently been investigated theoretically by our group[1] and by Eichler and Fritsch[2] and the experiments reported here are the first ones to test these theories.

Prior angular distribution measurements of ejected electrons have only been done as a function of ϑ, the angle of electron ejection measured with respect to the incident ion beam, but averaged over all orientations of the scattering plane. At best, these data allow the determination of the moduli of sublevel population amplitudes.[3] The present experiment fixes ϑ at 135° and varies

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