Interference Narrowing at Crossings of Sodium Stark Resonances

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We have observed a striking line narrowing in the photoionization spectrum of sodium atoms in an electric field. At crossings of two relatively broad resonances, one of them is narrowed by 2 to 3 orders of magnitude as a result of interference between the discrete and continuum coupling amplitudes that mediate their ionization. The observations agree quantitatively with WKB quantum-defect Stark theory and also with a simpler model involving two discrete states and two continua.

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In the course of investigating resonance widths in the sodium Stark photoionization spectrum, we have observed a striking line-narrowing effect. Near the crossings of otherwise relatively broad Rydberg-Stark levels having the same orbital magnetic quantum number (m_L), we have measured decreases of up to 2 orders of magnitude in the spectral width of one of the resonances over a few percent change in the electric field. This narrowing is a spectral manifestation of a phenomenon first observed temporally by Feneuille *et al.*[1] in Rb, where the ionization rates of certain levels in the Rb Stark spectrum were observed to decrease by factors up to 50 over a narrow range of fields. We have found excellent agreement of our observations and those in Ref. 1 with calculated spectra based on the recently developed WKB quantum-defect (WKB-QD) theory of the Stark effect in alkali metals.[2] These calculations confirm the interpretation of this narrowing (or stabilizing) effect as an interference between the ionization processes of nearby hydrogenic resonances mixed by the core interaction.

In the field-energy domain (FED) under consideration, the dominant contribution to the spectral width of observed Stark resonances with $m_L = 0$, 1, or 2 is from core-induced ionization via degenerate continua. The coupling to these continua arises from the nonhydrogenic part of the potential due to the core electrons which mixes the parabolic basis states of the hydrogen-Stark problem. In the vicinity of an avoided crossing of two such levels, the bound-continuum coupling amplitudes will interfere to some extent as a result of mixing of the anticrossing states. This produces a partial decoupling of one of these levels from the continua resulting in a decrease of its ionization rate and corresponding spectral width. This interference is analogous to that in dipole transition matrix elements which can sharply inhibit radiative transitions.[3] Related effects have been observed in the autoionization spectra of strontium[4] and barium[5] in an electric field, and in cesium[6] at low fields where a large number of closely spaced states approximate a continuum.

In our experiments, Na atoms in a thermal beam are stepwise excited by pulsed dye lasers via the $3^2P_{1/2}$ state to high-lying levels in an electric field.[7,8] The photoionization signal is measured as a function of the frequency of the linearly polarized second laser (~ 410 nm). We operate in a FED situated about 150 cm^{-1} below the zero-field ionization limit, E_0, with a field of from 2 to 4 kV/cm. This domain is appreciably above the classical saddle-point energy, E_{sp}, which defines the onset of rapid field ionization for the most anodic hydrogen levels (i.e., most downward going with field).[2] In this FED, many downward-going levels have broadened into continua because of their rapid ionization, with the result that sharp resonance peaks are fairly sparse. Furthermore, Na resonances are shifted by at most 0.5 cm^{-1} from their hydrogen energies, so that hydrogenic parabolic quantum numbers n, n_1, and m_L can be unambiguously assigned to them except near crossings of levels with the same m_L.

Figure 1(a) shows a succession of ionization spectra at several field values near the crossing of the $(n,n_1,m_L) = (20,19,0)$ and $(21,17,0)$ levels. (An $m_L = 1$ peak labeled X occurs in the data but does not couple with the $m_L = 0$ resonances and is not included in the calculations.) At fields below 3.6 kV/cm and above 4.1 kV/cm, the widths of these levels are seen to be on the order of 1 to 2 cm^{-1}, but interference is clearly noticeable as the levels approach the crossing field. At a field of 3.95 kV/cm, the width of one resonance exhibits the laser mode width of 0.7 GHz, indicating that the true atomic linewidth has been reduced by at least 2 orders of magnitude. Theoretical spectra at the same field values are shown in Fig. 1(b). These are calculated with the WKB-QD method[2] and use the experimental Na quantum defects but otherwise have no adjustable parameters. In light of this, the agreement between the calculated and observed spectra is excellent. The field and energy were calibrated to within 0.5% by comparing the observed spacing of $m_L = 1$ resonances to WKB-QD calculations for the $m_L = 1$ manifold.

Figure 2 shows the field dependence of the FWHM of the dominant spectral feature in the region of the

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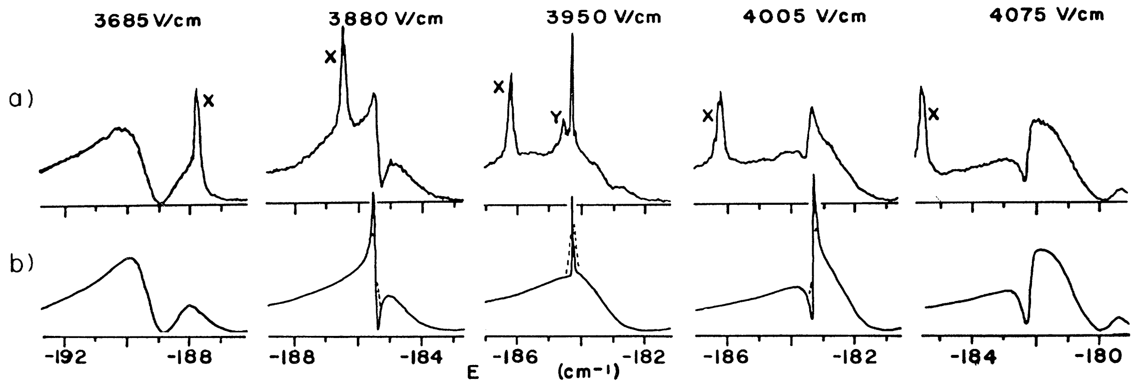

FIG. 1. (a) Experimental and (b) theoretical photoionization spectra of sodium Stark levels $(n, n_1, m_L) = (20, 19, 0)$ and (21,17,0) near their crossing. Excitation is via the $3^2P_{1/2}$ intermediate level. X is an $m_L = 1$ peak and is not included in the calculations. Y in the 3950-V/cm scan is an experimental artifact, probably due to some excitation from $3^2P_{3/2}$. The theoretical curves in the second row were computed with WKB-QD Stark theory (Ref. 2). Dashed lines show a 6-GHz instrumental width folded in.

(20,19,0)-(21,17,0) crossing for both the theoretical and the experimental spectra. There is good agreement at all widths larger than the laser resolution. Figure 3 shows the calculated field dependence of the width of the (20,19,0) level over a larger range of field values, together with the widths of other levels near their crossing with (20,19,0). For this plot, the width is taken to be the imaginary part of a complex eigenvalue calculated from a Hamiltonian matrix representation of the WKB-QD results. For isolated resonances and for the most narrow lines, widths computed this way agree well with fits of the calculated photoionization cross section versus energy to a Fano line shape. From 3 kV/cm, where the level begins to be

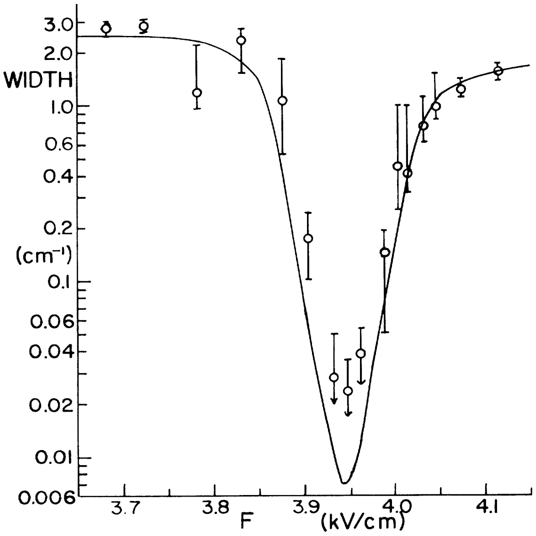

FIG. 2. Ionization width vs electric field for the (20,19,0) level near its crossing with (21,17,0) from experiment (data points) and from WKB-QD theory (solid line). Because the line shapes are quite asymmetric (except for very narrow lines), the width in this figure is taken to be the FWHM of the dominant feature corresponding to the (20,19,0) level in the photoionization cross section. For the narrowest lines, experimental widths are limited by the 0.7-GHz laser mode width. Error limits are asymmetric because of the peculiar line shapes and because of uncertainties due to the overlapping $m_L = 1$ resonance.

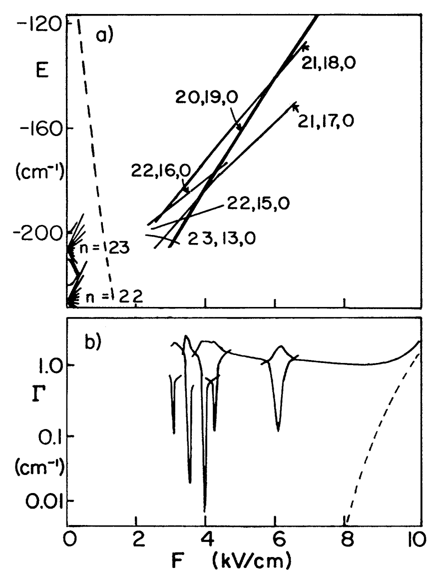

FIG. 3. Calculated energy and ionization width vs electric field for the (20,19,0) level from 3 to 10 kV/cm. Near each crossing region in (a), widths of the two intermixed components are shown in (b). The classical saddle-point energy is shown in (a) as a dashed line, and levels in the highly intermixed region near E_{sp} are omitted. In (b), the dashed line represents the hydrogenic width of the (20,19,0) level.

identifiable with the hydrogenic state, up to the point at which the hydrogenic width becomes appreciable, the width is relatively constant except near crossings, where one of the intermixed levels narrows drastically and the other broadens slightly.

To better illustrate the line-narrowing phenomenon, we consider a theoretical model of an isolated crossing of two discrete states coupled to one another and to two continua by a core interaction.[9] This is an extension of the work by Fano[10] and is equivalent to a model developed by Mies[11] and used recently by Saloman, Cooper, and Kelleher.[5] Since no assumptions are made concerning the character of the continua, they may be taken to be prediagonalized under the core interaction and form a basis for the continuum decay modes of the discrete states. This model can be generalized to treat the interaction of N discrete states which necessitates the introduction of N continua to describe their decay modes.

A complete solution of this model is necessary to obtain spectral shapes; however, the line-narrowing effect can be exhibited by a much simpler approach. Unperturbed discrete states $|\phi_i\rangle$ are coupled by the core interaction (V_c) to continua $|\psi_{jE}\rangle$, as indicated by matrix elements V_i^{jE}. The core-induced ionization width of an isolated state $|\phi_i\rangle$ is then

$$\Gamma_i = 2\pi \sum_j |\langle\phi_i|V_c|\psi_{jE}\rangle|^2 = 2\pi \sum_j |V_i^{jE}|^2. \qquad (1)$$

The states $|\phi_i\rangle$ are also mixed with each other by the core interaction, so that near a crossing of $|\phi_i\rangle$ and $|\phi_2\rangle$, the normalized eigenstates may be written

$$|\phi_+\rangle = \cos\theta\,|\phi_1\rangle + \sin\theta\,|\phi_2\rangle,$$
$$|\phi_-\rangle = -\sin\theta\,|\phi_1\rangle + \cos\theta\,|\phi_2\rangle. \qquad (2)$$

Here, θ depends on the energy separation of the unperturbed states and the matrix element $\langle\phi_1|V_c|\phi_2\rangle$. This angle varies from 0 to $\pi/2$ through the avoided crossing and is $\pi/4$ at the center. The ionization width of the mixed states is then

$$\Gamma_\pm = 2\pi \sum_j |\langle\phi_\pm|V_c|\psi_{jE}\rangle|^2 = \bar{\Gamma} \pm [\delta\cos 2\theta + (\Gamma_1\Gamma_2)^{1/2}\cos\gamma\sin 2\theta]. \qquad (3)$$

Here, $\bar{\Gamma} = (\Gamma_1+\Gamma_2)/2$, $\delta = (\Gamma_1-\Gamma_2)/2$, and $\cos\gamma$ is defined by

$$\cos\gamma = \left(\sum_j V_1^{jE} V_2^{jE}\right) / \{[\sum_j (V_1^{jE})^2]^{1/2} [\sum_j (V_2^{jE})^2]^{1/2}\}, \qquad (4)$$

and is a measure of the normalized overlap of the continuum states into which the unperturbed discrete states decay.

Equation (3) exhibits the decrease in the ionization width of one state near the unperturbed crossing field with a corresponding increase in the other width. The minimum width, attained at a field shifted from the unperturbed crossing field if $\Gamma_1 \neq \Gamma_2$, is

$$\Gamma_{\min} = \bar{\Gamma} - (\bar{\Gamma}^2 - \Gamma_1\Gamma_2\sin^2\gamma)^{1/2}. \qquad (5)$$

The dependence of $\Gamma_{\min}$ on the decay-channel overlap angle arises because the line narrowing is produced by an interference between the decay processes of the two bound states. Complete overlap of the decay channels, implying $\gamma = 0$ and equivalent to considering only one continuum, results in complete cancellation of the decay amplitudes for one state and therefore $\Gamma_{\min} = 0$. For $\gamma = \pi/2$, the decay channels are orthogonal and there is no interference. In this case, $\Gamma_{\min}$ will be the lesser of Γ_1 and Γ_2.[12]

A complete solution of the above model gives the spectral line shape as well as the linewidth functions. A connection with WKB-QD theory is established by separating off the parabolic channels that contain the hydrogenic resonances of interest[2,13] (effectively, the $|\phi_i\rangle$ above) and identifying the continuum channels that couple with them. Such a restatement of WKB-QD results in terms of a model with interacting discrete and continuum manifolds (to be discussed more fully in a future paper) provides resonance and resonance-interference parameters directly from the quantum-defect theory. For the (20,19,0)-(21,17,0) crossing, these calculations yield widths of 1.46 and 0.82 cm^{-1}, a 0.29-cm^{-1} coupling matrix element, and an overlap angle of 0.11 rad. The spectral shapes obtained from the two-continuum model above with these parameters (and the similarly calculated relative oscillator strengths) are essentially indistinguishable from those of Fig. 1(b) except for the extraneous $m_L = 0$ resonances in the wings of Fig. 1(b) which are not included in the model. The small overlap angle obtained for this crossing determines an overlap of almost 90% indicating considerable similarity of the two quasibound wave functions over the core region.

Our calculations, based on a WKB-QD approach extended to include spin-orbit effects, reproduce the Rb Stark spectra observations as reported by Feneuille *et al.*[1] These calculations indicate that, as in the Na narrowings reported here, core-mediated interactions between resonances within an m_L manifold and not spin-orbit coupling are responsible for the observed temporal stabilizations in Rb.

Littman, Zimmerman, and Kleppner[14] investigated the increase in the ionization rate of the (12,6,2) level in Na near its anticrossing with the relatively rapidly ionizing (14,0,2) level at 15.5 kV/cm. They found good agreement with a Bethe-Lamb treatment[15] which does not incorporate interference effects.[16] Interference effects are not significant at this crossing because the hydrogenic width of (14,0,2) is much greater than that of (12,6,2) and core broadening is very slight here.

Our observations are of interest as particularly transparent examples of a phenomenon that occurs whenever two autoionization peaks are close to each other. The possibility of interference narrowing in autoionization processes is implicit in equations developed by Fano[10] but has seldom been singled out for explicit discussion. However, it is dramatically evident when the resonance separation can be varied at will, as in Stark-Rydberg anticrossings.

There may also be practical applications of interference line narrowing for improved measurement precision. The occurrence of stabilized Stark levels over a small range in field strength may lead to a precise, reproducible electric field calibration procedure.

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[1]S. Feneuille, S. Liberman, E. Luc-Koenig, J. Pinard, and A. Taleb, J. Phys. B **15**, 1205 (1982).

[2]D. A. Harmin, Phys. Rev. A **24**, 2491 (1981), and **26**, 2656 (1982), and **30**, 2413 (1984).

[3]W. Davis, H. Metcalf, and W. Phillips, Phys. Rev. A **19**, 700 (1979); C. Kocher, Phys. Rev. A **6**, 35 (1972); J. Rubbmark, M. Kash, M. Littman, and D. Kleppner, Phys. Rev. A **23**, 3107 (1981).

[4]R. Freeman and G. C. Bjorklund, Phys. Rev. Lett. **40**, 118 (1978).

[5]E. B. Saloman, J. W. Cooper, and D. E. Kelleher, following Letter [Phys. Rev. Lett. **55**, 193 (1985)].

[6]C. Chardonnet, D. Delande, and J. C. Gay, Opt. Commun. **51**, 249 (1984).

[7]T. S. Luk, L. DiMauro, T. Bergeman, and H. Metcalf, Phys. Rev. Lett. **47**, 83 (1981).

[8]L. DiMauro, T. Bergeman, P. McNicholl, and H. Metcalf, J. Phys. (Paris), Colloq. **43**, C2–167 (1982).

[9]P. McNicholl and T. Bergeman, unpublished work.

[10]U. Fano, Phys. Rev. **124**, 1866 (1961).

[11]F. H. Mies, Phys. Rev. **175**, 164 (1968).

[12]If Γ_1, Γ_2 are not much smaller than $\langle \phi_1 | V_c | \phi_2 \rangle$, the widths $\Gamma_\pm$ given by Eq. (3) are not in complete agreement with the temporal decay rates obtained from a more rigorous analysis. {See W. E. Lamb, Phys. Rev. **85**, 259 (1952). Interference effects can be treated within the context of the Bethe-Lamb approach by adding an imaginary term, proportional to the decay-channel overlap, to the off-diagonal Hamiltonian matrix elements. [In M. G. Littman, M. L. Zimmerman, and D. Kleppner, Phys. Rev. Lett. **37**, 486 (1976), and Lamb, *loc. cit.*, only the diagonal elements are complex.]}

[13]W. van de Water, D. R. Mariani, and P. M. Koch, Phys. Rev. A **30**, 2399 (1984).

[14]Littman, Zimmerman, and Kleppner, Ref. 12.

[15]Lamb, Ref. 12.

[16]Interference effects can be treated within the context of the Bethe-Lamb approach by adding an imaginary term, proportional to the decay-channel overlap, to the off-diagonal Hamiltonian matrix elements. (In Refs. 14 and 15, only the diagonal elements are complex.)