sitions (Q branch), anisotropy splittings cannot occur. This has been verified by our recent highresolution measurements of the Q(1) line shape of hydrogen (4155.2 cm<sup>-1</sup>) in an applied field. The line was not split, but was shifted toward lower frequency in accordance with Eq. (5) of Ref. 4.

In summary, optical Stark splittings of rotational Raman transitions have been observed in highresolution inverse Raman spectra. The results are described well by calculations employing the optical polarizability anisotropy and time-averaged optical fields. Measurements show that the magnitudes of the splittings are significant even at moderate pulsed-laser powers. Thus, these laser-induced Stark effects are an important new consideration in high-resolution, nonlinear spectroscopy involving rotational transitions.

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## Measurement of the Lamb Shift in Hydrogenic Cl<sup>16+</sup>

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The Lamb-shift splitting in hydrogenic chlorine is measured to be 31.19(22) THz. The experiment was performed on a relativistic beam of chlorine ions using a CO<sub>2</sub> laser resonance method. The result agrees with one of the two existing calculations and is the most sensitive test to date of QED in high-*Z* systems.

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The  $2s_{1/2}-2p_{1/2}$  Lamb-shift splitting, \$, in the hydrogenic system  $Cl^{16^+}$  has been measured in a laser resonance experiment using a relativistic beam of chlorine ions. The result is  $31.19 \pm 0.22$ THz, in agreement with one of the two quantum electrodynamics (QED) calculations in the literature.<sup>1,2</sup> This result is one of the most sensitive tests to date of QED in bound systems because of the scaling of \$ with  $(\alpha Z)^n$ ,  $n \ge 4$ , and is important for interpreting the result of the recently reported Lamb-shift measurement in atomic hydrogen,<sup>3</sup> for assessing relativistic calculations of energy levels in heavy atoms, and for predicting QED effects in the limit of short distances and high field strengths.

The only previously reported high-Z Lambshift resonance measurement has been for the Z =9,  $F^{8^+}$  system.<sup>4</sup> However, nonresonance measurements of 8 have been reported for several systems<sup>5</sup> of which hydrogenic argon has the highest atomic number.<sup>6</sup> Other experiments on high-Z hydrogenic systems are in progress or are

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planned<sup>7</sup> and low-Z higher-precision measurements are also being continued.<sup>3</sup>

The n = 1 and 2 levels of hydrogenic chlorine are schematically indicated in Fig. 1. Our experimental technique is based on resonance quenching of the metastable  $2s_{1/2}$  level and is conceptually similar to the classic work of Lamb and Retherford.<sup>8</sup> The  $2s_{1/2}$ - $2p_{1/2}$  transition in a relativistic beam of chlorine ions is driven by radiation from a powerful carbon dioxide laser.<sup>9</sup> The resonance condition is monitored by detection of the subsequent Lyman- $\alpha$  x-ray emission at 2.96 keV, as a function of the CO<sub>2</sub> laser frequency. Because the expected Lamb-shift splitting overlaps the normal emission frequencies of the  $CO_2$  laser<sup>9,10</sup> (when Doppler shifted by about 3%) frequency tuning can be achieved by the Doppler effect and by discrete changes of the laser frequency. The tuning range of the laser is limited to about one half-width of the resonance at a fixed particle-beam-laserbeam intersection angle because of the short lifetime of the  $2p_{1/2}$  level which leads to a very broad resonance.

Figure 2 is a schematic diagram of the experimental setup at the Brookhaven National Laboratory tandem accelerator laboratory. A  $Cl^{14^+}$  beam at about 190 MeV is post-stripped to bare chlorine nuclei,  $Cl^{17^+}$ , by passage through ~  $60-\mu g/cm^2$ 



FIG. 1. Partial energy-level diagram of hydrogenic chlorine.

carbon foils. The beam (typically 2-3 particle nA) is then partially promoted into the metastable  $2s_{1/2}$  state of  $Cl^{16^+}$  by electron pickup in a thin movable (0.5 to 3  $\mu$ g/cm<sup>2</sup>) carbon foil. It is brought to a focus at the center of the interaction chamber, 5 to 10 cm beyond the foil, and then stopped in a Faraday cup for normalization purposes. Particle-beam divergence caused by multiple scattering in this foil was minimized with the use of adjustable irises prior to the interaction point.

The CO<sub>2</sub> laser beam enters the interaction chamber forming an angle  $\theta$  with respect to the particle beam, is focused to a waist coincident with the particle beam waist, and is reimaged onto the surface of a power meter. The laser system consists of a grating-tuned oscillator stage that produced 175-W pulses of 120- $\mu$ sec duration at 480 Hz and a 13-m-long amplifier stage that provided a saturated gain of 14. The amplified laser beam has good spatial coherence and was focused with f/3.8 KCl optics to a 150- $\mu$ m spot full width at half maximum (FWHM) to intersect with the  $Cl^{16^+}$  beam at angle  $\theta$ . A Ge:Au ir detector received a small portion of the laser oscillator signal and its output is used to trigger gating circuitry defining a "laser-on" state.

Two 300-mm<sup>2</sup> intrinsic Ge x-ray spectometers are mounted perpendicular to the intersection plane of the particle and laser beams. Thin Be windows allow detection of the 2.96-keV  $2p_{1/2}$ - $1s_{1/2}$  x rays while aluminum collimators minimized x-ray detection from other than the interaction volume. Typical spectra consisting of spontaneous two-photon and *M*1 radiation from the  $2s_{1/2}$  state and background x rays from Cl of lower ionization states were published in a prelimi-



FIG. 2. Schematic diagram of experimental apparatus (not to scale).

nary report<sup>11</sup> of this work and will be discussed in more detail in a subsequent publication.

A fast scaler bank was used to count timing pulses and the Lyman- $\alpha$ -like counts from the Ge x-ray spectrometers when both the laser and particle beams traversed the chamber. A second scaler bank stored counts at all times that the particle beam traversed the chamber. The presence of laser-induced counts was determined by the excess of counts when the laser was "on" over counts when the laser was "off" multiplied by the fraction of time the laser was on. The ungated counting rate was of order 70 kHz/detector while the laser-induced counting rate was of order 15 Hz/detector. In order to enhance the signal-to-background ratio, the CO<sub>2</sub> laser-system design was optimized so that a figure of merit defined by the average laser power (which yields signal) divided by the square root of the laser-on time (which is proportional to random background fluctuation) was a maximum.

Since the laser power is far too low to saturate the transition, the expression for laser-induced signal becomes to a good approximation a simple Lorentzian with the natural linewidth. In this limit, the  $2s_{1/2}-2p_{1/2}$  transition probability per unit time,  $\mu$ , is given by the Wigner-Weisskopff theory:

$$\mu = \frac{e^2 \Gamma S_m}{2\pi c \hbar^2} \frac{|r|^2}{(\nu_m - \mathbf{S})^2 + (\Gamma/4\pi)^2},$$
 (1)

where  $|r|^2$  is the square of the transition matrix element,  $S_m$  is the laser power density,  $\nu_m$  is the laser frequency in the ion rest frame,  $\Gamma(=1/\tau_{2p})$ is the natural linewidth, and  $\hat{s}$  is the Lamb shift. The quantity  $\mu$  is a function of intersection angle as both  $S_m$  and  $\nu_m$  must be transformed to the moving ionic reference frame:

$$\nu_{m} = \frac{\nu_{L} (1 - \beta \cos \theta)}{(1 - \beta^{2})^{1/2}}; \quad S_{m} = \frac{S_{L} (1 - \beta \cos \theta)^{2}}{1 - \beta^{2}}, \quad (2)$$

where  $\nu_L$  and  $S_L$  are the frequency and power of the laser in the laboratory frame and  $\beta$  is the ion velocity of light.

Figure 3 shows an example of our resonance data for particle beam energy of 190 MeV and  $\theta$ =130°. The data presented (as solid triangles) were the result of 8 h of acquisition time using eight different laser frequencies from within the 4-THz tunability range of the CO<sub>2</sub> laser.<sup>9,10</sup> The frequencies were changed every 3 min in a random order to average out any slow variations of the system. Also shown, in Fig. 3 (as the solid curve) is a best fit to the experimental data with



FIG. 3. Typical Lamb-shift resonance data, accumulated in 8 h, with one set of experimental conditions,  $E(C1^{16+}) = 190 \text{ MeV}, \ \theta = 130^{\circ}$ . The solid curve is the best fit. The agreement of points close in frequency are indicative of lack of systematic error. (See text.)

the Lamb shift and an amplitude factor as the only parameters. The final value for the Lamb shift was obtained from an overall fit to more than 50 h of data acquisition with one free frequency parameter \$ and free amplitude parameters for each individual resonance curve. This fit yielded the value \$ = 31.19(22) THz with  $\chi^2 = 29$  for 41 degrees of freedom. The error bound is the statistical one standard deviation. As a check of this procedure, a separate fit was employed for each data set, and averaging the results yielded a similar value for the Lamb shift, while estimating the error in the mean from the scatter of the various results yielded the same 0.7% relative error.

The two sources thought to have the greatest potential for producing systematic errors were possible laser-frequency-dependent counts induced in the counting system by nonresonance effects (e.g., electronic noise from the laser current pulsing) and variation in beam overlap with laser frequency changes. Electrical noise and/or rf pickup was minimized by careful shielding and grounding and by having the electrical signals for the laser system totally independent of the xray spectrometers and digital counting system. The resonance counting rate dropped to zero when there was no physical overlap between laser and particle beams, and close-lying wavelengths of differing power yielded equivalent counting rates, indicating that spurious laser frequency-dependent counts were not important.

The resonance signal is proportional to the product of laser power density, interaction time, and the overlap of the two beams. Overlap was acVolume 48, Number 6

complished to  $\pm 25 \ \mu$ m using a precision alignment block and an alignment telescope. The actual transverse intensity profile of the particle beam (Fig. 4) was determined by observing characteristic Mo x rays as a  $25-\mu m$  wire was moved through the beam. The laser intensity profile was determined by monitoring the power transmission of an attenuated beam past a stainless-steel knife edge. The overlap between the beams was directly determined by monitoring the induced counting rate as the laser beam was translated across the particle beam and is also shown in Fig. 4. As the focused laser beam was observed to be smaller than the focused particle beam for all frequencies, frequency-dependent variations in beam overlap were not considered to be important.

The hyperfine splitting for <sup>35</sup>Cl is calculated to be of order  $0.02\Gamma$  and need not be included in the fitting with the theoretical line shape because of its small size and the fact that it does not shift the center of the resonances. Possible errors due to uncertainty in ion beam velocity, intersection angle, and neglect of the  $2p_{3/2}$  level have been considered and are estimated to contribute an uncertainty less than  $\pm 0.03$  THz, far less than the final statistical uncertainty. Other possible systematic errors, for example, due to possible laser resonances with high-n state components in the beam were searched for in several ways. For example, changing the ion beam energy by 40 MeV (to 150 MeV) and (independently) varying the distance from exciter foil to interaction point by one-half decay length caused no change in the experimental result within one standard deviation. Reversing the direction of the laser beam through the interaction chamber to scan the resonance

curve at  $\theta = 50^{\circ}$  yielded a result within experimental uncertainty of the mean. Greatly reduced laser power because of a longer air path and many mirror surface losses, as well as possible systematic effects from an unstable beam waist position in this configuration, resulted in only a  $\pm 2\%$  statistical accuracy, however. All systematic test runs were included in the best value quoted above.

The experimental result for the Lamb shift in hydrogenic chlorine is 31.19(22) THz in agreement with the calculation of Mohr,<sup>1</sup> 31.35(2) THz, and with the series expansion in powers of  $\alpha Z$ given in the review article by Taylor *et al.*,<sup>12</sup> 31.27 THz. The result is three standard deviations below the calculation of Erickson,<sup>2</sup> 31.93(13)THz. Scaling the discrepancy recently reported for the Z =1 Lamb shift<sup>3</sup> with an order higher than Z<sup>5</sup> would yield a result which disagrees with the present result and suggests that the inconsistency reported there is due to low-order corrections other than QED terms. These corrections may include proton size or structure effects<sup>13</sup> or corrections due to relativistic recoil effects.

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FIG. 4. Horizontal cross sections, at intersection point, of particle beam intensity (dashed curve), laser power intensity (solid curve), and overlap measured by laser-induced counts (solid triangles).

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