$\pi n^4 a_0^2$, the geometrical cross section of an excited atom, are given. In Fig. 3 the experimental cross sections are plotted versus n. Fitting the argon points by a straight line implies that σ is proportional to $n^{3.8(5)}$.

The interaction responsible for the angular momentum mixing is probably the interaction of the sodium valence electron with the strong short-range potential of the rare-gas atom while it is within the region of the valence-electron wave function. This interaction is orders of magnitude stronger than the energy differences between the sodium $l \geq 2$ states, and as a result these l states are completely mixed by the presence of the rare-gas atom. Furthermore, since the interaction is so strong, it is insensitive to minor differences in the potentials of the rare-gas atoms. Consequently the cross sections for collisional angular momentum mixing should simply reflect the geometrical cross section of the excited sodium atoms for any atomic or nonpolar-molecular collision partner. This hypothesis is in agreement with our observations in that the observed cross section does increase as n^4 as does the geometrical cross section of the excited sodium atom and seems to be independent of the rare gas in spite of the fact that the polarizabilities of argon and neon differ by a factor of 5.

Here we have directed our attention mainly to collisions in which l but not n is changed. The fact that the effective decay times τ_{eff} were not noticeably changed by pressures of up to 1 Torr suggests that the radiationless quenching cross sections are < 1 Å^2.

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Infrared–X-Ray Double-Resonance Study of $2P_{3/2}$-$2S_{1/2}$ Splitting in Hydrogenic Fluorine

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Hydrogenic fluorine atoms in the $2S_{1/2}$ metastable state were produced at 64 MeV by use of a tandem electrostatic accelerator and suitable combinations of stripper and adder carbon foils. A pulsed HBr laser centered at 2382.52 cm^{-1} was Doppler-shift tuned through the $2S_{1/2}$-$2P_{3/2}$ energy separation to yield a resonance in Lyman-α x rays at 826 eV. This technique yields a value for the Lamb-shift splitting ($2S_{1/2}$-$2P_{1/2}$) equal to 3339 ± 35 GHz which is presently in agreement with several quantum electrodynamic calculations.

We have observed Lyman-α radiation at 826 eV induced via resonant absorption of infrared radiation (2328.52 cm^{-1}) produced by a pulsed HBr laser incident upon a high-energy beam of $^{19}F^{8+}$ in the metastable $2S_{1/2}$ state. The fixed-frequency laser was tuned through the $2S_{1/2}$-$2P_{3/2}$ resonance ($\Delta E - \delta$, where ΔE is the fine-structure splitting and δ the Lamb shift) by use of the Doppler shift in the frame of the particle beam ($v/c = 0.085$). This measurement for the splitting $\Delta E - \delta$ can be used to obtain a value for the Lamb shift ($2S_{1/2}$-$2P_{1/2}$) of 3339 ± 35 GHz which can be compared with quantum electrodynamic (QED) calculations[1-3] in a strong Coulomb field. The error quoted represents 1 standard deviation; possible systematic effects are still under investigation. As this new technique is refined, an order-of-magnitude improvement in precision is

anticipated.

Since the classic experiments of Lamb and Retherford,[4] and subsequent theoretical developments, agreement between the precision measurements of S and the calculations has been one of the important tests and outstanding triumphs of QED theory. The specific motivations of the present work and other recent studies include the following: (1) The present QED formalism involves calculations in the form of a perturbation series expansion in α and $(Z\alpha)$ with $(Z\alpha)^4$ the lowest-order term. The convergence of this expansion has never been established and recent attempts to obtain the Z dependence of S in closed form[1,3] have yielded results which differ somewhat from those obtained by use of the series expansion. (2) The high-energy, short-distance limits of the present QED theory have never been established. Thus the amplification of the Lamb-shift effect which derives from its strong Z dependence [$S(Z=9) \sim 3200 S(Z=1)$] offers the prospects for significant experimental tests (of various calculations) using new techniques developed in the study of highly ionized atoms and high-power lasers.

Previous experiments on high-Z hydrogenic atoms ($Z=6$, 8, and 9)[5-7] utilized the dc Stark effect to couple the $S_{1/2}$ and $P_{1/2}$ levels. The Lamb shift is obtained indirectly from the mixed-state lifetime. The present experiment utilizing resonance quenching is a return to the precision techniques used for hydrogen, helium, and lithium. A similar experiment for measuring $\Delta E - S$ in the $(\mu^4\mathrm{He})^+$ ion has recently been reported by Bertin *et al.*[8]

As in the previous high-Z Lamb-shift studies,[5-7] hydrogenic atoms are produced by post stripping a high-energy beam from an electrostatic accelerator (The Rutgers University–Bell Telephone Laboratories tandem Van de Graaff). For these experiments F^{7+} at 64 MeV was converted by stripping in a thin foil to F^{9+} atoms. The F^{9+} beam is passed through a second thin carbon foil (adder foil) from which a small fraction of the emerging atoms are in the F^{8+} metastable $2S_{1/2}$ state. The average beam current was 18 nA (9^+ particle charge) of which about 1% was estimated to be in this metastable state. Two thin-window gas-flow proportional counters located 1 m beyond the adder foil detect radiation from the particle beam. This radiation consists primarily of two-photon spontaneous emission from the hydrogenic $2S_{1/2}$ state and radiation from metastable heliumlike states present in the beam. The count rate in these detectors (~50 kHz) is much greater than the induced Lyman-α rate on resonance (~1 per pulse). However, the energy resolution of the detectors (~30%) does not permit complete discrimination between single-photon emission at 826 eV and photons in the upper half of the two-photon emission spectrum.

Optical radiation to excite the $\Delta E - S$ transition nominally located at 2296 cm^{-1} (in the rest frame of the $^{19}F^{8+}$ atoms[1]) is provided by a pulsed HBr chemical laser.[9] This laser, operated with flowing argon, hydrogen, and bromine at a total pressure of 55 Torr and at 1.5 pulses/sec, simultaneously produces numerous frequencies in the range of 2296 cm^{-1}. The nonmonochromatized laser output was directed into the interaction chamber through a KCl window by a rotatable-mirror system consisting of the mirrors M2 and M3 and the lens L (see Fig. 1). The lens was used to focus the laser beam (1.5-mm spot size) so as to intersect the particle beam on the axis of the rotating-mirror system and between the two proportional counters (PC1 and PC2). This optical system, together with the fixed mirror M1, allows angle changes without realignment of the various optical components, and is designed to assure negligible vertical motion (with respect to the particle beam) of the laser beam throughout the angles of interest. The entire system was aligned with slits and a helium-neon laser to a precision of $\pm 0.1°$.

For the present experiments, the HBr laser power was not monochromatized. The angle θ between the laser and the particle beams was adjusted to use one of the strongest HBr laser lines —the $P_{2-1}(5)$—for the $\Delta E - S$ excitation of F^{8+}. This transition occurs as a doublet arising from the $H^{79}Br$ and the $H^{81}Br$ molecules in the chemical laser. These two lines are of almost equal

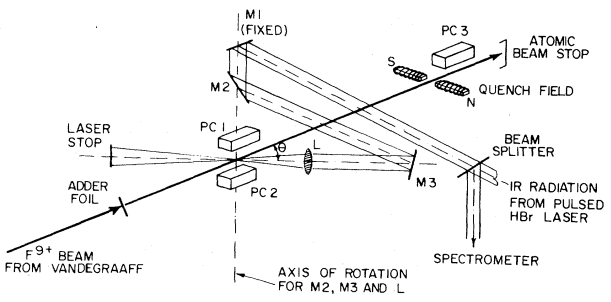

FIG. 1. Schematic drawing of particle-beam–laser-beam interaction. The rotating-mirror system used for Doppler-shift tuning is also indicated.

intensity[9] and are located at 2382.35 and 2383.68 cm^{-1}. The peak power output on these two transitions was ~10 kW in a 500-nsec-duration pulse. It is important to note that the frequency spacings between the various HBr laser lines[10] are such that if all lines were of equal intensity the contributions from the nearest-neighbor laser lines at a given angle would effect the $\Delta E - S$ excitation rate at about half the peak rate. For the present selection of angle θ and the corresponding HBr laser line $[P_{2-1}(5)]$ used for the exploration of the $\Delta E - S$ resonance curve, the nearest-neighbor HBr laser line on one side, the $P_{2-1}(6)$ at 2364.36 cm^{-1}, is missing because of atmospheric CO_2 absorption,[9] while the nearest neighbor on the other side, the $P_{2-1}(4)$ at 2400.47 cm^{-1}, is about $\frac{1}{5}$ the intensity of the $P_{2-1}(5)$ line. Thus the resonance curve should have a sizable asymmetric contribution to its line shape. The known and reproducible relative intensities[10] of all the HBr laser lines which are simultaneously interacting with the F^{8+} atomic beam in the interaction chamber were included in a consistent manner in the data analysis.

The frequency ν_F observed by the fluorine beam moving at a velocity v with respect to the laboratory (refer to Fig. 1) is

$$\nu_F = \frac{\nu_L(1 - v\cos\theta/c)}{(1 - v^2/c^2)^{1/2}}, \quad (1)$$

where ν_L is the rest-frame frequency of the laser. Data were obtained, as a function of angle θ, by observing the time coincidence between laser pulses and detected x rays with a coincidence resolving time of 1 μsec. In this way the signal-to-background ratio at the peak of a resonance curve was better than 3 to 1. Figure 2 is a time-to-amplitude-converter spectrum obtained at one angle.

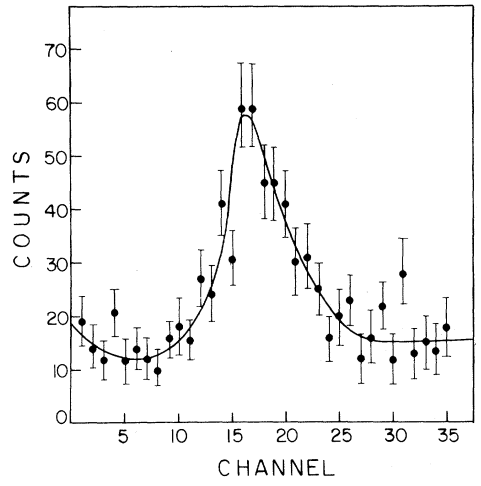

FIG. 2. Typical time-to-amplitude-converter spectrum. Start pulses are generated by the Ge(Au) infrared detector; stop pulses, by a proportional-counter signal. The width of the peak (full width at half-maximum) is about 1 μsec.

For each laser-beam–particle-beam angle chosen, coincidence data were accumulated for about 2 h. Each datum point of Fig. 3 represents total coincidence counts corrected for random events and normalized to integrated particle current and integrated laser power. A third proportional counter in an electromagnet downstream from the apparatus detected Stark-effect-induced Lyman-α radiation as a monitor of the F^{8+} metastable fraction in the particle beam. For the present experimental conditions the calculated resonant quench rate was about 1 per pulse. The actual detection rate was reduced by a factor of 4 to 8, probably because of uncertainties in proportional-counter efficiency and the actual metastable fraction.

The solid curve in Fig. 3 is a nonlinear least-squares fit to the data points with the line center as a parameter. ^{19}F has a nuclear spin ($I = \frac{1}{2}$) so that the $S_{1/2}$ and $P_{3/2}$ levels are doublets consisting of four and eight degenerate substates, respectively. Selection rules allow three distinct but unresolved transitions between the various hyperfine levels. To lowest order, the hyperfine structure leads only to a broadening of the resonance line but not to a shift in its center of gravity. The signal $F(\theta)$, given below, is pro-

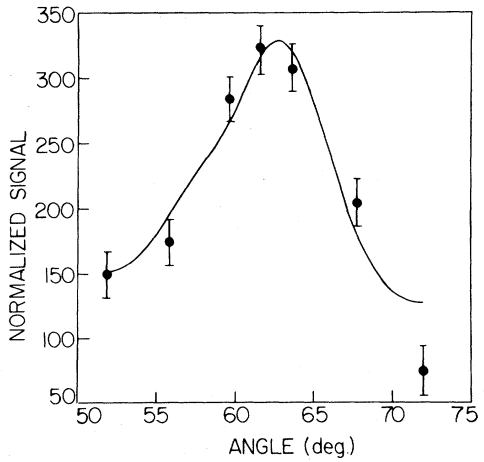

FIG. 3. Resonance curve used to obtain $\Delta E - S$ line center.

portional to the fraction of metastables driven to the $2P_{3/2}$ state with the transition probability μ given by the Wigner-Weiskopff formula[4] applied in the rest frame of the atoms.

$$F(\theta)=\sum_k\{1-\exp[-\textstyle\sum_{ij}\mu_{ij}{}^k(\theta)t(\theta)]\}\,, \qquad (2)$$

where the sum over k is over the four initial s-state components; over i, over final p states; and over j, over laser frequencies present. The θ dependence of μ involves the Doppler effect [Eq. (1)] and the Lorentz-transformed laser power. The parameter $t(\theta)$ is the laser-beam–particle-beam interaction time and is inversely proportional to $\sin\theta$.

The best-fit line center thus far obtained is 68 853.9 ± 35 GHz. As ΔE, the $P_{3/2}$-$P_{1/2}$ splitting, is quite insensitive to radiative corrections and is given to good accuracy by the Dirac theory with corrections[11] (ΔE = 72 192.96 Hz),[12] the Lamb shift can be extracted, yielding S = 3339 ± 35 GHz. For comparison the theoretical values are 3342 GHz using the series expansion[11] with the fourth-order term corrected,[13] 3349 GHz using Erickson's model,[12] and 3360 GHz using Mohr's calculation.[14] All of these calculations use 2.8 fm for the nuclear radius. Our anticipated final precision of 0.1%, based on improved statistics and signal-to-noise ratios, will allow discrimination between these various results.

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Saturation of Resonant Two-Photon Transitions in Thallium Vapor

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We present direct evidence on the saturation of resonant two-photon transitions in the optical region. Saturation manifests itself as a broadening of the transition linewidth and as a deviation from the low-level intensity dependence for both third-harmonic generation and two-photon–induced fluorescence. These effects may greatly limit the extent of attainable resonant enhancement in experiments which require high intensity for excitation.

Saturation of resonant two-photon transitions[1] in the optical region was first discussed before the advent of intense tunable dye lasers. In observing the fluorescence emission excited by a resonant two-photon transition in potassium vapor, Yatsiv *et al.*[1] noted on theoretical grounds that the transition should have been saturated; however, direct observation of saturation was not possible. In this Letter, we present the first direct evidence for the saturation of the resonant two-photon transition between the $6^2P_{1/2}$ and $7^2P_{1/2}$ levels of the thallium atom. Saturation manifests