Quantum-electrodynamic theory of overlapping spectral lines of multiply charged ions

V. G. Gorshkov, L. N. Labzovskiĭ, and A. A. Sultanaev

B. P. Konstantinov Leningrad Institute of Nuclear Physics, Academy of Sciences, USSR

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The dynamics of the decay of overlapping levels with identical quantum numbers and the formation of the spectral line contour are studied by the method of summation of diagrams for the S-matrix in the Furry picture. The result suggests that the shape of the contour differs significantly from the usual superposition of Lorentzian contours. The case of two adjacent levels, $2s^2$ and $2p^2$, with identical exact quantum numbers is considered in the spectrum of helium-like uranium under coherent excitation conditions of the initial state.

1. Multiply charged ions are objects in which many quantum-electrodynamic phenomena are more clearly manifest than for ordinary neutral atoms. As an example one may cite the observation of normally forbidden transitions in the spectra of multiply charged ions. These peculiarities are explained by the fact that the electrons participating in these transitions are highly relativistic. For example, in helium-like U^{90+} the velocity of the electron amounts to on the average to 0.67 of the velocity of light.[1]

In this paper we shall investigate one more effect which arises, apparently, only in the spectra of multiply charged ions, namely the overlap of the contours of spectral lines of two levels described by identical quantum numbers. We shall show that in this situation the total contour is substantially different from the usual superposition of the two Lorentzian contours corresponding to the individual lines. This situation is new to atomic spectroscopy (see Sec. 3 for more details).

For such relativistic systems as the multiply charged ions the construction of the contours of the spectral lines can be consistently carried out only on the basis of quantum electrodynamics. The quantum-electrodynamic theory of the line contour for one-electron atoms was fully developed first in the paper by Low.[2] The approach based on the summation of graphs for the S-matrix in the Furry picture is also convenient.[3] This approach permits readily the generalization of the results to the case of atoms (ions) with several electrons. First considered within this framework was the problem of overlapping levels for atoms.[4] In a recently published paper by Brown[5] a different quantum-electrodynamic method was proposed, based on the use of single-time Green functions; among other things the problem of overlapping levels was considered. One more method, employing the theory of the T-matrix, was applied to the problem of construction of the line contour in Ref. 6.

For the calculation of decay probabilities of states it is convenient to consider the evolution operator $\hat{S}(\infty,0)$, assuming that the system was prepared at the instant of time $t = 0$ and then decays. Here it is assumed that the decay does not depend on the method of preparation. Such a situation is typical of problems in spectroscopy. However in the case of two overlapping levels this statement of the problem needs refinement. It is asserted in Ref. 5 that the theory based on the evolution operator $\hat{S}(\infty,0)$ is nonrenormalizable. Nonetheless, in the resonance approximation the theory using $S(\infty,0)$ is renormalizable, as can be seen directly from the expression for probabilities containing only matrix elements of the electron self-energy operator in lowest order.[3] We emphasize that the entire theory of the line contour is formulated in the resonance approximation.

The characteristic process of excitation followed by radiation of a multiply charged ion is shown in Fig. 1. Here the thin solid line denotes the electron belonging to the ion, i, k, and f being the initial, intermediate, and final states of this electron. The double solid line depicts the incident particle—atom, ion, etc., which excites the ion under study. The wavy line denotes the emitted quantum (photon) with wave vector $\mathbf{k}_{\rm ph}$. Assuming that in the collision a certain level k is excited, and from it follows the transition to the final state f, then the probability of the emission process equals

$$dW_{kf} = |S_{kf}(\infty,0)|^2 \frac{d\mathbf{k}_{\rm ph}}{(2\pi)^3}, \tag{1}$$

where $S_{kf}(\infty,0)$ is the matrix element of the S-matrix.

In the general case, however, it is necessary to suppose that after the collision between the ion and the incident particle the state of the ion is described by the density matrix ρ_{kl}. Formula (1) should then be replaced by the formula

$$dW = \sum_{kl} \rho_{kl} S_{lf}^{*}(\infty,0) S_{kf}(\infty,0) \frac{d\mathbf{k}_{\rm ph}}{(2\pi)^3}. \tag{2}$$

Only the system ion + incident particle can be described by a definite wave function, the state of the subsystem (ion) is not pure.

The passage from (2) to (1) may be justified if the level is sufficiently well isolated in energy from other levels. But we are interested in precisely the opposite case: two adjacent levels 1 and 2 which are equally excited by the collision. Conditions for coherent excitation of the atom by light were

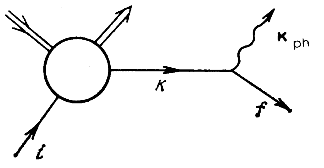

FIG. 1. Characteristic process of excitation and subsequent radiation by a multiply charged ion; i, k, f are the initial, intermediate, and final states of the electron.

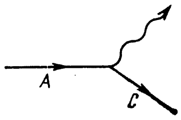

FIG. 2. Emission amplitude for the transition from an isolated level A to the ground state C to lowest order in the coupling constant.

discussed in Ref. 7. They consist in the overlap of the widths of the levels: in that situation levels 1 and 2 are excited by the same harmonics of the spectrum of the excitation force (the effective force acting on the atom upon irradiation by light, collision, etc.). Under such conditions the phases of the oscillations excited in the atom (transitions from the levels 1 and 2) are the same, and that means coherence. In the case of excitation by collision the argument is substantially the same. To coherent excitation corresponds the choice of the matrix ρ_{kl} in the form $\rho_{11}=\rho_{12}=\rho_{21}=\rho_{22}$. Then

$$dW_{1+2,f}=|S_{1f}(\infty,0)+S_{2f}(\infty,0)|^2\frac{d\mathbf{k}_{\rm ph}}{(2\pi)^3}. \tag{3}$$

Normalization of the matrix ρ_{kl} is specified by the condition

$$\int dW_{1+2,f}=1. \tag{4}$$

2. Before passing from overlapping levels to radiation we briefly consider the process of one-quantum transition from an isolated excited level A to the ground level C. To lowest order in the coupling constant this process is described by the diagram in Fig. 2. The transition amplitude, obtained by direct calculation of the corresponding matrix element of the S-matrix, equals

$$S_{AC}^{(1)}(\infty,0)=U_{CA}/(E_A-E_C-\omega_{\rm ph}), \tag{5}$$

$$U_{CA}=e(2\pi/\omega_{\rm ph})^{1/2}[(\boldsymbol{\alpha}\mathbf{e})^*\exp(-i\mathbf{k}_{\Phi}\mathbf{r}))]_{CA}, \tag{6}$$

where E_A, E_B are single-electron energies, $[\dots]_{CA}$ is the matrix element calculated using single-electron (Dirac) wave functions $\mathbf{k}_{\rm ph}$, $\omega_{\rm ph}$, $\mathbf{e}$ are the wave vector, frequency and polarization of the photon, $\boldsymbol{\alpha}$ are the Dirac matrices.

To obtain the usual Lorentzian line contour it is necessary that the self-energy insertions into the single-electron lines be taken into account. The summation of all such insertions into the initial state line in the resonance approximation[3] results in the replacement of E_A in Eq. (5) by $E_A+\Delta E_A{}^r$, where $\Delta E_A{}^r=(\Sigma^{(1)})_{AA}$ is the diagonal element of the self-energy operator in lowest order (radiative correction). Here

$$\Delta E_A=\delta E_A-{}^1/{}_2 i\Gamma_A, \tag{7}$$

where δE_A is the Lamb shift of the level and Γ_A is the radiative width of the level. Summation of all the insertions into the final state line results in the replacement of E_C by $E_C+\delta E_C$. In addition to the self-energy diagrams one must also sum the one-loop diagrams. In what follows the electron lines in the diagrams summed over the self-energy insertions in the resonance approximation will be represented by heavy lines.

After performing the indicated replacements in Eq. (5), substituting into Eq. (1), integrating over the directions of emission of the photon $\nu_{\rm ph}\equiv\mathbf{k}_{\rm ph}/\omega_{\rm ph}$ and summing over the polarizations e_α we obtain

$$dW_{AC}=\frac{1}{2\pi}\frac{\Gamma_{AC}\,d\omega_{\rm ph}}{(\omega_{AC}-\omega_{\rm ph})^2+{}^1/{}_4\Gamma_A{}^2}, \tag{8}$$

where

$$\Gamma_{AC}=2\pi\omega_{\rm ph}^2\sum_\alpha\int d\boldsymbol{\nu}_{\rm ph}|U_{CA}|^2 \tag{9}$$

is the partial width of the state A, due to the transition to the ground state C,

$$\omega_{AC}=E_A+\delta E_A-E_C-\delta E_C \tag{10}$$

is the transition frequency with the Lamb shift in the initial and final state taken into account.

3. Let us consider now the overlap of the contours of spectral lines of two adjacent levels A and B, from which transitions are possible to the ground state C. We suppose that the levels A and B are described by an identical set of quantum numbers. We consider first the amplitude S_{AC}. Now there occur, in addition to the usual resonance denominators due to keeping in sums over intermediate states just the state A, new resonance denominators corresponding to the state B. Diagrammatically the summation over such denominators is depicted in Fig. 3. Figure 3a describes the result of the summation of ordinary resonance denominators. In addition it is necessary, first, to sum over all insertions of the block X (see Fig. 3b) into the electron line of the initial state. The result of such a summation is

$$\frac{U_{CA}}{\tilde\omega_{CA}-\omega_{\rm ph}}\sum_{n=1}^{\infty}\left(\frac{X}{\tilde\omega_{AC}-\omega_{\rm ph}}\right)^n=\frac{U_{CA}}{\tilde\omega_{AC}-\omega_{\rm ph}-X}, \tag{11}$$

where

$$X=(\Delta E_{AB}{}^r)^2/(\tilde\omega_{BC}-\omega_{\rm ph}) \tag{12}$$

and we have introduced the notations:

$$\tilde\omega_{AC}=\omega_{AC}-{}^1/{}_2 i\Gamma_A, \tag{13}$$

$$\Delta E_{AB}{}^r=\Delta E_{BA}{}^r=(\hat\Sigma^{(1)})_{AB}\equiv\delta E_{AB}-{}^1/{}_2 i\Gamma_{AB}. \tag{14}$$

Second, one must add to expression (11) the result of the replacement of the vertex in the diagrams of Fig. 3b by the block Y (see Fig. 3c). This means that U_{CA} in Eq. (11) should be replaced by

$$Y=U_{BA}\Delta E_{AB}{}^r/(\tilde\omega_{AC}-\omega_{\rm ph}). \tag{15}$$

It is not hard to verify directly that upon such a summation the contribution of all resonant denominators is taken into account.

As a result of the indicated operations the following expression is obtained for the amplitude[7]:

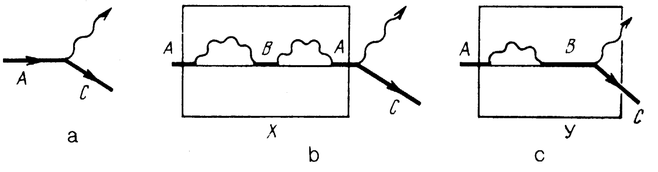

FIG. 3. Summation of resonance denominators: a—heavy line—the result of summing the resonance denominators corresponding to the state A, b—summation of insertions of block X, c—summation of insertions of block Y.

$$S_{AC} = [U_{CA}(\tilde{\omega}_{BC} - \omega_{ph}) + U_{CB}\Delta E_{AB}{}']/$$
$$[(\tilde{\omega}_{AC} - \omega_{ph})(\tilde{\omega}_{BC} - \omega_{ph}) - (\Delta E_{AB}{}')^2]. \qquad (16)$$

The denominator in formula (16) may be represented as follows

$$(\tilde{\omega}_{AC} - \omega_{ph})(\tilde{\omega}_{BC} - \omega_{ph}) - (\Delta E_{AB}{}')^2$$
$$= (\omega^{(1)} - \omega_{ph})(\omega^{(2)} - \omega_{ph}), \qquad (17)$$
$$\omega^{(1,2)} = {}^1/_2(\tilde{\omega}_{AC} + \tilde{\omega}_{BC}) \pm {}^1/_2[(\tilde{\omega}_{AC} - \tilde{\omega}_{BC})^2 + 4(\Delta E_{AB}{}^r)^2]^{1/2}, \qquad (18)$$

where values of $\omega^{(1,2)}$ indicate the location of the reconstructed levels. We may therefore write instead of Eq. (16)

$$S_{AC} = a_{AC}^{(1)}/(\omega^{(1)} - \omega_{ph}) + a_{AC}^{(2)}/(\omega^{(2)} - \omega_{ph}), \qquad (19)$$

$$a_{AC}^{(1,2)} = \pm[U_{CA}(\omega^{(1,2)} - \tilde{\omega}_{BC}) - U_{CB}\Delta E_{AB}{}^r]/(\omega^{(1)} - \omega^{(2)}). \qquad (20)$$

The amplitude S_{BC} is calculated analogously. Treating the excitation of the levels A and B as coherent, i.e., employing now formula (3), integrating over the directions, and summing over the polarizations we obtain the following expression for the combined line contour:

$$dW_{A+B,C} = \frac{\Gamma_{AC}|f_{AC}|^2 + \Gamma_{BC}|f_{BC}|^2 + 2\,\mathrm{Re}\,\Gamma_{AB,C} f_{AC}{}^* f_{BC}}{|(\omega^{(1)} - \omega_{ph})(\omega^{(2)} - \omega_{ph})|^2}\, d\omega_{ph}, \qquad (21)$$

where

$$f_{AC} = f_{AC}(\omega_{ph}) = \tilde{\omega}_{BC} + \Delta E_{AB}{}^r - \omega_{ph} \qquad f_{BC} = \tilde{\omega}_{AC} + \Delta E_{AB}{}^r - \omega_{ph}, \qquad (22)$$

and the mixed partial width $\Gamma_{AB,C}$ is determined by the formula

$$\Gamma_{AB,C} = 2\pi\omega_{ph}^2 \sum_{\alpha} \int d\mathbf{v}_{ph} U_{CA}{}^* U_{CB}. \qquad (23)$$

It follows from Eq. (21) that the line contour arising in the case of overlap differs from the superposition of two Lorentzian contours. The reason for this difference is twofold: first, the influence of the off-diagonal "shift" $\Delta E_{AB}{}^r$ and, second, the presence of interference terms. We note that the interference terms for the differential cross sections (probabilities) can also arise in the case of overlap of levels with different quantum numbers. Such a situation was discussed, for example, in Ref. 7. However in passing over to total probabilities the nonLorentzian form of the contour is obtained only for levels with identical quantum numbers.

Indeed, in the case of overlapping levels with different quantum numbers we have as a consequence of symmetry, first, $\Delta E_{AB}{}^r = 0$ and, second, $\Gamma_{AB,C} = 0$, so that both above reasons are inactive. For the differential cross sections, although $\Delta E_{AB}{}^r = 0$, the partial probability $\Gamma_{AB,C}$ does not vanish as there is no integration over angles and the interference terms are preserved regardless of the nature of the quantum numbers of levels A and B.

4. We shall investigate the overlap of identical levels in the spectra of multiply charged ions, using two-electron ions as an example. For the characteristics of the levels, with the electrons assumed to be relativistic, we shall make use of the *jj*-coupling scheme, i.e., we shall specify the set of quantum numbers $(nlj, n'l'j')$, where nlj is the set of single-electron quantum numbers (n is the principal quantum number, j is the total angular momentum, l is the orbital angular momentum), and J—the total angular momentum of the ion. The orbital angular momentum l is not conserved for a relativistic electron but indicates the parity of the state. The levels are degenerate in the quantum number l; this degeneracy is lifted upon taking into account the interaction between the electrons and the Lamb shift. The splitting of the energy levels according to the values of J also occurs when the electron–electron interaction is taken into account. For an ion the magnitude of this splitting is of the order of $\Delta E^{\text{int}} \approx E_0/Z$, where E_0 is the binding energy. We note that among levels belonging to the same *jj*-configuration $\Sigma\, ll'(nlj, n'l'j')$, there may be several levels with the same quantum numbers J and the same parity, determined by the sum $l + l'$. Further, for large values of the nuclear charge $Z(\alpha Z \sim 1$, where $\alpha \approx 1/137$ is the fine structure constant) the radiative width is $\Gamma \sim \alpha E_0$. Consequently, for appropriate numerical coefficients it could happen that $\Gamma \approx \Delta E^{\text{int}}$, which is the situation that interests us.

As a concrete example one may cite the doubly excited levels $(2s_{1/2}, 2s_{1/2})_0$ and $(2p_{1/2}, 2p_{1/2})_0$. For such levels we have, within the limits of *jj*-coupling and with the electron–electron interaction completely ignored, that formula (21) again reduces to the superposition of two Lorentzian contours. Indeed, $\hat{\Sigma}$ is a single-particle operator and its off-diagonal elements vanish in this case. Therefore a nontrivial result is obtained only when the electron–electron interaction is taken into account. The wave functions of the mixed states have the form

$$\psi^{(A,B)} = a_1^{(A,B)}\psi((2s_{1/2}, 2s_{1/2})_0) + a_2^{(A,B)}\psi((2p_{1/2}, 2p_{1/2})_0), \qquad (24)$$

where the mixing coefficients are determined by diagonalization of the electron–electron interaction V. It follows from the orthogonality condition of $\psi^{(A,B)}$ that:

$$a_1{}^{(A)} = -a_2{}^{(B)}, \quad a_2{}^{(A)} = a_1{}^{(B)}. \qquad (25)$$

For intermediate values of Z there is admixed to (24) one more level $(2p_{3/2}, 2p_{3/2})_0$ belonging to another *jj*-configuration. We shall, however, ignore this admixture.

All quantities entering formula (21) may be obtained from a knowledge of the coefficients $a_i{}^{(A,B)}$:

TABLE I.

Z	$Z^{-1}V(2s^2\,2s^2)$	$Z^{-1}V(2p^2\,2p^2)$	$-Z^{-1}V(2s^2\,2p^2)$	$Z^{-1}\Delta E_A^{\text{int}}$	$Z^{-1}\Delta E_B^{\text{int}}$	$a_1^{(A)}$	$a_2^{(B)}$
50	0.158	0.194	0.037	0.217	0.135	0.530	0.850
60	0.162	0.200	0.040	0.225	0.137	0.534	0.845
70	0.166	0.205	0.044	0.234	0.138	0.545	0.838
80	0.171	0.213	0.048	0.245	0.139	0.547	0.837
90	0.177	0.220	0.053	0.256	0.140	0.559	0.830
100	0.183	0.230	0.058	0.269	0.143	0.560	0.830

TABLE II.

Z	ΔE^{int}	δE_{2s}	δE^r_A	δE^r_B	Γ_A	Γ_B	δE^r_{AB}	Γ_{AB}
50	4.10	1.64	0.9	2.4	1.52	0.59	1.5	−0.3
60	5.28	3.30	1.9	4.8	2.91	1.16	3.0	−0.5
70	6.72	6.03	3.7	8.5	5.46	2.31	5.6	−0.9
80	8.48	10.42	6.3	14.7	8.93	3.83	9.6	−1.4
90	10.44	17.62	11.1	24.3	13.71	6.24	16.4	−2.1
100	12.60	29.25	19.0	40.0	20.39	9.28	27.2	−2.5

$$\Delta E_A{}^r=2(a_1{}^{(A)})^2\Delta E_{2s}{}^r+2(a_2{}^{(A)})^2\Delta E_{2p}{}^r, \tag{26}$$

$$\Delta E_B{}^r=2(a_1{}^{(B)})^2\Delta E_{2s}{}^r+2(a_2{}^{(B)})^2\Delta E_{2pr}, \tag{27}$$

$$\Delta E_{AB}{}^r=2a_1{}^{(A)}a_2{}^{(A)}(\Delta E_{2s}{}^r-\Delta E_{2p}{}^r). \tag{28}$$

Here we made use in Eq. (28) of the relation (25). Further, if one were to study, for example, the transition from the states A, B to the state[1)] $C=(1s, 2p)_1$, then

$$\tilde{\omega}_{AC}=E_2-E_1+\Delta E_A{}^r-\delta E_{1s}+\Delta E_A^{int}, \tag{29}$$

$$\tilde{\omega}_{BC}=E_2-E_1+\Delta E_B{}^r-\delta E_{1s}+\Delta E_B^{int}. \tag{30}$$

Here E_n ($n=1,2$ is the principal quantum number) is the energy of the electron in the Coulomb field and $\Delta E^{\text{int}}_{A,B}$ are the corrections, due to the interaction, obtained in diagonalizing the matrix V.

Matrix elements of the interaction V, borrowed from Ref. 8, and calculated on that basis values of $\Delta E^{\text{int}}_{A,B}$ and $a_i{}^{(A,B)}$, are given Table I. All quantities with the dimension of energy are given in atomic units. We note that the matrix V is represented in Ref. 8 in the form of just the first two terms of its expansion in αZ, and this could result in substantial errors for large Z.

Matrix elements of the interaction $\Delta E^{\text{int}}=\Delta E^{\text{int}}_A-\Delta E^{\text{int}}_B$, values of δE_{2s} borrowed from Ref. 9, and values of $\delta E_A{}^r$ and $\delta E_B{}^r$ calculated from formulas (26) and (27) are given in Table II. We do not list here values of δE_{2p} since for arbitrary Z one has the inequality $\delta E_{27}\gg\delta E_{2p}$.[9] The level widths Γ_A and Γ_B, taken from Ref. 10, are also given in Ref. 9. We determine these widths from the most intense transitions tabulated in Ref. 10. In addition the doubly excited levels have also a width due to autoionization, which, however, becomes smaller than the radiative width for large Z. Further we list in Table II the quantity $\delta E_{AB}{}^r$, calculated directly from formula (28). Lastly, the quantity Γ_{AB} may be expressed with the help of Eqs. (26) and (27) in terms of Γ_A and Γ_B:

$$\Gamma_{AB}=-\frac{a_1{}^{(A)}a_2{}^{(A)}(\Gamma_A-\Gamma_B)}{(a_2{}^{(A)})^2-(a_1{}^{(A)})^2}. \tag{31}$$

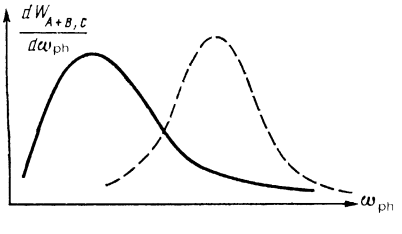

FIG. 4. Calculation of the line contour from formula (21) for transition from overlapping levels A and B to the final state $C(J=1)$.

It follows from the data in the Tables that, first, for $Z>50$ we have indeed $\Delta E^{\text{int}}\approx\Gamma_{A,B}$ and, second, that the off-diagonal element $\Delta E_{AB}{}^r\approx\Delta E^r{}_{AB}$ i.e., is not small. Consequently, in this region the difference between the total contour and the superposition of two Lorentzian contours should be substantial.

The final result is shown in Fig. 4 for $Z=90$. We have assumed here that $\Gamma_{A,C}=\Gamma_A$, $\Gamma_{B,C}=\Gamma_B$, $\Gamma_{AB,C}=(\Gamma_{A,C}\Gamma_{B,C})^{1/2}$ which should not cause too much of an error in the calculations. The solid line in Fig. 4 depicts the total contour obtained in the absence of interference terms and for $\Delta E^r_{AB}=0$, i.e., the superposition of two Lorentzian contours corresponding to the individual lines. The dashed line depicts the result of the calculation according to formula (21) in the case when $\Gamma_{AB,C}$ and $\Delta E_{AB}{}^r$ are different from zero.

The two expressions for $dW_{A+B,C}$ are normalized in the same fashion using formula (4). The result indicates that the form of the contour strongly differs from the usual superposition of Lorentzian contours. Let us note a certain analogy between the pair of levels here studied and the familiar pair of neutral kaons: the latter are also characterized by identical quantum numbers (with the weak interaction taken into account) and level overlap, although the interaction that determines the mixing of the initial states and decay lifetimes has a different physical origin.

The authors are grateful to V. M. Shabaev for discussion.

[1)] In the preceding discussion we have considered the final state C to be stable, which cannot be said about the state $(1s, 2p)_1$. It may be supposed, however, that allowance for the instability of the final state does not change the form of the contour, as happens in the case of an isolated level.

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