Effects of external electric fields on high Rydberg states formed in foil and gas interactions of 85-MeV Ne^{6+} ions

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High Rydberg states in neon ions are formed in interactions of 85-MeV $^{20}Ne^{6+}$ ions with a thin carbon-foil target and with a helium-gas target. The effect of a weak, transverse, static electric field on the Rydberg states is investigated by measuring the electron yield due to field ionization in the strong electric field of an electron spectrometer. For both targets, marked oscillatory structure is observed in the dependence of the electron yield on the strength of the weak field. The Fourier analysis of the data reveals the dominance of a narrow band of fundamental frequencies in the oscillations and indicates the presence of second-harmonic contributions. The oscillations are interpreted in terms of quantum interferences among Stark states coherently developing in the weak electric field. The relevance of experiments of the present type to the understanding of the mechanisms of Rydberg state formation is discussed.

I. INTRODUCTION

In recent years, a large number of investigations have dealt with the production of high Rydberg states in fast ions interacting with solid or gaseous targets. The ultimate goal of the investigations has been to obtain some understanding of the specific mechanisms leading to the formation of these Rydberg states.

Most of the studies performed so far have been concerned with the production of Rydberg states in beam-foil interactions (for general aspects of beam-foil excitation, see Refs. 1–3). The results of measurements[4–8] of delayed x-ray emission following beam-foil excitation have been analyzed by means of cascade calculations.[4] Conflicting results regarding the initial population of the Rydberg states have emerged[5–8] from these analyses. Betz and co-workers[5,7] have concluded that foil excitation leads to a preferential population of states with high orbital angular momentum l (within the manifold of substates with given principal quantum number n). Such a behavior of the l distribution is at variance with the usual assumption that direct capture of target electrons ("last-layer capture") is the dominant production mechanism of high Rydberg states. The analysis of Pálinkás *et al.*[8] led the authors to conclude that the distribution of l substates within an n manifold is essentially uniform.

The study of foil-excited Rydberg states by means of *field-ionization* techniques was initiated by Vager *et al.*[9] who realized that Rydberg electrons field-ionized in the electric field of the electron spectrometer can contribute to the yield of "cusp" electrons emerging at an energy corresponding to the beam velocity. With the initial aim to separate the Rydberg contribution from the contribution of "convoy" electrons produced in the foil target, Vager *et al.*[10] and Kanter *et al.*[11,12] have performed systematic studies of the influence of different types of electric fields on foil-excited Rydberg states in fast ions. A remarkable result of these studies was the observation[10] of oscillatory structure, as a function of the strength of a weak longitudinal field, in the integral electron yield following field ionization of hydrogen Rydberg atoms in the electron spectrometer (see also Ref. 13). The oscillations were associated with quantum interferences ("Stark beats") among Stark states coherently developing in the weak field. The principal quantum numbers n of the Rydberg states giving rise to the oscillations were estimated to be about 50. Electric-field-induced structure had been observed earlier[14–16] in the delayed light or x-ray emission following foil excitation of various ions. The production mechanisms of foil-excited Rydberg states were discussed by Kanter *et al.*[11] on the basis of measurements using the field-ionization technique. Kanter *et al.* argue that their results are compatible with the assumption that last-layer capture is the dominant production mechanism, a conclusion which Betz and co-workers[5,7,17] claim to have disproved on the basis of their analyses of delayed x-ray emission.

Not much information has been obtained so far on the production of high Rydberg states in interactions of fast ion beams with gaseous targets. Betz *et al.*[4] found the delayed x-ray emission following gas-target excitation to be not significantly different from that observed in foil excitation. Cusp electron spectra have been studied (see, e.g., Refs. 18 and 19) also for the case of beam-gas interactions, but no separation of the Rydberg contribution to the electron yield has been attempted.

In the present work, we employ the field-ionization technique in a study of neon Rydberg states formed in interactions of 85-MeV Ne^{6+} ions with a carbon-foil target and with a helium-gas target. For both cases, the influence of a weak, transverse "deflector" field on the integral field-ionization yield in the electron spectrometer is examined. Regular oscillations in the yield as function of the strength of the weak field are observed for both foil and gas excitation. The observation of field-induced oscilla-

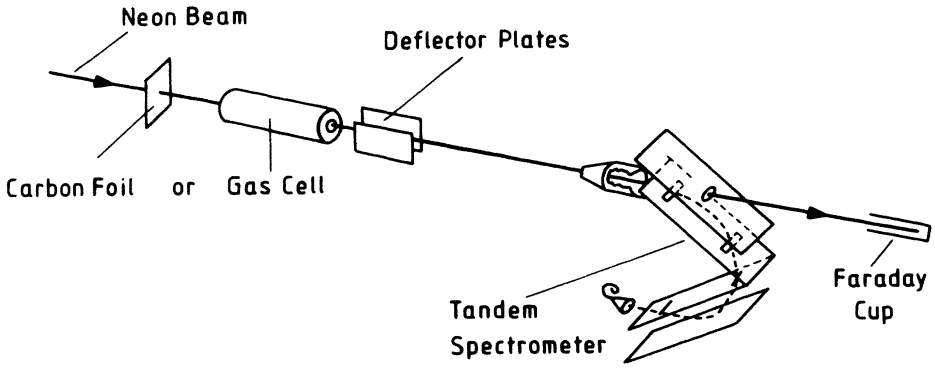

FIG. 1. Schematic diagram of the experimental setup.

tions in the Rydberg-electron yield following *gas* excitation represents what we believe to be the first observation of this kind.

In Sec. II we describe the setup used in the present experiment. Our experimental results as well as the results of a Fourier analysis of the data are presented in Sec. III. In Sec. IV the oscillatory structure observed in the field-ionization yield is interpreted in terms of quantum interferences among Stark states coherently developing in the deflector field. We also discuss the possibility of obtaining information on the production mechanisms of Rydberg states from experiments which employ the field-ionization technique. A summary of the paper is given in Sec. V.

II. EXPERIMENTAL SETUP

The experimental setup, which is similar to the one used in Refs. 10 and 12, is depicted schematically in Fig. 1. A tightly collimated beam of 85-MeV $^{20}Ne^{6+}$ ions accelerated in the Berlin heavy-ion facility VICKSI traverses a carbon-foil target or a helium-gas target. The thickness of the carbon foil is 20 $\mu g/cm^2$. The gas cell is 4 cm long; the pressure in the cell is kept at 10^{-2} Torr to ensure single-collision conditions. Downstream from the target, the beam passes through a transverse deflector field, i.e., a static, homogeneous electric field oriented perpendicular to the beam axis. The deflector field is generated by two parallel electrodes which are 2.1 cm long and separated by a distance of 1.3 cm. The distance between target exit and field entrance is 70 and 0.3 cm for the foil target and the gas cell, respectively.

After having traveled a distance of 6 cm from the deflector field exit, the ion beam traverses the first stage of an electrostatic 45° parallel-plate tandem electron spectrometer.[20] For purposes of normalization, the beam current is recorded by stopping the beam in a Faraday cup. The electric field of the first stage of the spectrometer serves to field ionize neon Rydberg states and to separate the ionized electrons from the beam. The purpose of the second stage of the spectrometer is to reduce the background due to slit-scattered beam ions and to improve the energy resolution to 3.2% full width at half maximum (FWHM) by using narrow slits of size 1.5×10 mm^2 (slit size of the first stage: 4×10 mm^2). The electrons are finally recorded by means of an electron multiplier (Thorn EMI, Type 9642/3B).

Typical electron spectra are shown in Fig. 2 for the gas-target case. The spectrum for *zero* deflector field F_D is dominated by the cusp peak centered about an energy of 2314 eV, which corresponds to electrons traveling with the beam velocity. The cusp electrons are ascribed to excitation into projectile continuum states in the target. Narrow lines superimposed on the tails of the cusp can be assigned[21] mainly to the decay of autoionizing Ne^{6+} $1s^2 2pnl$ ($n \geq 7$) configurations excited and decaying in the gas target. Electrons resulting from field ionization of projectile Rydberg states in the first stage of the spectrometer are expected[12] to show up at an "apparent" energy somewhat larger than the cusp energy since field ionization will take place only after the Rydberg ions have traveled a short distance into the spectrometer. We associate the bump seen on the high-energy side of the cusp peak at an energy of about 2600 eV with field-ionized Rydberg electrons. In the spectrum for $F_D = 385$ V/cm, the cusp peak has almost disappeared because most of the projectile continuum electrons have been deflected out of

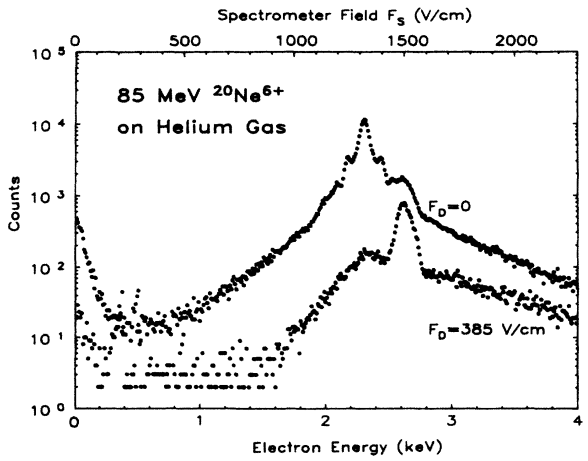

FIG. 2. Electron spectra following collisions of 85-MeV $^{20}Ne^{6+}$ ions with a helium-gas target, taken for zero deflector field and for a field strength $F_D = 385$ V/cm, respectively.

the beam. The Rydberg contribution at 2600-eV electron energy now shows up as a pronounced peak.

As compared with the cusp energy, the apparent energy of the Rydberg peak is shifted upwards by about 300 eV, independent of the strength of the deflector field. By calculating the electron trajectories in the first stage of the spectrometer, we have estimated that this shift roughly corresponds to an average distance of about 0.25 cm which the Rydberg ions penetrate into the spectrometer before field ionization takes place. In order to investigate the effect of the deflector field on the Rydberg states, the spectrometer field was kept fixed at $F_S = 1485$ V/cm, the field strength corresponding to an apparent energy of the Rydberg peak of 2600 eV. The total electron yield as function of the deflector field was studied by scanning F_D in the range 0—300 V/cm.

III. EXPERIMENTAL RESULTS AND DATA ANALYSIS

Figure 3 shows the F_D dependence of the electron yield measured for 85-MeV Ne^{6+} ions interacting with a thin carbon-foil target (a) and a helium-gas target (b). In both cases, the yield exhibits an oscillatory structure superimposed on a continuous background. While the background for the foil target decreases only slowly with increasing F_D, the background for the gas target decreases fairly rapidly in the range of small F_D values. The different behavior is related to the difference in the distance between target exit and deflector-field entrance in the two cases (cf. Fig. 1).

In order to Fourier analyze the oscillating structures seen in Fig. 3, we have subtracted from the total yield a smooth background contribution in such a way that the background-subtracted yield (Fig. 4) approaches zero for large F_D. If we assume the total yield $Y(F_D)$ to be the sum of a cusp contribution $Y_C(F_D)$ and an (oscillating) Rydberg contribution $Y_R(F_D)$, our procedure of background subtraction amounts to subtracting not only $Y_C(F_D)$, but also the average value $\overline{Y}_R(F_D)$ of $Y_R(F_D)$. Since $\overline{Y}_R(F_D)$ is expected to decrease, due to increased field ionization in the deflector field, slightly with increasing F_D, the Fourier spectrum of the background-subtracted yield may not fully reflect the Fourier spectrum of the Rydberg contribution to the yield.

The Fourier power spectra corresponding to the background-subtracted yield curves shown in Fig. 4 are displayed in Fig. 5. The spectrum resulting from the foil target exhibits two pronounced peaks at frequencies of 0.028 $(V/cm)^{-1}$ and 0.056 $(V/cm)^{-1}$ (field periods: 36 and 18 V/cm), respectively. In the spectrum resulting from the gas target, a pronounced peak appears at 0.033 $(V/cm)^{-1}$ (period: 30 V/cm), i.e., at a frequency slightly larger than the one at which the first peak in the foil-target spectrum shows up. A broad bump in the gas-

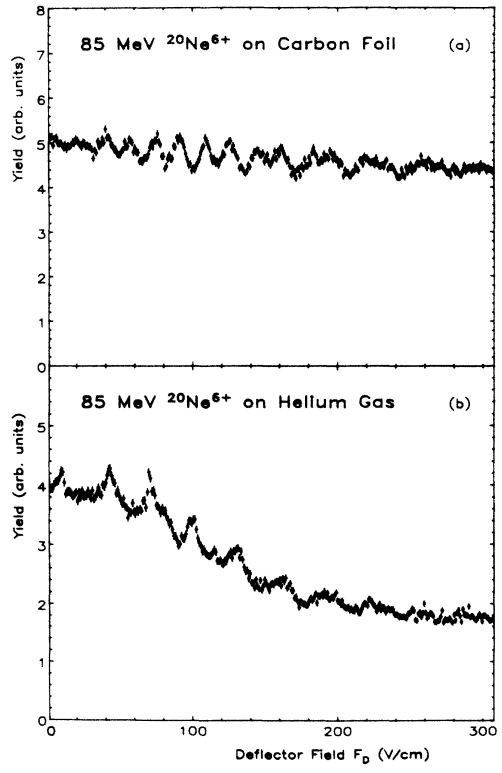

FIG. 3. Electron yield as function of the strength of the deflector field at fixed strength of the spectrometer field $F_S = 1485$ V/cm: (a) carbon-foil target; (b) helium-gas target.

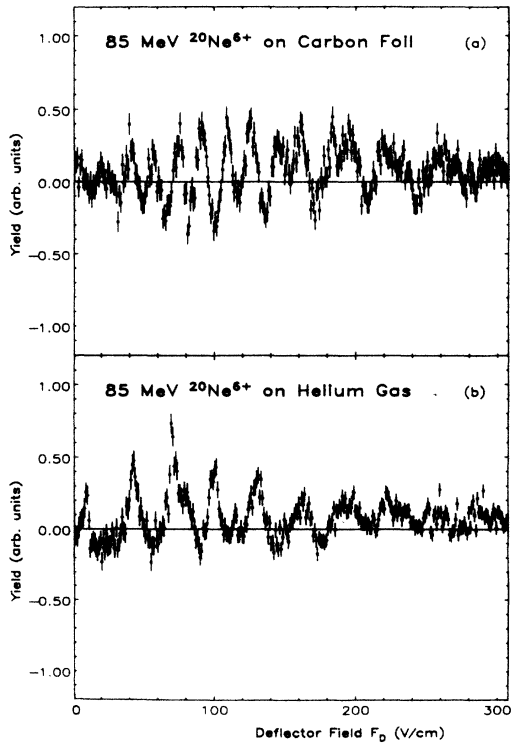

FIG. 4. Background-subtracted yield curves corresponding to the cases (a) and (b) of Fig. 3, normalized to zero for $F_D \to \infty$.

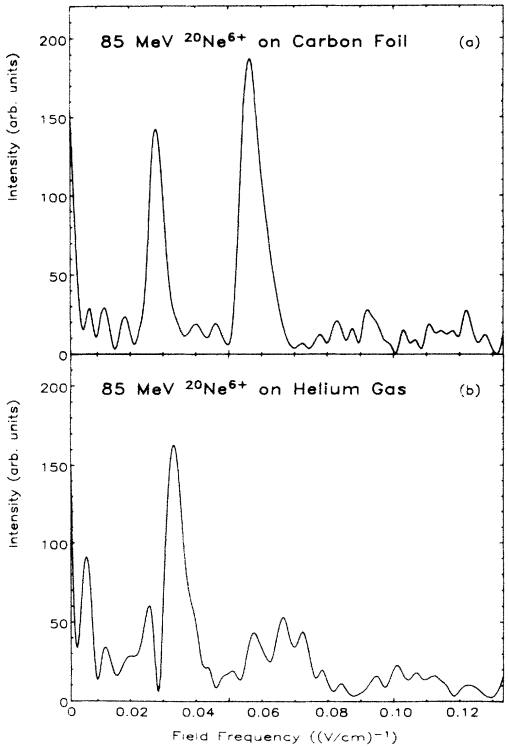

FIG. 5. Fourier-power spectra corresponding to the yield curves of Fig. 4.

target spectrum is centered about 0.065 $(V/cm)^{-1}$ (period: 15 V/cm), a frequency that is slightly larger than the frequency of the second peak in the foil-target spectrum. The fact that the ratio of centroid frequencies of the first and second peak is, in each of the two cases, very close to 1:2 suggests that the first peak represents a (narrow) band of fundamental frequencies and the second peak the corresponding second-harmonic contribution.

IV. INTERPRETATION OF THE RESULTS

We give a more or less qualitative interpretation of the oscillatory structure observed in the field-ionization yield by essentially following the ideas outlined by Vager *et al.*[10]

Since the energy spread associated with the time scale of the excitation processes is large in comparison with the energy splittings of the Rydberg states studied in the present case, the beam-target interaction will in general result in (fully or partially) *coherent* excitation of these states. Dealing with this general case requires the application of a density-matrix description.[2] However, for the arguments we wish to present here, it will be sufficient to separately consider individual hydrogenlike Rydberg states $\psi^{(Z)}_{nlm}(t)\equiv\phi^{(Z)}_{nlm}\exp(-iE^{(Z)}_n t)$ corresponding to an effective nuclear charge Z (unless stated otherwise, we use atomic units).

Under the conditions of the present experiment, the time evolution of the state $\psi^{(Z)}_{nlm}(t)$ in the deflector field may be calculated in first order in the field strength F_D. This follows from the fact that those Rydberg states which give the main contribution to the field-ionization yield in the strong spectrometer field are only weakly affected by the deflector field. Expanding the stationary state $\phi^{(Z)}_{nlm}$ in terms of Stark states $\phi^{(Z)}_{n_1n_2m}$ labeled[22] by parabolic quantum numbers n_1 and n_2 $(n=n_1+n_2+|m|+1)$, we can write the time-dependent state $\psi^{(Z)}(t)$ evolving from $\psi^{(Z)}_{nlm}(t)$ for times t in the interval $[0,t_d]$ corresponding to the passage of the Rydberg ion through the deflector field as

$$\psi^{(Z)}(t)=\sum_k \begin{bmatrix} (n-1)/2 & (n-1)/2 & l \\ (m+k)/2 & (m-k)/2 & m \end{bmatrix} \times\phi^{(Z)}_{nkm}\exp(-iE^{(Z)}_{nk}t)\ , \quad (1)$$

where $k\equiv n_1-n_2$. The expansion coefficients[23] denoted by $\begin{bmatrix} & & \\ & & \end{bmatrix}$ are the usual Clebsch-Gordan coefficients. The first-order energy of the Stark states is given by

$$E^{(Z)}_{nk}=E^{(Z)}_n+\frac{3}{2}\frac{F_D}{Z}nk\ .$$

The "dwell time" t_d of the ion in the deflector field may be expressed as $t_d=L/v$, where L is the axial length of the field and v is the beam velocity. In writing Eq. (1), we have assumed the quantization axis to lie parallel to the field direction.

When the ion leaves the deflector field, the field-dependent part of the phase factors in Eq. (1) becomes "frozen in" at its value for $t=t_d$, and the electronic wave function for $t>t_d$ reads

$$\psi^{(Z)}(t)=\left[\sum_k \begin{bmatrix} (n-1)/2 & (n-1)/2 & l \\ (m+k)/2 & (m-k)/2 & m \end{bmatrix}\phi^{(Z)}_{nkm} \times\exp\left[-i\frac{3}{2}\frac{F_D}{Z}nkt_d\right]\right]\exp(-iE^{(Z)}_n t)\ . \quad (2)$$

For given dwell time t_d, this function evidently is a periodic function of the field strength F_D with a fundamental period $4\pi Z/(3nk_0t_d)$, where $k_0=1$ or 2 depending on whether $n-|m|$ is even or odd.

While the effect of the (weak) deflector field on individual Rydberg states can be calculated in a simple fashion, the detailed theoretical evaluation of the field-ionization processes taking place in the electron spectrometer is a formidable task. However, for the purposes of a qualitative discussion of our data, it seems appropriate to elaborate only on the conditions which must be met in order that the periodicity properties of the wave function after the deflector field entail corresponding properties of the field-ionization yield.

A very simple picture would arise if the effect of the spectrometer would consist merely in inducing *sudden* transitions of the Rydberg electrons into the final continuum states sampled by the spectrometer, i.e., into eigenstates of the momentum operator. The amplitude $a_f^{(Z)}$ for field ionization of Rydberg electrons into the specific final state $|f\rangle$ is in this case given directly by the projection of

the wave function $\psi^{(Z)}(t)$ [Eq. (2)] onto $|f\rangle$, i.e., by an expression of the general form

$$a_f^{(Z)} = \sum_k C_k^{(Z)} \exp\left[-i\frac{3}{2}\frac{F_D}{Z}nkt_d\right], \tag{3}$$

where the coefficients $C_k^{(Z)}$ comprise both the Clebsch-Gordan coefficients of Eq. (2) and the overlaps of the Stark states with the final continuum states. This expression evidently preserves the periodicity properties of the wave function $\psi^{(Z)}(t)$ with respect to variations of the field strength F_D.

The assumption of a sudden ionization of the Rydberg electrons is not justified in our case, as can be inferred from the fairly large average distance the Rydberg electrons travel into the spectrometer before field ionization occurs (cf. Sec. II). However, the small width of the Rydberg peak (cf. Fig. 2) suggests that the region where field ionization takes place is sharply localized in space (and, accordingly, in time). Therefore, we may assume that the effect of the spectrometer can be separated into two successive steps.

In the first step, the Rydberg states undergo a *coherent* time evolution in the gradually increasing spectrometer field. In the second step, strong mixing of the Stark states and field ionization occur in a very short time interval in that spatial region where the field strength is just about to reach its full value. The time evolution in the first step results in a wave function $\psi^{(Z)}(t)$ modified by phase factors which depend on the integral of the field strength over the effective time that elapses until field ionization takes place. However, these phase factors do not disturb the periodicity of the wave function with respect to variations of the deflector field F_D. Then, if we assume the field-ionization process itself to be represented by a projection of $\psi^{(Z)}(t)$ onto momentum eigenstates, we arrive again at an expression of the form (3) for the ionization amplitude, albeit with coefficients $\tilde{C}_k^{(Z)}$ depending on the strength of the spectrometer field. The periodicity properties of $a_f^{(Z)}$ remain unaltered if the quantization axis does not coincide with the direction of the deflector field (in this case, the ionization amplitude includes rotation-matrix elements depending on the relative angle between quantization axis and field direction).

The field ionization yield $Y_R(F_D)$ can be schematically written as

$$Y_R(F_D) \propto |a_f^{(Z)}(F_D)|^2 = \sum_{\kappa=0,\pm1,\ldots}\left[\sum_K \tilde{C}_{K+\kappa}^{(Z)}\tilde{C}_{K-\kappa}^{(Z)*}\right]\exp\left[-2\pi i\frac{F_D}{\Delta F_D}\kappa\right]. \tag{4}$$

This expression consists of an F_D-independent part ($\kappa=0$), and a part periodic in F_D [$|\kappa| \geq 1$; note that the quantum number k in Eqs. (1)–(3) varies, for fixed n and $|m|$, in steps of *two* units] with the fundamental period ΔF_D given by

$$\Delta F_D = \frac{2\pi}{3}\frac{Z}{nt_d}. \tag{5}$$

The total field-ionization yield comprises contributions from a certain band of n values. Because of the n dependence of the fundamental period ΔF_D, the possibility to observe deflector-field-induced oscillations in this yield depends on the width of this band. Marked oscillations are expected to show up only if this width is sufficiently small. The n band is limited on the low-n side by the threshold for field ionization in the spectrometer field; its extension to high-n values depends on the mechanisms of Rydberg-state formation.

The threshold for field ionization in the spectrometer can be estimated from classical conditions.[22] The simplest (one-dimensional) classical estimate gives $n_S = (Z^3/16F_S)^{1/4}$ for the threshold n value n_S above which ionization in a field of strength F_S takes place. In the present case ($F_S = 1485$ V/cm), $n_S = 93$ for $Z=7$ and $n_S = 120$ for $Z=10$. A quantum-mechanical estimate which takes into account the finite dwell time of the Rydberg ions in the spectrometer, is obtained by using the "semiempirical" formula of Damburg and Kolosov[24] for the lifetimes of hydrogenlike atomic states in electric fields. From this formula, we get, e.g., for $Z=7$, n_S values ranging between 136 and 111 when the parabolic quantum number n_2 varies from its smallest to its largest value.

Identifying now the fundamental period given by Eq. (5) with the corresponding periods obtained from the Fourier analysis of the field-ionization yield, we arrive at the following interpretation of our data.

From the period (36 V/cm) associated with the first peak in the *foil-target* spectrum of Fig. 5, the n values contributing to the field-ionization yield are estimated to be centered about $n=72$ for $Z=7$ and about $n=103$ for $Z=10$. The latter value may be most appropriate for a discussion of foil-target data since one expects multiple collisions in the foil target to result in fully stripped cores of the Rydberg ions. From the first peak (period 30 V/cm) in the *gas-target* spectrum of Fig. 5 we obtain $n=87$ for $Z=7$ and $n=124$ for $Z=10$. For discussing gas-target data, the n value calculated for $Z=7$ should be appropriate.

The absolute n values derived with the help of Eq. (5) are somewhat smaller than the estimates for the threshold values n_S given above. The discrepancy can presumably be explained by uncertainties in the estimates for n_S. The ratio 103:87 of the n values determined from the foil-target data ($Z=10$) and the gas-target data ($Z=7$), respectively, is in fairly good agreement with the ratio 120:93 of the corresponding n_S values calculated from the classical estimate.

The small widths of the peaks in the spectra of Fig. 5 suggest that only a narrow band of n values in the vicinity of the threshold value for field ionization contributes significantly to the yield. A plausible explanation for this result can be provided by assuming that the probability for forming Rydberg states decreases rapidly with increasing principal quantum number n. This would be the case, for example, if electron capture from target atoms is the dominating mechanism of Rydberg-state formation. More detailed information on the initial n population can possibly be obtained from an analysis of the *shape* of the peaks in

Fig. 5, in particular of their asymmetry.

According to our interpretation, the first peak in each of the spectra shown in Fig. 5 corresponds to the term with $\kappa=1$ in Eq. (4). Second-harmonic contributions associated with $\kappa=2$ appear in both the foil-target and the gas-target spectrum. The observation of these contributions in the present experiments is the first observation of this kind; the analysis[10] of hydrogen Rydberg states formed in beam-foil interactions had not given any indication for the presence of higher-than-first harmonics. The significance of the second-harmonic contributions observed in our case (as well as of the absence of clear evidence for third and higher harmonics) is difficult to assess. The high complexity of the coefficients $\tilde{C}_k^{(Z)}$ which weigh the contributions to the yield from different κ values renders a quantitative analysis in terms of mechanisms of Rydberg-state formation prohibitively complicated. In particular, the extraction of detailed information on the population of angular-momentum substates in the excited Rydberg manifold seems impossible.

Certain qualitative trends in the dependence of the field-ionization yield on the initial population of the Rydberg states can be indicated by making extreme assumptions on this population. Assuming, for example, only orbital angular momenta l close to the maximum value $n-1$ to be excited with non-negligible probability, the magnitude of the relevant coefficients $\tilde{C}_k^{(Z)}$ can be estimated to decrease rapidly with increasing value of $|k|$ (the decrease in the magnitude of the Clebsch-Gordan coefficients contained in $\tilde{C}_k^{(Z)}$ overcompensates by far the increase in the field-ionization width of the Stark states for large n_2, as can be inferred by applying the semiempirical formula for this width given by Damburg and Kolosov[24]). In this case, only a few terms corresponding to the smallest κ values will give large contributions to the yield, i.e., only the lowest harmonics are expected to show up in the Fourier spectra. Unfortunately, one cannot turn round the argument and immediately conclude from the appearance of only second harmonics in our case that *high-l* values are preferentially populated [note that the index κ in Eq. (4) involves only *differences* of k quantum numbers]. In the case of low-l values, the Clebsch-Gordan coefficients depend only weakly on the quantum number k. Then, the increase of the field-ionization width of the Stark states with increasing n_2 may give rise to large contributions to the yield from large-κ values.

V. SUMMARY AND CONCLUSIONS

By employing the field-ionization technique, we have studied in the present paper the influence of a weak electric field on high Rydberg states formed in gas and foil interactions of fast neon ions. For both types of target, the measured ionization yield exhibits regular oscillations as function of the weak field. The Fourier analysis of the oscillating part of the yield reveals the presence of a narrow band of fundamental frequencies as well as of second-harmonic contributions. The theoretical interpretation of the data has provided fairly strong evidence that quantum interferences among Stark states coherently developing in the weak field ("Stark beats") are responsible for the oscillations in the yield.

Stark beats among Rydberg states with principal quantum numbers larger than 100, as they have been observed in the present experiments, had not been observed previously. The identification of these beats clearly represents an interesting effect by itself. From the analysis of the present data, only information on the range of *principal* quantum numbers excited in the beam-target interaction has been obtained. We have shown that details on the initial population of angular-momentum substates are hidden in highly complicated coefficients which may depend, in particular, on specific properties of the electron spectrometer. In order to disclose these and other details, further systematic studies will be necessary, in which the type and the degree of ionization of the projectile as well as the angle between the directions of the spectrometer field and of the deflector field should be varied.

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