Influence of a strong external electromagnetic field on autoionizing states of atoms

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An analysis is made of the influence of a strong electromagnetic field on autoionization resonances in an atom with N closely spaced discrete levels below the first ionization threshold, which is in a multiphoton resonance with the ground state, and with M autoionizing levels above this threshold. The multilevel structure of a discrete state can be in the form of many closely spaced levels of highly excited states of an atom or it may form a multiplet. A study is made of the possibility of narrowing resonances as a function of the intensity and frequency of an external field, and generation of the third harmonic is considered.

INTRODUCTION

The influence of a strong electromagnetic field on autoionizing states of an atom is currently attracting much attention.[1-11] The nature and the origin of the autoionizing states are as follows: if two or more electrons are excited in an atom, there may be coupled states whose energies exceed the energy needed to detach one electron. Such a system is highly unstable and it is subject to autoionization (preionization), i.e., one electron may be transferred to the continuum. Autoionization occurs, for example, due to the interaction of electrons as a result of which one of the excited electrons is transferred to a bound state and the other becomes free. The probability of autoionization is many times greater than the probability of spontaneous emission of radiation by an excited atom. If the characteristic lifetime of an excited state is 10^{-8} sec, autoionization can reduce it to 10^{-14}–10^{-15} sec and it is then autoionization that determines the total width of an autoionizing state.

The influence of external electromagnetic radiation on autoionizing states of atoms is not only of theoretical but also of practical interest. For example, autoionizing states of atoms and transitions to the continuum have become particularly important in the processes of laser isotope separation and generation of coherent radiation in the vacuum ultraviolet range.[12,13]

An autoionizing state in an external electromagnetic radiation field was investigated in Ref. 6 and it was shown there that narrowing of the photoelectron spectrum occurs near a Fano minimum.[14,15] Photoionization from an autoionizing state and degeneracy of the continuum are allowed for in Refs. 2 and 3. Inclusion of these transitions, which always occur, has the effect that complete narrowing of resonances is impossible and the minimum width increases on increase in the external field intensity. A theory describing radiative decay of an autoionizing state of an atom in an external electromagnetic field is developed in Refs. 9 and 10. This theory allows systematically for radiative decay of the unperturbed continuum. A detailed analysis is made of the effects of radiative decay on the Fano profiles and photoelectron spectra. An analysis is also made of the fluorescence originating from an autoionizing state in an external field.

An external electromagnetic field may give rise to autoionization-like resonances in the continuous spectrum of a one-electron atom. A study of these resonances is desirable because the width, energy, etc., of these autoionization-like resonances depend on the intensity and frequency of the external field. A theoretical analysis of such resonances can be found in many papers, for example, in Refs. 16 and 17.

We shall consider the influence of an external electromagnetic field on autoionization resonances and consider the possibility of narrowing these resonances in the case of N closely spaced levels below the first ionization threshold of an atom, which are in a multiphoton resonance with the ground state, and M autoionizing levels above this threshold. The multilevel structure of a discrete state can be in the form of a large number of closely spaced excited levels or it may form a multiplet. In real atoms one frequently encounters a situation when the frequency of laser radiation is close to a resonance with a group of closely spaced autoionizing levels. Such a situation occurs, for example, in the Sr atom where the closely spaced discrete levels can be the highly excited states $5s10s(^3S)$, $5s11s(^3S)$, $5s7s(^3S)$, etc. and the closely spaced autoionizing levels can be multiplets of the $4d\,6p(^3P)$ state. We shall consider also multichannel decay of autoionizing and discrete states under the influence of a laser field. As first shown in Refs. 2 and 3, noninterfering channels have a considerable influence on the final results. A discussion of the influence of these channels can be found in §4. In calculations we shall ignore spontaneous transitions in an atom and assume that the laser field is monochromatic. Spontaneous transitions and the finite duration of application of the laser field may result in partial narrowing (down to the larger of the widths of radiation and laser pulses).

§1. GENERAL ANALYSIS

For the sake of simplicity we shall assume that the continuum is nondegenerate and that noninterfering transitions, such as photoionization from autoionizing states or a transition to a second continuum (when an ion is in an excited state), are absent. The influence of these transitions will be discussed in §4.

We shall consider a many-electron atom in the field of strong electromagnetic radiation. The first field couples the ground state to discrete levels ν and the second field couples intermediate levels ν to autoionizing states μ and to the continuum (Fig. 1).

The basis wave functions for the discrete spectrum of an

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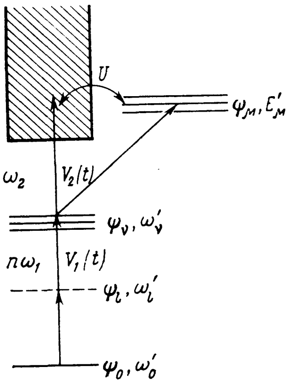

FIG. 1.

atom in a field are assumed to be quasienergy wave functions derived in the approximation of a multiphoton resonance in the case when the following periodic perturbation is applied adiabatically:

$$V_1(t)=V_1^+e^{i\omega_1 t}+V_1^-e^{-i\omega_1 t}. \qquad (1)$$

These functions are described by

$$\Phi_0(t)=\exp\Big[-\frac{i}{\hbar}\lambda_0 t\Big]\Big(C_0^0\psi_0+e^{-in\omega_1 t}\sum_i C_i^0\psi_i\Big),$$

$$\Phi_k(t)=\exp\Big[-\frac{i}{\hbar}(\lambda_k-n\hbar\omega_1)t\Big]\Big(C_0^k\psi_0+e^{-in\omega_1 t}\sum_i C_i^k\psi_k\Big),$$

$$\Phi_\mu(t)=\exp\Big[-\frac{i}{\hbar}E_\mu' t\Big]\psi_\mu$$

$$\times(i,k=0,\ldots,N;\ \mu=N+1,\ldots,N+M), \qquad (2)$$

where λ_ν ($\nu=0,1,\ldots,N$) are the quasienergies of an atom in an electromagnetic radiation field, which are described by an equation of degree $N+1$. When the interaction is switched off, i.e., when $V_1(t)\to 0$, the quasienergy λ_ν reduces to the energy of levels of a free atom, and the wave functions are reduced to functions of a free atom:

$$\Phi_\nu(t)\to\psi_\nu e^{-i\omega_\nu' t}\quad(\nu=0,\ldots,N). \qquad (3)$$

Discrete states ψ_k ($k=0,\ldots,N$) have energies below the first ionization threshold and states ψ_μ ($\mu=N+1,\ldots,N+M$) have energies above this threshold.

The Schrodinger equation for this problem is as follows:

$$i\hbar\frac{\partial\Psi}{\partial t}=(H_0+V_1(t)+V_2(t)+U)\Psi(t), \qquad (4)$$

where H_0 is the free Hamiltonian, V_2 is the interaction with an ionizing electromagnetic field, and U is the "configurational" Fano interaction.[14] The complete solution of the Schrödinger equation allowing for the continuum will be represented as follows:

$$\Psi(t)=\sum_k a_k(t)\Phi_k(t)+\int d\lambda a_\lambda(t)\varphi_\lambda(t), \qquad (5)$$

where $\varphi_\lambda(t)=\varphi_\lambda\exp(-i\lambda t/\hbar)$ is the unperturbed wave function for a continuous spectrum of an atom of energy λ.

Substituting Eq. (5) into Eq. (4), we obtain a system of differential equations for the coefficients $a_k(t)$ ($k=0,\ldots,N+M$) and $a_\lambda(t)$, which is converted by the Fourier transformation

$$a_k(t)=\exp\Big[\frac{i}{\hbar}E_k t\Big]\int dE\,a_k(E)\exp\Big[-\frac{i}{\hbar}Et\Big],$$

$$a_\lambda(t)=\exp\Big[\frac{i}{\hbar}\lambda t\Big]\int dE\,a_\lambda(E)\exp\Big[-\frac{i}{\hbar}Et\Big]$$

$$\times(k=0,\ldots,N+M), \qquad (6)$$

where

$$E_k=\begin{cases}\lambda_0+n\hbar\omega_1+\hbar\omega_2, & k=0,\\ \lambda_k+\hbar\omega_2, & k=1,\ldots,N,\\ E_k', & k=N+1,\ldots,N+M,\end{cases} \qquad (7)$$

to a system of algebraic equations for their Fourier transforms. The solution of these algebraic equations can be found in Ref. 18. Using the results of Ref. 18, we shall describe the complete system of orthonormalized quasienergy wave functions by

$$\Psi_E(t)=z(E)\Big(\frac{1}{z^2(E)+\pi^2}\Big)^{1/2}\exp\Big[-\frac{i}{\hbar}Et\Big]$$

$$\times\Big\{\sum_n\Big(\sum_m\frac{A_{nm}\tilde\vartheta_{Em}}{E-\tilde E_m}\Big)$$

$$\times\exp\Big[\frac{i}{\hbar}E_n t\Big]\Phi_n(t)+\int d\lambda\exp\Big[\frac{i}{\hbar}\lambda t\Big]$$

$$\times\Big[\frac{P}{E-\lambda}\sum_k\frac{\tilde\vartheta_{\lambda k}\tilde\vartheta_{Ek}^*}{E-\tilde E_k}+\delta(E-\lambda)\Big]\varphi_\lambda(t)\Big\}, \qquad (8)$$

where

$$z(E)=2\pi\Big[\sum_{m=0}^{N+M}\frac{\tilde\Gamma_m(E)}{E-\tilde E_m}\Big]^{-1}, \qquad (9)$$

$$\Gamma_m(E)=2\pi|\tilde\vartheta_{mE}|^2, \qquad (10)$$

$$\tilde\vartheta_{mE}=\sum_n\vartheta_{nE}A_{nm}. \qquad (11)$$

The system of equations for the determination of the elements of a unitary matrix A and of $\tilde E_m$ is as follows:

$$E_m A_{mn}+\sum_k F_{mk}(E)A_{kn}=A_{mn}\tilde E_n, \qquad (12)$$

where

$$F_{mk}(E)=\vartheta_{mk}+P\int\frac{\vartheta_{\lambda m}\vartheta_{\lambda k}^*}{E-\lambda}d\lambda \qquad (13)$$

and P denotes the principal value. The matrix elements $\vartheta_{n\lambda}$ and ϑ_{nk} are described by the following expressions:

$$\vartheta_{n\lambda}=\begin{cases}\sum_k C_k^n V_{k\lambda}^{(2)}, & \text{if}\quad 0\leqslant n\leqslant N,\\ U_{n\lambda}, & \text{if}\quad n\geqslant N+1,\end{cases} \qquad (14)$$

$$\vartheta_{nk}=\begin{cases}0, & \text{if}\quad n\leqslant N.\quad k\leqslant N,\\ \sum_l C_l^n V_{lk}^{(2)} & \text{in the remaining cases.}\end{cases} \qquad (15)$$

where

$$V_{n\lambda}^{(2)}=\langle\psi_n|V_2^-|\varphi_\lambda\rangle,\quad V_{nk}^{(2)}=\langle\psi_n|V_2^-|\psi_k\rangle,\quad U_{n\lambda}=\langle\psi_n|U|\varphi_\lambda\rangle.$$

The general solution of the Schrödinger equation can be represented in the form of an expansion in functions $\Psi_E(t)$. We shall assume that the exciting resonance field is applied adiabatically, whereas the ionizing field is applied at a moment $t=0$. Then, at the time of application of the ionizing field an atom is in a quasienergy state Φ_0. The solution of the Schrödinger equation subject to this initial condition is

$$\Psi(t)=\int_{-\infty}^{\infty}\langle\Psi_E(0)\,|\,\Phi_0(0)\rangle\Psi_E(t)\,dE. \tag{16}$$

Using Eqs. (2) and (8), we find that the wave function is described by

$$\Psi(t)=\int_{-\infty}^{\infty}dE\exp\left[-\frac{i}{\hbar}Et\right]\frac{z^2(E)}{z^2(E)+\pi^2}\left(\sum_m\frac{A_{0m}\tilde{\vartheta}_{mE^*}}{E-\tilde{E}_m}\right)\times\left\{\sum_n\left(\sum_m\frac{A_{nm}\tilde{\vartheta}_{Em}}{E-\tilde{E}_m}\right)\times\exp\left[\frac{i}{\hbar}E_nt\right]\Phi_n(t)+\int d\lambda\exp\left[-\frac{i}{\hbar}\lambda t\right]\times\left[\frac{P}{E-\lambda}\sum_m\frac{\tilde{\vartheta}_{\lambda m}\tilde{\vartheta}_{Em}^*}{E-\tilde{E}_m}+\delta(E-\lambda)\right]\varphi_\lambda(t)\right\}. \tag{17}$$

We shall assume that all the matrix elements, including $F_{mn}(E)$, depend weakly on the energy E and that they can be regarded as constants. As pointed out in Ref. 19, the dependences of these functions on the energy E is indeed weak, since the characteristic interval for the variation of the functions ($\sim$Ry = 13.26 eV) is large compared with the values of the functions.

Projecting the wave function $\Psi(t)$ on $\Phi_n(t)$ $(n=0,...,N+M)$ and on $\varphi_\lambda(t)$, we find that $a_n(t)$ and $a_\lambda(t)$ are described by the following expressions:

$$a_n(t)=\int_{-\infty}^{\infty}dE\,\frac{\alpha_0^*(E)\alpha_n(E)}{f(E)f^*(E)}\exp\left[-\frac{i}{\hbar}(E-E_n)t\right], \tag{18}$$

$$a_\lambda(t)=\int_{-\infty}^{\infty}dE\,\frac{\beta(E)\alpha_0^*(E)}{f(E)f^*(E)}\frac{e^{-i(E-\lambda)t/\hbar}}{E-\lambda-i0}+\frac{z(\lambda)}{z(\lambda)+i\pi}\sum_m\frac{A_{0m}{}^*\tilde{\vartheta}_m{}^*}{\lambda-\tilde{E}_m}, \tag{19}$$

where

$$\beta(E)=\sum_n\Gamma_n\left[\prod_{\substack{m\\ m\neq n}}(E-\tilde{E}_m)\right], \tag{20}$$

$$f(E)=\prod_m(E-s_m), \tag{21}$$

$$\alpha_n(E)=\sum_m A_{nm}\tilde{\vartheta}_m\left[\prod_{\substack{k\\ k\neq m}}(E-\tilde{E}_k)\right]. \tag{22}$$

The quantities s_m in Eq. (21) (where $m=0,...,N+M$) are roots of a complex equation of degree $N+M+1$:

$$f(E)=\prod_m(E-\tilde{E}_m)+\frac{i}{2}\sum_k\Gamma_k\left[\prod_{\substack{m\\ m\neq k}}(E-\tilde{E}_m)\right]=0, \tag{23}$$

which in turn is a characteristic equation of the matrix

$$\mathbf{D}=\|E_\nu\delta_{\nu\mu}+F_{\nu\mu}-i\pi\tilde{\vartheta}_{\nu E}{}^*\tilde{\vartheta}_{\mu E}\|. \tag{24}$$

It is shown in Ref. 18 that the roots of Eq. (23), which is of degree $N+M+1$, lie in the lower complex half-plane of E. Integration with respect to E in Eqs. (18) and (19) yields the following expressions for $a_n(t)$ $(n=0,...,N+M)$ and $a_\lambda(t)$:

$$a_n(t)=-2\pi i\sum_m\frac{\alpha_0{}^*(s_m)\alpha_n(s_m)}{f'(s_m)f^*(s_m)}\exp\left[-\frac{i}{\hbar}(s_m-E_n)t\right], \tag{25}$$

$$a_\lambda(t)=\sum_m\frac{\alpha_0{}^*(s_m)}{f'(s_m)}\frac{1-\exp[-i(s_m-\lambda)/\hbar]}{\lambda-s_m}. \tag{26}$$

We shall consider the case when all the roots of Eq. (23) are different; the case when there are some multiple roots will require a separate investigation.

In the limit $t\to\infty$ we find that the amplitude of the distribution of photoelectrons is described by

$$a_\lambda(\infty)=\sum_m\frac{\alpha_0{}^*(s_m)}{f'(s_m)}\frac{1}{\lambda-s_m}, \tag{27}$$

which shows that this amplitude has $N+M+1$ different maxima located at points $\lambda=\mathrm{Re}s_m$ and with widths $2\,\mathrm{Im}\,s_m$.

The total probability of ionization at a moment t is described by the following expression:

$$W(t)=\int_{-\infty}^{\infty}|a_\lambda(t)|^2\,d\lambda.$$

Substituting here $a_\lambda(t)$, we find that the total probability of ionization as a function of time t is described by

$$W(t)=2\pi i\sum_{m,n}\frac{\alpha_0{}^*(s_m)}{f'(s_m)}\left[\frac{\alpha_0{}^*(s_n)}{f'(s_n)}\right]^*\times\frac{1}{s_n{}^*-s_m}\left(1-\exp\left[\frac{i}{\hbar}(s_n{}^*-s_m)t\right]\right). \tag{28}$$

§2. EFFECTS OF NARROWING OF AUTOIONIZATION RESONANCES IN A STRONG ELECTROMAGNETIC FIELD

As pointed out in §1, the amplitude of the distribution of photoelectron energies has $N+M+1$ maxima at points $\lambda=\mathrm{Re}\,s_n$ with widths $2\,\mathrm{Im}\,s_n$. In the present section we shall show that the positions and widths of these maxima (resonances) may depend not only on their atomic characteristics, but also on the intensity of an external laser field. For simplicity, we shall ignore noninterfering channels such as spontaneous decay,[9,10] photoionization from autoionizing states,[2,3] photodecay to a different continuum,[11] etc. We shall also assume that the laser field is monochromatic.[8] Photoionization from autoionizing state degeneracy of the continuum, and inelastic transitions will be discussed in §4.

In the present section we shall show that complete narrowing of resonances is possible under the assumptions made above.

We shall only consider the case when the eigenvalues of the matrix $\|E_n\delta_{nm}+F_{nm}\|$ are different. Narrowing occurs if Eq. (23) has at least one real root E. Substituting this real root E into Eq. (23) and separating the real and imaginary parts of the equation, we obtain

$$\prod_{n=0}^{N+M}(E-\tilde{E}_n)=0,\quad \sum_n \Gamma_n \prod_{\substack{m\\ m\neq n}}(E-\tilde{E}_m)=0. \tag{29}$$

It follows from the first equation in the system (29) that $E=\tilde{E}_k$ ($k=0,\ldots,N+M$) and then, substituting this into the second equation of the system (29), we obtain

$$\Gamma_k \prod_{\substack{m\\ (m\neq k)}}(\tilde{E}_k-\tilde{E}_m)=0. \tag{30}$$

Since we are assuming that all the values $\tilde{E}_k$ are different, it follows that

$$\Gamma_k=0, \tag{31}$$

whereas Eqs. (10) and (11) give

$$\sum_n \vartheta_n A_{nk}=0. \tag{32}$$

The condition (32) can be interpreted as follows. The second field mixes discrete levels causing "splitting" of each level (Autler-Townes effect). Each new "dressed" state contains "old" discrete states with amplitudes A. A transition to the continuum from each old state occurs proportionally to ϑ. Hence, we find that the effective matrix element which links a dressed state with the continuum is proportional to the sum $\Sigma_n \vartheta_n A_{nk}$; if this matrix element vanishes because of interference, it follows that narrowing of a dressed resonance takes place.

Moreover, we can show that the condition (32) is a generalization of the condition derived by Rzazewski and Eberly.[6] They stated that narrowing occurs if zero of the ionization amplitude coincides with the energy of a dressed state. In fact, if $\lambda=\tilde{E}_k$ is substituted into Eq. (27), we obtain

$$\alpha_0(\tilde{E}_k)=\tilde{\vartheta}_k A_{0k}\prod_{\substack{n\\ (n\neq k)}}(\tilde{E}_k-\tilde{E}_n)=0.$$

According to our hypothesis on the multiplicity of $\tilde{E}_k$, we obtain $\tilde{\vartheta}_k=0$ or $\Sigma_n \vartheta_n A_{nk}=0$.

Substituting the eigenvalues E_k into Eq. (12), we shall represent the condition (32) in a more convenient form. If all the roots $\tilde{E}_k$ of the characteristic equation

$$|(E_n-\tilde{E})\delta_{nm}+F_{nm}|=0 \tag{33}$$

are different, then—as is known—the coefficients A_{kn} are proportional to the cofactors of the determinant (33), where $\tilde{E}$ is replaced by the corresponding value of $\tilde{E}_n$. We shall denote these cofactors by Δ_{pk} [p is selected so that one of the cofactors Δ_{pk} ($k=0,\ldots,N+M$) differs from zero; the existence of such an index follows from the assumption about the multiplicity of $\tilde{E}$]. It follows from this assumption that the matrix $\|E_n\delta_{nm}+F_{nm}\|$ is of rank $N+M$. This allows us to write down

$$A_{kn}=C_n\Delta_{pk}(\tilde{E}=\tilde{E}_n). \tag{34}$$

where C_n are deduced from the condition of unitarity of the matrix A. Substituting Eq. (34) into Eq. (32), we obtain

$$\sum_k \vartheta_k \Delta_{pk}=0. \tag{35}$$

We can find the narrowing condition if we solve simultaneously Eqs. (35) and (33) or, in other words, Eqs. (35) and (33) should have at least one root in common. We can show that Eq. (35) applies irrespective of the index p. If we represent the left-hand sides of Eqs. (35) and (33) in the form of polynomials, we can readily formulate the condition of simultaneous narrowing of k resonances. This is true when and only when the matrix $(2n-k)\times(2(n-k)+1)$ ($n=N+M+1$), composed of coefficients of the polynomials (35) and (33), is of rank smaller than $2(n-k)+1$ (Ref. 20). It follows from this formalism that, in general, one can expect simultaneous narrowing of $N+M$ resonances. It should be pointed out that if the matrix D of Eq. (24) is normal, i.e., if $DD^+=D^+D$, then $k=N+M$.

We shall illustrate this method by considering the case when $N+M=1$ and $N+M=2$. The case when $N+M=1$, i.e., when there is one discrete level below the first ionization threshold of an atom and one level above this threshold, has been considered by many authors.[6-8] This case is illustrated schematically in Fig. 2.

In this case the matrix D of Eq. (24) is normal subject to the following condition which is both necessary and sufficient:

$$E_1-E_0=\frac{F_{01}}{\vartheta_0^*\vartheta_1}(|\vartheta_1|^2-|\vartheta_0|^2). \tag{36}$$

It follows from Eq. (36) that there are always such frequencies of the laser field that narrowing cannot be achieved simply by altering the radiation intensity. Consequently, it would be of interest to investigate the narrowing of resonances by varying the radiation frequency.

If $N+M=2$, we can distinguish three different cases illustrated in Fig. 3. The cases labeled a and b are obtained if the first perturbing field is weak; then, a transition from the ground state of an atom can be allowed for by using perturbation theory in respect of the interaction V_1. In the case of narrowing effects we need to consider only the "upper part" of the ionization process shown in Figs. 3a and 3b. The case when the interaction V_1 mixes strongly the states 0 and 1 is illustrated in Fig. 3c.

It is particularly interesting to derive the condition ensuring that the amplitude of the distribution of photoelectrons obtained in the case of ionization exhibits simultaneous narrowing of two resonances. The necessary and sufficient condition is that the matrix D of Eq. (24) is normal. Hence, we obtain

$$E_1-E_0=F_{02}\vartheta_2^*/\vartheta_0-F_{21}\vartheta_2/\vartheta_1, \tag{37}$$

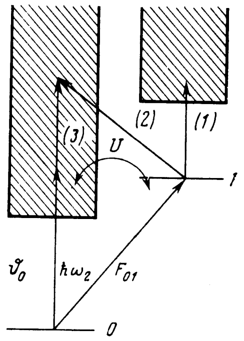

FIG. 2.

$$E_2-E_0=F_{02}\vartheta_2/\vartheta_0-F_{02}\vartheta_0/\vartheta_2-F_{12}\vartheta_1/\vartheta_2. \tag{38}$$

We can readily show that the condition (37) of simultaneous narrowing of two resonances is not satisfied in the case illustrated in Fig. 3c. In fact, it follows from Eqs. (14), (15) and (7) that

$$E_1-E_0=\lambda_1-\lambda_0-2\hbar\omega_1=0. \tag{39}$$

Substituting λ_1 and λ_0 of Ref. 21 into Eq. (39), we obtain

$$\varepsilon'^2+4|f_{01}|^2=0, \tag{40}$$

where ε' is two-photon detuning and f_{01} is an effective matrix element which couples atomic states ψ_0 and ψ_1. Since $f_{01}\neq 0$, it follows that the condition (40) cannot be satisfied. This is due to the fact that the state ψ_0 is not coupled directly to the continuum. We can similarly consider the cases when $N+M=3,4,\ldots$.

§3. GENERATION OF THE THIRD HARMONIC

We shall consider nonlinear mixing of frequencies of the $\omega_3=2\omega_1+\omega_2$ type for the scheme of transitions shown in Fig. 4. The interaction V_1 is in resonance with the two-photon transition $0\to 1$. For the sake of simplicity, we shall allow for V_1 using perturbation theory and include the interaction V_2, which couples the states 1 and 2, in a resonance approximation. In the present section we shall show that when the condition (36) is satisfied, the signal at the frequency $2\omega_1+\omega_2$ increases. This is due to the fact that the ionization losses decrease[7] if the condition (36) is satisfied.

A complex nonlinear polarization $P_{\omega 3}(t)$ at the frequency $\omega_3=2\omega_1+\omega_2$ is given by

$$P_{\omega_3}(t)=\left\{a_0(t)C_0^0\exp\left[-\frac{i}{\hbar}\lambda_0 t\right]+a_1(t)C_0^1\exp\left[-\frac{i}{\hbar}(\lambda_1-2\hbar\omega_1)t\right]\right\}\times\left\{a_2^*(t)\exp\left[\frac{i}{\hbar}E_2't\right]\mu_{02}+\int d\lambda\exp\left[\frac{i}{\hbar}\lambda t\right]a_\lambda^*(t)\mu_{0\lambda}\right\}. \tag{41}$$

Here the coefficients $a_n(t)$ ($n=0,1,2,\lambda$) are found from the relationships (25) and (26); μ_{02} is a matrix element of the dipole moment between the ground and autoionizing levels; $\mu_{0\lambda}$ is a matrix element of the dipole moment of a transition from the ground state to the continuum. When the first resonance interaction is weak, i.e., in the limit $V_1\to 0$, it follows from Eq. (2) that

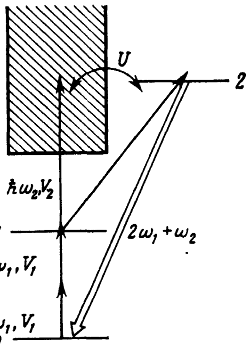

FIG. 4.

$$C_0^0\to 1,\ C_0^1\to 0,\ C_1^1\to 1,\ C_1^0\to 0,\ \lambda_0\to\omega_0',\ \lambda_1\to\omega_1'. \tag{42}$$

If we retain in Eqs. (25) and (26) only the terms of the first order in V_1 and assume that the interaction time satisfies the conditions

$$\hbar/|\operatorname{Im} s_{1,2}|\ll t\ll\hbar/|\vartheta_0|^2, \tag{43}$$

where $\vartheta_0=C_1^0\vartheta_1$, we find that simple transformations yield $a_0(t)=1$,

$$a_2(t)=\frac{f_{01}}{f'(E_0)}(F_{21}-i\pi\vartheta_1\vartheta_2^*)\exp\left[-\frac{i}{\hbar}(E_0-E_2)t\right],$$

$$\int d\lambda a_\lambda(t)\exp\left[-\frac{i}{\hbar}\lambda t\right]=-\frac{-i\pi f_{01}}{f'(E_0)}[\vartheta_1(E_0-E_2)+\vartheta_2F_{12}]\exp\left[-\frac{i}{\hbar}E_0t\right], \tag{44}$$

where

$$f'(E_0)=(E_0-s_1)(E_0-s_2),$$

whereas f_{01} is an effective two-photon matrix element coupling the states 0 and 1 (Ref. 21). Substituting Eq. (44) into Eq. (41), we find that $P\omega_3(t)$ is described by

$$P_{\omega_3}(t)=\exp[i(2\omega_1+\omega_2)t]\frac{f_{01}^*}{(E_0-s_1^*)(E_0-s_2^*)}\times\{\mu_{02}(F_{12}+i\pi\vartheta_1^*\vartheta_2)+i\pi\mu_{0\lambda}[\vartheta_1^*(E_0-E_2)+\vartheta_2^*F_{21}]\}. \tag{45}$$

The expression (45) and the narrowing condition (36) were obtained in Ref. 7. However, the condition (36) was deduced in Ref. 7 on the assumption that the interaction

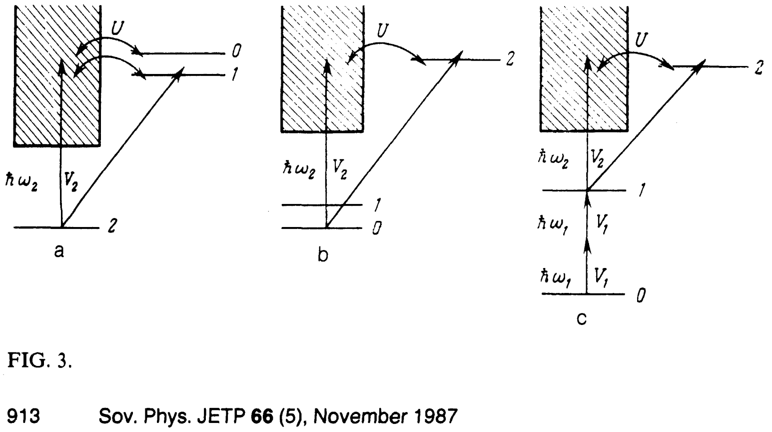

FIG. 3.

between the levels 1 and 2 is many times greater than the Fano configurational interaction.[14] It is clear from the results of the preceding section that the condition (36) is independent of this assumption.

If the two-photon interaction V_1 is weak, it follows—as pointed out already—that only the "upper part" of the ionization process (Fig. 4) is important in the narrowing and the field frequency is then to be found from the expression

$$E_2-E_1=\vartheta_2F_{12}/\vartheta_1-\vartheta_1F_{21}/\vartheta_2. \quad (46)$$

If the condition (46) is satisfied, the polarization of an atom reaches its maximum when

$$E_1-E_0=\vartheta_1F_{21}/\vartheta_2. \quad (47)$$

It follows from Eqs. (46) and (47) that the frequency of the output radiation is

$$2\omega_1+\omega_2=\hbar^{-1}(E_2'-\hbar\omega_0'-\vartheta_2F_{12}/\vartheta_1). \quad (48)$$

If a two-photon resonance transition is strong or the conditions of Eq. (43) are not obeyed, an analysis of the final results becomes very difficult and further calculations for these cases must be carried out numerically on a computer.

§4. INFLUENCE OF NONINTERFERING DECAY CHANNELS ON THE RESONANCE NARROWING EFFECTS

As pointed out in Refs. 2 and 3, the noninterfering channels of decay of autoionizing states can alter significantly the results obtained in §2. If a transition from autoionizing states to the second continuum (when an ion is in an excited state) occurs only at a specific frequency of a laser field, the photoionization with a transition to this continuum and the degeneracy of the continuum always occur. However, the photoionization from autoionizing states [such as transition (2) in Fig. 2], which is due to a two-electron dipole transition, is forbidden in the one-configuration approximation because autoionizing states belong to a shifted term of an atom (when two or more electrons are excited simultaneously). Little study has been made of such transitions and they can be resolved only if we allow for the configurational interaction. The matrix elements for such transitions consist of two parts. The first (factorized) part describes transitions following the absorption of an additional photon in the continuous spectrum [such as transition (3) in Fig. 2] after autoionization. Such a transition can occur and experimental investigations of the above-threshold ionization[22] demonstrate that a free-free transition $V_{EE'}$ is really observed. It reduces by a factor $[1+\pi^2|V_{EE'}|^2]^{1/2}$ the matrix elements of transitions[23] from discrete and autoionizing states to the first (lower) continuum[23] and gives rise to an additional peak, proportional to $|V_{EE'}|^2$, in the photoelectron distribution. The second part of the matrix element which corresponds to a two-electron transition is due to an integral representing the principal value in the composite matrix element and is clearly small. Usually the oscillator strengths of two-electron transitions are 10^{-3}–10^{-4} (Refs. 11 and 15).

In the present section we shall consider the influence of these channels on the narrowing condition. We shall do this by investigating the behavior of the eigenvalues of the matrix D of Eq. (24):

$$D=\left\|E_n\delta_{nm}+F_{nm}-i\pi\sum_i \vartheta_n^{*(i)}\vartheta_m^{(i)}\right\|, \quad (49)$$

where $\vartheta_n^{(i)}$ is a matrix element which couples a discrete or autoionizing state (n) to the continuum (i).

It is shown in Ref. 18 that the imaginary parts of the eigenvalues of the matrix D of Eq. (49) lie within the interval from $\min\{\lambda_c\}$ to $\max\{\lambda_c\}$, where λ_c are the eigenvalues of the matrix C which is of the following form:

$$C=-{}^1/_2 i(D-D^+)=-\pi\left\|\sum_i \vartheta_n^{*(i)}\vartheta_m^{(i)}\right\|. \quad (50)$$

Since each $-\pi\|\vartheta_n^{*(i)}\vartheta_m^{(i)}\|$ matrix is of rank 1, the rank of the matrix C is $r_c\le K$, where K is the number of continuums. Hence, it follows that the matrix C has $N+M+1-K$ zero eigenvalues. Consequently, in narrowing of resonances it is essential that the number K of the continuum should be less than the number of discrete levels in the continuum. However, it is difficult to achieve such a situation unless such "selection rules" as those suggested by Bethe[15] reduce the number of continuums.

For the sake of simplicity we shall consider a scheme (Fig. 3b) allowing for photoionization from an autoionizing state or for photodecay from this state. Simple calculations give the following narrowing conditions:

$$E=E_0,\quad F_{20}=F_{21}(\Gamma_0/\Gamma_1)^{1/2}. \quad (51)$$

The second condition of Eq. (51) can be satisfied at random, since it is independent of an external magnetic field. The condition (51) can be derived on the basis of the following considerations. An external field mixes discrete states with amplitudes A. One of the states becomes narrower if $A_{i2}=0$ (Fig. 3b) if naturally the matrix element of the photoionization does not vanish, and moreover if $A_{i1}=-A_{i0}\vartheta_0/\vartheta_1$. The condition (51) is obtained by substituting these elements into Eq. (12).

We shall now consider the case when there is one autoionizing level and one discrete level, and photoionization takes place [transition (2) in Fig. 2)][2,3] or the transition (1) to the second continuum takes place from an autoionizing state.[11] In this case the matrix D becomes

$$D=\begin{Vmatrix}E_0-i\Gamma_0/2 & F_{01}-i(\Gamma_0\Gamma_1)^{1/2}/2\\ F_{10}-i(\Gamma_0\Gamma_1)^{1/2}/2 & E_1-i(\Gamma_1+\Gamma_i)/2\end{Vmatrix}, \quad (52)$$

where Γ_i is the photoionization width of an autoionizing state. In this case the matrix C of Eq. (50) becomes

$$C=\begin{Vmatrix}-\Gamma_0/2 & -(\Gamma_0\Gamma_1)^{1/2}/2\\ -(\Gamma_0\Gamma_1)^{1/2}/2 & -(\Gamma_1+\Gamma_i)/2\end{Vmatrix}, \quad (53)$$

whereas the eigenvalues are

$$\lambda_c^{(\pm)}=-{}^1/_4\{\Gamma_1+\Gamma_i+\Gamma_0\pm[(\Gamma_1+\Gamma_i-\Gamma_0)^2+4\Gamma_i\Gamma_0]^{1/2}\}. \quad (54)$$

The narrowing condition can be obtained if we assume that the matrix (52) is normal, which gives

$$E_1-E_0=F_{01}(\Gamma_1+\Gamma_i-\Gamma_0)/(\Gamma_0\Gamma_1)^{1/2}. \quad (55)$$

This condition is identical with the conditions in Refs. 2 and 3. Since the matrix $(-C)$ is positive definite, it follows that an increase in the intensity increases the eigenvalues, i.e., the minimum width increases the intensity of the external field.

CONCLUSIONS

We investigated narrowing of the photoelectron spectrum and the influence of such narrowing on generation of the third harmonic. Ignoring spontaneous transitions and using the approximation of a monochromatic wave, we found the conditions for narrowing of the photoelectron spectrum in respect of the detuning and intensity of the external field. However, as first shown in Refs. 2 and 3, in the presence of noninterfering channels such as a two-electron transition to the first continuum and a one-electron transition to a higher second continuum accompanied by the absorption of a photon, or in the case of degeneracy of the continuums we found that the minimum width obtained due to interference of the channels rises monotonically with intensity of the external field, i.e., there is no narrowing of the intensity in the photoelectron spectrum.

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