Photoabsorption of Ba and Na in an electric field: Manifestation of strong-field-mixing resonances

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We have observed large oscillations in the photoabsorption cross sections of Ba and Na in the presence of an electric field. The two-step laser excitation technique allowed the use of various polarization combinations for studying the oscillations in the vicinity of the zero-field ionization limits. Thus, we could for the first time determine the relative contributions from two entirely different theoretical models: The quasibound $m = 0$ states (strong-field-mixing resonances) on the one hand, and symmetry-induced oscillator-strength cancellations on the other. We found that the model of strong-field-mixing resonances alone is able to account for all our experimentally observed cross-section oscillations.

I. INTRODUCTION

The photoabsorption of atoms in an electric field, particularly at final-state energies near the zero-field ionization limit, has recently begun to be investigated both experimentally[1,2] and theoretically.[2-4] Perhaps the most striking experimental observations were resonancelike oscillations in the absorption cross section, which extended even above the zero-field limit. First attempts to explain these resonances have been based on the fact that the final state of this process lies in the "strong-field-mixing" regime, where the Coulomb field of the ionic core, and the applied external electric field, have roughly the same magnitude and neither of them can be considered as a perturbation. Recently, Rau[3] has shown very generally that an electron's motion in comparable Coulomb and external fields exhibits characteristic resonances whose spacing follows well-defined laws, depending on the nature of the external field. This picture has been used to explain the observed absorption resonances of atoms in magnetic and electric fields; furthermore, it predicts that such a pattern of strong-field-mixing resonances is a quite general phenomenon found in various branches of physics.

In contrast, Luc-Koenig and Bachelier[4] have pointed out that experimentally observed cross-section variations do not necessarily reflect equally strong variations in the density of final states (i.e., resonances). Instead, they calculated resonancelike modulations in the H-atom absorption cross section that arise largely from cancellations in the distribution of oscillator strengths, due to the relative symmetry of the initial states and the absorbed light with respect to the electric field. More precisely, the latter structures are generally predicted for π-polarized light absorptions (polarization parallel to the field) from a symmetric lower state, with zero-field quantum numbers $l+m$ even, and for σ-polarized light absorption (polarization perpendicular to the field) from antisymmetric states with $l+m$ odd.

Unfortunately, as we shall see below, the experimental results to date do not allow the separation of the two effects. Therefore, it has been the aim of the present investigation to confirm unambiguously the relative contribution from the proposed strong-field-mixing resonances and the oscillator-strength-induced cross-section modulations, respectively.

If we first summarize the previous investigations in more detail, we find that the only experiment so far was a one-photon absorption in Rb by Freeman *et al.*,[1] who first observed equally spaced cross-section oscillations of about 10–15% modulation. The oscillations started below and extended beyond the zero-field ionization limit $E = 0$. The fact that they could only be observed in absorption of π-polarized light led to the postulation of nearly stable $m = 0$ states, which was supported by classical trajectory calculations for the electron.[2] From this model, the spacing of the resonances has been calculated using a one-dimensional model potential. The result was in excellent agreement with the experiment and with the analytical expression derived by Rau for this case of strong field mixing,[3] which reads

$$\frac{dE}{dn} = (22.5\ \mathrm{cm}^{-1})\left(\frac{F}{4335}\frac{\mathrm{V}}{\mathrm{cm}}\right)^{3/4}. \tag{1}$$

While the spacing was also in agreement with the calculations of Luc-Koenig and Bachelier,[4] the depth of the observed resonances disagreed drastically from their quantum-mechanical treatment of the photoabsorption of hydrogen in an electric field. In their work, Luc-Koenig and Bachelier found that the density of upper $m = 0$ states exhibits resonancelike modulation of about 3%, which is much less than observed in Rb, and in addition, is of the same order of magnitude as the calculated 1% modulation in the density of $m \neq 0$ states. However, calculations of the actual

absorption oscillator strengths, or cross sections reproduced qualitatively the experimentally observed strong modulations. It could be shown that this phenomenon arises from periodically occurring partial cancellations in the density of oscillator strengths, df/dE, which lead to an additional 15% modulation in the cross sections for π-polarized light from the symmetric $^1S_0$ ground state. These cancellations, in turn, have been explained by symmetry properties of both the wave functions and the dipole operator in the excitation matrix element. In essence, certain states in a combined Coulomb and strong electric field are known to be symmetric with respect to the plane $z = 0$ (z being the electric field direction). In these states the electron's charge distribution is *not* distorted towards either the anode or the cathode side of the atom. These symmetric states occur periodically in energy, with the periodicity being approximately given by Eq. (1). Considering both the symmetry (depending on the polarization) of the dipole operator, and the $l_i + m_i$ symmetry of the initial state (n_i, l_i, m_i), the mentioned selection rules for the cancellation in the excitation matrix elements to these particular states have been established.

It is worth noting at this point that, so far, the obviously important symmetry properties of atomic states in an electric field have only been studied for a pure Coulomb potential of the atom. Moreover, the considerations have been confined to the region in the vicinity of the nucleus, where even the alkali Rydberg electrons are known to experience non-Coulombic forces, leading to nonzero quantum defects. In fact, Luc-Koenig and Bachelier point out that the modulation depth caused by cancellation effects may vary from atom to atom, depending on the particular core potential. Thus they explained the fact that the modulation observed in Rb was actually smaller than calculated for H. Nevertheless, since the observed modulation was still much greater than the calculated structure in the density of H states, the authors claimed that also in the photoabsorption in Rb the oscillator-strength structure rather than the strong-field-mixing resonances accounted for most of the observed modulations.

It follows immediately from the selection rules for these oscillations that, in any one-photon absorption from a symmetric atomic ground state, the oscillations can only occur in $m = 0$ final states. Hence, in the experiment of Freeman *et al.*,[1] they cannot be separated from the $m = 0$ strong-field-mixing resonances. In a two-step excitation of atoms in an electric field, however, the two effects can be observed separately by the appropriate choice of light polarizations in each of the two excitation steps. This is because the initial state of the crucial second excitation step (which goes to the strong-field-mixing region) is no longer confined to be the ground state of the atom; rather, it is some intermediate bound state, whose symmetry can be chosen by the polarization of the first laser. Consequently, in the present investigation we used resonant two-step excitation of atomic states in an electric field to study the effects of either of the proposed theoretical models separately.

We chose to study the cross-section modulations in both an alkali atom (Na) and an earth-alkali atom (Ba). The two competing theoretical models are both expressed in terms of orbital angular momentum quantum numbers and their projections, hence we had to consider possible complications due to spin-orbit splitting (in the case of Na), and configuration mixing (in the case of Ba), respectively. Since, however, all our experimental data were internally consistent and strongly supported one of the theories (strong-field-mixing resonances), we feel confident that the results are not largely affected by spin or many-particle effects. Consequently, our experiments may serve as an aid in interpreting results of related experiments, like the previous one-photon-absorption experiment in Rb.[1,2]

II. EXPERIMENTAL

The apparatus used for these investigations has been described before.[5] In principle, it it a crossed-beam apparatus, where the interaction region is defined by the crossing of an atomic beam (density about 10^8 atoms/cm^3) with the two collinear, pulsed laser beams. The interaction region is centered between two parallel electric field plates which provide either the static electric field or, alternatively, a field-ionizing pulse of about 5 kV/cm, occurring several hundred nanoseconds after the laser pulses and serving to ionize the long-living Rydberg states. The ions are pulsed through a grid in the upper plate and detected by a secondary-electron multiplier. The multiplier signal is fed into a boxcar averager and finally recorded as a function of the wavelength of the second laser. The scan rate of the second laser was measured simultaneously by monitoring and recording its transmission through an etalon, after passing the interaction region. The absolute wavelength calibration was made using the well-known spectroscopic data for zero-field excitation.[6-8] The polarization of the laser beams, being initially polarized at 45° with respect to the electric field direction, was defined by two calcite prism polarizers. In the case of Na, it was necessary to com-

pensate for laser-power fluctuations by monitoring the laser power separately and normalizing the ion signal with respect to it.

III. PHOTOABSORPTION IN BARIUM

A. General considerations

Starting from Ba in its $6s^2$ ground state, we excited in a first step the $6s6p(^1P_1^0)$ intermediate state with either π- or σ-polarized light of 5537 Å, leading to a $\Delta m = 0$ or a $\Delta m = \pm 1$ transition, respectively. Using a second, tunable pulsed dye laser with wavelengths around 4200 Å we could then excite the strong-field-mixing states near $E = 0$ in the presence of an external static electric field.

If we disregard for the moment any possible reduction of the symmetry due to configuration interaction and think of the intermediate state as of a $6p$ electron surrounding a spherically symmetric $6s(^2S)$ core, then we can easily verify that now the effect of the expected oscillator-strength minima is completely separated from $m = 0$ final-state resonances. Let us consider, for example, two consecutive π-polarized transitions (π-π transition), which lead to a pure $m = 0$ final state. Since the intermediate state is antisymmetric with $l + m$ odd, no additional structure due to the oscillator strength should be observed and all of the oscillations stem from particular $m = 0$ upper-state properties. A σ-π transition, as well as a π-σ transition, leads to a pure $|m| = 1$ final state, in which case practically all of the observed structure is expected to be due to the transition oscillator strength caused by the respective symmetry of the light and the electric field. In the present experiment, we have investigated the modulations in the experimental cross sections for all the possible combinations of linear light polarization, at two diffferent values of the electric field. For convenience, the expected structures for the various polarizations are summarized in Table I.

Before discussing the polarization dependence of our results, we may discuss the general features of the Ba excitation spectrum using Fig. 1(a) as an example. The spectrum was obtained at 4.8-kV/cm field strength with the first laser being polarized perpendicular to, the second parallel to, the field (σ-π transition). Starting from the high-energy side, the spectrum exhibits the following features: at 42 117.4 cm^{-1} a Beutler-Fano profile, which is caused by the autoionization of a low-lying member of a $J = 2$, even-parity series, converging to the $5d_{5/2}$ ionization limit.[9] Below the zero-field 6s ionization limit I_0 (at 42 035 cm^{-1}) the main structure is a broad absorption dip centered around 41 841 cm^{-1}. It arises from the interaction of a $5d\,7d(^1D_2)$ perturber with the continuum of the fast ionizing Stark manifold of a 6s core configuration. Towards lower energies, this continuum is increasingly structured due to the existence of long-living Stark components. Additionally, we could observe a perturbing $J = 1$, even-parity state at about 41 930 cm^{-1}, which also lies below the zero-field limit I_0. In contrast to the $5d\,7d(^1D_2)$ perturber, it appears as a peak rather than an absorption dip in the spectrum. An extensive and detailed experimental study of the interaction of these perturbers with the electric-field-induced continuum has been completed and will be published separately.[10]

TABLE I. Comparison of expected cross-section structures in Ba arising from the model of $m_l = 0$ final-state resonances (Refs. 2 and 3), or from oscillator-strength cancellations (Ref. 4). A pure $6s6p(^1P_1)$ intermediate state has been assumed when calculating the results given in this table.

Laser polarization combination	Final-$m_l = 0$-state resonances	Oscillator-strength cancellations
$(\sigma - \pi)$	no	yes
$(\pi - \sigma)$	no	yes
$(\pi - \pi)$	yes	no
$(\sigma - \sigma)$	yes	no

It is evident both from Fig. 1(a) and from detailed MQDT studies[7,11] that there is practically no perturbation extending to the vicinity of the zero-field ionization limit I_0 which could interfere with the study of the polarization-dependent strong-field-mixing resonances. For the following discussions of the experimental results, we will restrict ourselves to the energy region near I_0, between 41 950 and 42 100 cm^{-1}. The observed slight increase in the measured intensity towards lower energies is caused by a monotonic increase in the laser power due to the particular dye efficiency. The spectrum has not been corrected for laser-power variations; however, the laser power has been recorded simutaneously and showed short-time fluctuations of less than 3%.

B. Results and discussion

Figures 1(a)–(d) show the polarization dependence of the strong-field-mixing oscillations in Ba at an electric field strength of 4.8 kV/cm, obtained with σ-π, π-σ, π-π, and σ-σ transitions, respectively. In the energy region of interest (between 41 950 and 42 100 cm^{-1}) we observe the following behavior.

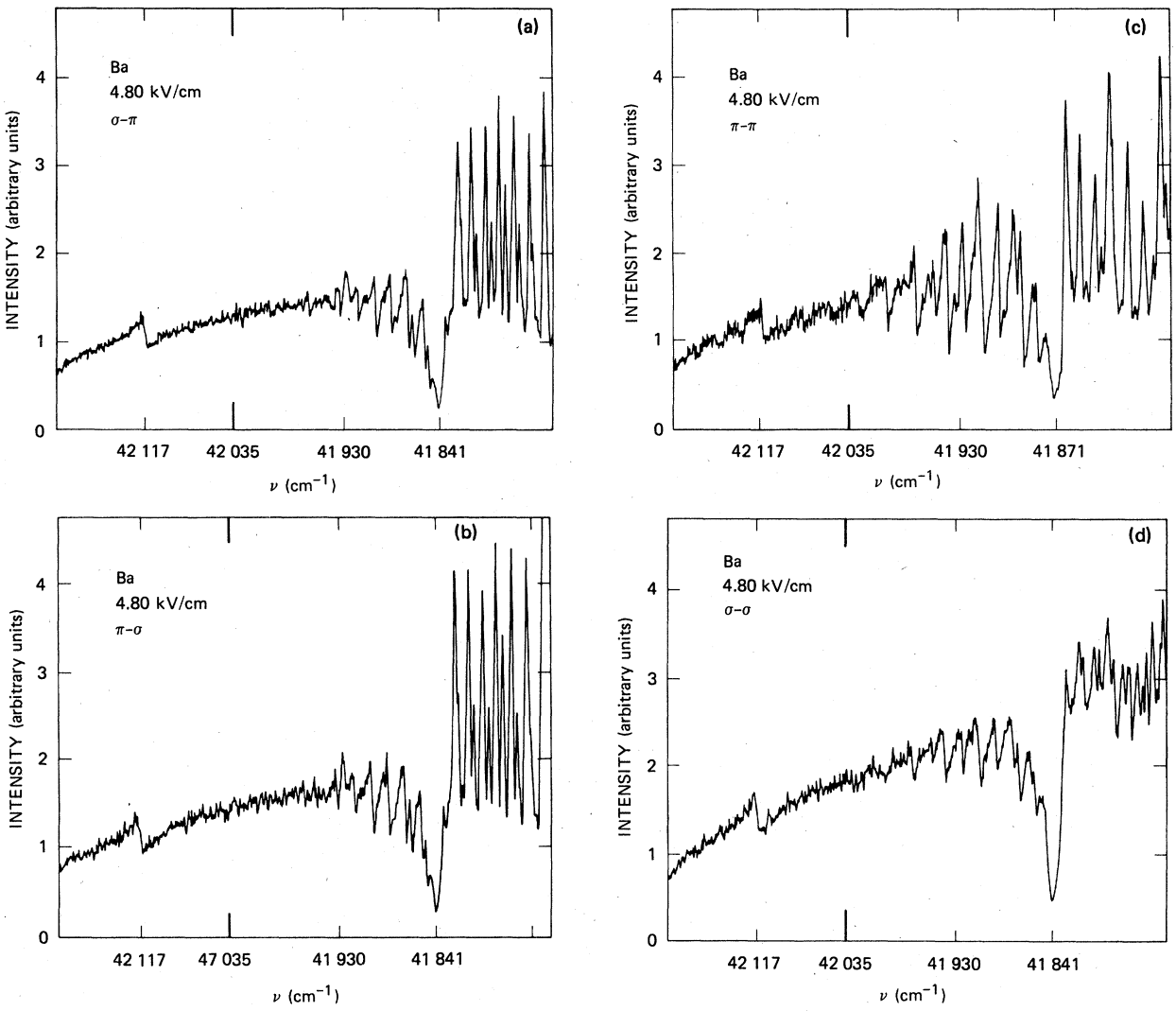

FIG. 1. (a)–(d) Experimental photoabsorption signal in Ba at an electric field strength of 4.80 kV/cm, obtained by resonant two-photon absorption with $\sigma-\pi$, $\pi-\sigma$, $\pi-\pi$, and $\sigma-\sigma$ polarization, respectively. The details of the spectra are explained in the text.

(1) In the case of a $\sigma-\pi$ [Fig. 1(a)] transition, there is practically no oscillation larger than the statistical fluctuation of the signal. A $\sigma-\pi$ transition leads to a pure $|m|=1$ state, whose density variation is expected to account for oscillations on the order of 1%.[4] According to the oscillator-strength model, a considerable amount of additional structure could have been expected from the transition because the selection rule for the second step ($l+m$ even and π-polarized light) predict zeros in the density of oscillator strengths to the upper state. From the examples calculated in Ref. 4, we may estimate that in our case the oscillations would also have been on the order of ~10%, which, in principle, could easily be observed.

There are two possible reasons for this disagreement from theoretical predictions: First, as mentioned in Ref. 4, the *absolute magnitude* of the oscillations is dependent on the nature of the core potential, which already in the case of Rb led to considerable deviations from the hydrogenic model. This leaves the possibility that in the case of Ba the oscillations are less than our statistical fluctuations. Second, our particular intermediate state $6s6p(^1P_1^0)$ contains about 30% admixture of $5dnp(^1P_1^0)$ character due to the presence of configuration interaction.[12] The $5d$ core configuration is no longer spherically symmetric, which could, in principle, affect the

symmetry considerations leading to the postulated structures in the oscillator strengths.

We do not expect, however, any additional effect due to the presence of electron spin, which is also neglected in the hydrogen photoabsorption calculation: It is known from the zero-field spectra that by far the most probable transition is the one to the singlet states, hence the electron spin remains completely unaffected in the transitions in Ba.

Similar considerations, only with reversed symmetries of both the light and the intermediate state, apply to the case of the π-σ transition, which is shown in Fig. 1(b). They lead to exactly the same result, that there is no experimental evidence for any oscillator-strength-induced modulations larger than the experimental uncertainty.

(2) In the case of a π-π transition [Fig. 1(c)], which leads to a pure $m = 0$ upper state, we find strong oscillations around the zero-field limit, which can easily be traced to energies as high as about 42 170 cm^{-1}. The spacing of the oscillations around I_0 is about 24.5 ± 0.5 cm^{-1}, which is in excellent agreement with the value of 24.28 cm^{-1} calculated from Eq. (1). The shape of the observed resonance structure is clearly asymmetric, which is in good qualitative agreement with the calculated density of $m = 0$ states in the case of hydrogen excitation in an electric field.[4] The modulation depth, however, which is about 20% in our experiment, exceeds by far the calculated 3% structure depth of $m = 0$ states in hydrogen. We note, from Table I, that no additional structure is expected from the oscillator strengths, since the pure $6s\,6p(^1P_1^0)$ $m = 0$ state has odd symmetry, leading to no structure in a π transition. A configuration-mixed intermediate state is even more unlikely to have a well-defined even symmetry ($l + m$ even) with respect to the electric field, which would be necessary to introduce additional amplitude in the observed oscillations. Consequently, we conclude that the observed 20% modulation in the absorption cross section is caused by the density variations of the $m = 0$ upper state alone.

(3) The σ-σ transition shown in Fig. 1(d) in general leads to mixture of $m = 0$ and $|m| = 2$ states. In the case of a field-free atom, with well-defined l values for the upper states, the ratio of $m = 0$ to $|m| = 2$ population can be calculated to be 1 : 7.[13] Consequently, to a zeroth-order approximation, we might expect the modulation amplitude to be reduced by a factor of about 7, compared to the case of a π-π polarization. The actually observed modulation (5% to 10% around I_0) appears to be reduced by only a factor 2–4. This is, for our purpose, still a satisfactory qualitative agreement, since no additional modulation from the oscillator-strength cancellation is predicted by the respective selection rules.

(4) To confirm the experimental results we have taken another set of spectra at a different field strength, $F = 2.67$ kV/cm. The observations were qualitatively identical to those of 4.8 kV/cm. In addition, as may be seen from the π-π transition shown as an example in Fig. 2, the spacing of the oscillations was much narrower (16.1 ± 0.5 cm^{-1}), again in agreement with the calculated value of 15.6 cm^{-1}. Thus the $F^{3/4}$ dependence of the spacing, which is characteristic for the strong-field-mixing resonances, was confirmed.

IV. PHOTOABSORPTION IN SODIUM

A. Experimental results

Starting again from ground-state atoms, the first laser now drives the $3s_{1/2} \rightarrow 3p_j$ ($j = \frac{1}{2}, \frac{3}{2}$) transitions in Na around 5890 Å, and the second laser ionizes the atom. The experiment is carried out in the same way as the Ba experiment. However, for reasons given below, only one polarization configuration (π-π polarization) has been studied in detail with both the $^2P_{3/2}$ and the $^2P_{1/2}$ state as the intermediate state, at an electric field strength of 8.91 kV/cm. In addition, one spectrum at 4.46 kV/cm was taken with the $^2P_{3/2}$ intermediate state, using again the π-π polarization. Typical scans are shown in Figs. 3 and 4. The modulation spacing and depth results are tabulated in Table II. Again, the observed spacings are in good agreement with the theoretical values of 38.6 cm^{-1} at 8.91 kV/cm and 23.0 cm^{-1} at 4.46 kV/cm as given by Eq. (1).

In order to understand the modulation depths we now must take into account the effects of the electron spin. Since the $3p$ states have a large spin-orbit splitting (~17 cm^{-1}) compared to our laser linewidth (~1 cm^{-1}), we may never populate a pure m_l state with either polarization. This makes the interpretation substantially more complicated, since we can no longer argue qualitatively by merely looking at the presence or absence of modulations. Instead, we have to calculate, for any given polarization combination, the m_l substate population fraction, from which the expected modulation depth according to either theory may be evaluated and compared with the experiment.

B. Theoretical modulation and comparison with experiment

We first define a modulation amplitude μ in the observed photoabsorption cross section $\sigma(E)$ near the ionization limit by decomposing the cross

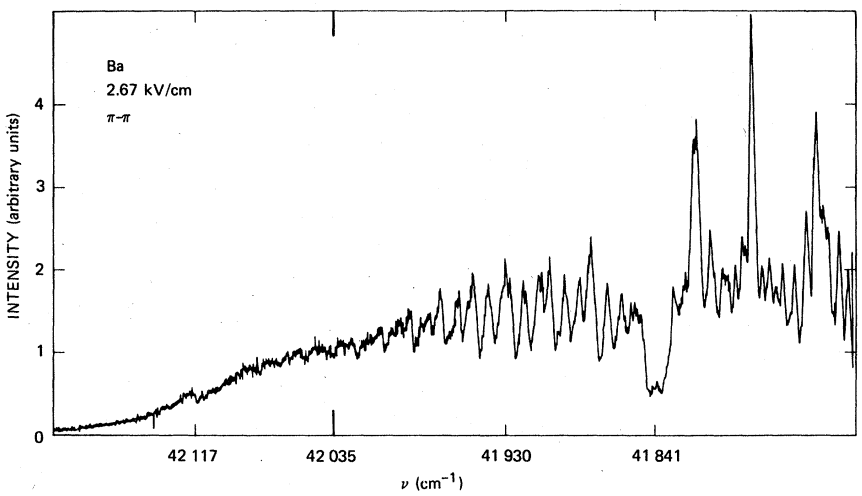

FIG. 2. Same as Fig. 1(c), except for a different value of the electric field strength (2.67 kV/cm).

section into a smooth and an oscillating component:

$$\sigma(E) = \bar{\sigma} + \mu g(E), \tag{2}$$

where $\bar{\sigma}$ denotes the average value of $\sigma(E)$, averaged over several oscillations. The function $g(E)$ describes the shape of the modulations; we only require $g(E)$ to be a periodic function of E [the periodicity is given by Eq. (1)], with unit amplitude and vanishing average over one period, i.e., $\overline{g(E)} = 0$. We will mainly be interested in the relative modulation amplitude M, which is defined by

$$M = \mu/\bar{\sigma}\ . \tag{3}$$

For the sake of simplicity we assume that the modulation amplitude is approximately constant within a sufficiently small energy region around the zero-field ionization limit. In order to account for this observed behavior theoretically, we now decompose the transition matrix element for the second excitation step in a similar way into an energy-independent part plus one or more oscillating components, $\alpha(E)$, which we will have to specify according to the theoretical model under consideration:

$$\langle\psi_f(E)|\hat{p}\cdot\vec{r}|\psi_i\rangle = \langle\psi_f|\hat{p}\cdot\vec{r}|\psi_i\rangle[1+\alpha(E)]. \tag{4}$$

Here again we require $\overline{\alpha(E)} = 0$ and define the first factor to be

$$\langle\psi_f|\hat{p}\cdot\vec{r}|\psi_i\rangle = \langle\psi_f(E)|\hat{p}\cdot\vec{r}|\psi_i\rangle_a \tag{5}$$

averaged over a suitable energy region around the zero-field limit. ψ_i and ψ_f denote the intermediate and the final state, respectively, and $\hat{p}$ is the polarization vector of the second laser.

If we now consider the model of the strong-field-mixing resonances, where the oscillations depend on the density of final m states $\psi_f(m_f)$, we identify

$$\alpha(E) = \alpha_{m_f}\,\delta_{m_f,0} \tag{6}$$

with the implicit understanding, that the (yet unknown) density modulation α_{m_f} has still the desired

TABLE II. Experimental results for the spacing and the relative modulation M of the cross-section oscillations in Na. The relative modulation M is defined as the average difference of two adjacent cross-section extrema, divided by half their sum, in an energy region of about 50 cm^{-1} below and 90 cm^{-1} above the zero-field ionization limit. The value of M given in this table is the weighted average of several (typically 3) recorded spectra.

Electric field strength F (kV/cm)	Laser polarizations	Intermediate state	Observed relative modulation M (%)	Observed spacing (cm^{-1})
8.91	π-π	$3\,^2P_{1/2}$	11 (2)	39.5 (45)
8.91	π-π	$3\,^2P_{3/2}$	21 (3)	35.9 (36)
4.46	π-π	$3\,^2P_{3/2}$	13 (2)	20.4 (21)

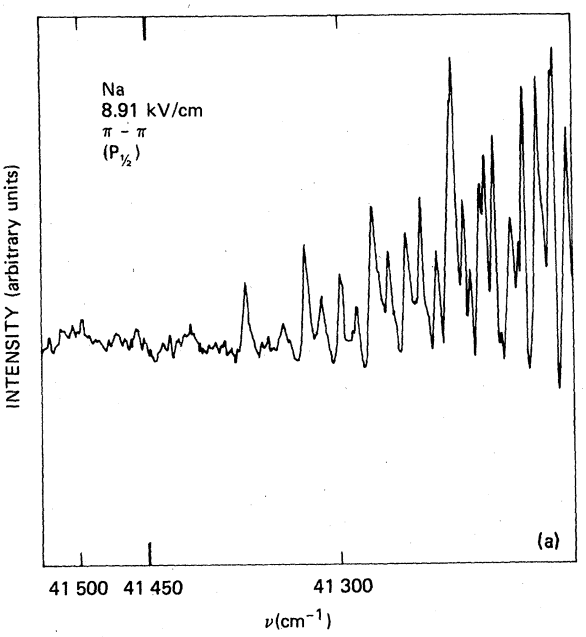

FIG. 3. (a), (b) Experimental photoabsorption signal in Na at an electric field strength of 8.91 kV/cm, obtained by resonant two-photon transition (π-π polarization) via the $3\,^2P_{1/2}$ intermediate state, and the $3\,^2P_{3/2}$ intermediate state, respectively. The energy of 41 450 cm^{-1} corresponds to the zero-field ionization limit.

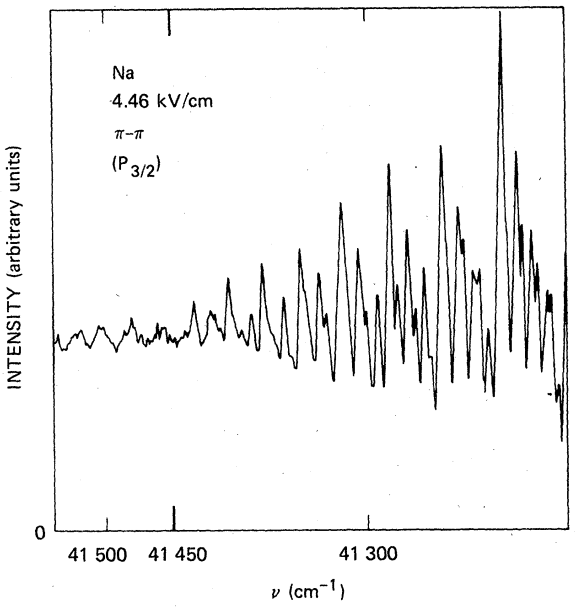

FIG. 4. Same as Fig. 3(b), except for a different value of the electric field strength (4.46 kV/cm).

oscillatory energy dependence. The Kronecker symbol accounts for the fact that only the $m_f = 0$ states are expected to exhibit a substantial density modulation and establishes a selection rule for this kind of modulation. The transition matrix element now reads

$$\langle\psi_f(E,m_f)|\hat{p}\cdot\vec{r}\,|\psi_i\rangle = \langle\psi_f(m_f)|\hat{p}\cdot\vec{r}\,|\psi_i\rangle(1+\alpha_{m_f}\delta_{m_f,0})\,. \tag{7}$$

Equation (7) also holds in the model of Luc-Koenig and Bachelier,[4] but eventually becomes multiplied by another factor which accounts for the modulation due to oscillator-strength cancellations. These, in turn, depend on the $(l_i + m_i)$ symmetry of the intermediate state ψ_i, and on the polarization of the light with respect to the electric field. To formulate this we may first expand the intermediate state $\psi_i(jm_j)$ into the (l,m,s,m_s) basis, since it is not substantially affected by the applied electric field. Then, after including another modulation factor β_{m_i} to account for possible oscillator-strength cancellations, we finally obtain the most general form for the matrix element:

$$\langle\psi_f(E,m_f)|\hat{p}\cdot\vec{r}\,|\psi_i(j,m_j)\rangle = \sum_{m_i,m_s}\langle l_i m_i \tfrac{1}{2} m_s | l_i \tfrac{1}{2} j m_j\rangle\langle\psi_f(m_f)|\hat{p}\cdot\vec{r}\,|\psi_i(l_i,m_i,m_s)\rangle(1+\alpha_{m_f}\delta_{m_f,0})$$
$$\times(1+\beta_{m_i}\delta_{\epsilon(\hat{p}),\epsilon(m_i)})\,. \tag{8}$$

We easily convince ourselves that the Kronecker symbol in the last factor yields the desired selection rules for the oscillator-strength cancellations,[4] if we define the symmetries $\epsilon(\hat{p})$ and $\epsilon(m_i)$ of the light and the intermediate state as follows:

$$\epsilon(\hat{p}) = \hat{p} \cdot \hat{F} \quad (= 0 \text{ or } 1, \text{ respectively}), \tag{9a}$$

$$\epsilon(m_i) = \begin{cases} 1 & \text{for } l_i + m_i \text{ even}, \\ 0 & \text{for } l_i + m_i \text{ odd}. \end{cases} \tag{9b}$$

Except for irrelevant numerical constants, the photoabsorption signal $\sigma(E)$ is then given by

$$\sigma(E) \propto \Sigma_{m_f} \Sigma_{m_j} C(m_j) \left| \langle \psi_f(E, m_f) | \hat{p} \cdot \vec{r} | \psi_i(j, m_j) \rangle \right|^2 . \tag{10}$$

The coefficients $C(m_j)$ describe the relative population of intermediate m_j states by the first excitation step. The $C(m_j)$ were readily determined since we saturated the first transition.

It is now straightforward, by inserting the matrix element (8) or (5) into Eq. (10) and observing the dipole selection rules, to obtain the theoretical expression for $\sigma(E)$ or $\overline{\sigma(E)}$, respectively. From this, the theoretical relative modulation $M_j^{p'p}$ can be derived, which is labeled by the light polarizations p' and p and by the angular momentum j of the intermediate state $3p_j$. The final result is (to first order in α, β)

$$M_{1/2}^{\pi\pi} = M_{1/2}^{\sigma\pi} = M_{3/2}^{\sigma\pi} = \frac{2\alpha_0 Z_{00}^2 + 4\beta_1 Z_{11}^2}{Z_{00}^2 + 2Z_{11}^2}, \tag{11a}$$

$$M_{3/2}^{\pi\pi} = \frac{4\alpha_0 Z_{00}^2 + 2\beta_1 Z_{11}^2}{2Z_{00}^2 + Z_{11}^2} . \tag{11b}$$

Thus, we are left with only two independent equations, which contain four unknowns: the two modulation amplitudes α_0 and β_1, which we finally want to determine, and two unknown transition matrix elements which we have abbreviated by the symbol $Z_{m_f m_i}$:

$$Z_{m_f m_i}^2 = \left| \langle \psi_f(m_f) | \hat{p} \cdot \vec{r} | \psi_i(m_i) \rangle \right|^2 . \tag{12}$$

We have already restricted ourselves to π polarization in the second step ($\Delta m = 0$), since σ polarization with $\Delta m = \pm 1$ would have introduced even more unknown matrix elements. We note that, due to the selection rules (6), (9a), and (9b), only quadratic expressions in α and β survived, while all terms containing products ($\alpha\beta$) vanished. This allows us to reduce the number of unknowns by considering only the *ratio of relative modulations* for two selected cases, which we also had studied experimentally:

$$\frac{M_{1/2}^{\pi\pi}}{M_{3/2}^{\pi\pi}} = \left(\frac{r + 2R}{2r + R} \right) \left(\frac{2r + 1}{r + 2} \right), \tag{13}$$

where we have used

$$R = \frac{\beta_1}{\alpha_0} \text{ and } r = \frac{Z_{00}^2}{Z_{11}^2} . \tag{14}$$

The expression enclosed in the second parentheses in Eq. (13) is equal to the ratio $\bar{\sigma}_{3/2}/\bar{\sigma}_{1/2}$ of the average absorption cross sections via the two intermediate states, $3p_{3/2}$ and $3p_{1/2}$, respectively. This ratio can be measured independently by simply sweeping the first laser over both resonance lines and keeping the second laser fixed. The oscillations in the cross sections can be averaged out by repeating the measurement at several nearby wavelengths of the second laser. We obtained

$$\left(\frac{\bar{\sigma}_{3/2}}{\bar{\sigma}_{1/2}} \right)_{\text{expt}} = 1.08 \pm 0.03$$

from which a value of $r_{\text{exp}} = 1.26 \pm 0.03$ can be computed. Finally, entering the experimental result for r, $M_{1/2}^{\pi\pi}$, and $M_{3/2}^{\pi\pi}$ into Eq. (13), we find the experimental value for the ratio R of modulations caused by oscillator-strength cancellations versus strong-field-mixing resonances to be

$$R_{\text{expt}} = -(0.02 \pm 0.17) .$$

Since, by definition (14), R is a positive number, we may conclude that R_{exp} is zero. In other words, all our observed modulations in sodium are caused by $m = 0$ final states (strong-field/mixing resonances) while any possible contribution from the oscillator-strength minima is less than the experimental uncertainty. This result is in agreement with the results in Ba.

IV. SUMMARY

In this paper we have reported the study of photoabsorption cross-section modulations in Ba and Na in the presence of a static electric field. We used a resonant two-step laser excitation technique, with independently selectable polarization of the two lasers. Thus we were able to prepare intermediate and final states in well-defined m_i quantum states in Ba, or, because of the large spin-orbit splitting, in a well-defined superposition of m_l substates in Na, respectively. This allowed, for the first time, the distinction between two theoretical models, which both claimed to account for previously observed cross-section modulations in Rb in the vicinity of the zero-field ionization limit: the model of quasistable $m_l = 0$ final states (strong-field-mixing resonances), or the model of cross-section modulations caused by symmetry-induced oscillator-strength cancellations. Both theories predicted

the same periodicity of the modulation, which scales with the $\frac{3}{4}$ power of the electric field strength. Our observed periodicity was in excellent agreement with the theoretical value in all cases where substantial cross-section modulations have been found.

By comparing the observed modulation amplitude with postulated m_l selection rules for either theory, we found that the model of quasistable $m_l = 0$ final states alone accounted for *all* our observed modulations, whereas any possible contribution from the oscillator-strength cancellations was less than our experimental uncertainty. The effects of the electron's spin or configuration mixing have been discussed, but are not believed to account for the failure of the latter theory. Rather, we may conclude that the symmetry-induced oscillator-strength cancellations are sensitive to deviations from the pure hydrogenic case, for which they have been calculated.

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