

Nonlinear polarization resonances in the continuum

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The influence of nonlinear autoionization-like resonances, induced in the continuum by strong optical radiation, on the optical activity of the transitions to the continuum is investigated. The role of the distribution of the oscillator strengths in the continuum and the effect of the spin-orbit interaction on the induced optical activity of the medium is made clear. It is shown that the largest effect should be expected in the region of the minima of the photoabsorption in the continuum (Cooper minima). Allowance for the spin-orbit interaction in the continuum leads to a dependence of the principal parameters of the nonlinear resonance on the projection of the total angular momentum, and affects adversely the optical activity of the medium. The possibilities of experimentally observing nonlinear autoionization-like resonances in cesium vapor and of nonlinear spectroscopy of transitions between the discrete and continuous spectra are discussed, as is the possibility of measuring some integral characteristics of the continuum.

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1. INTRODUCTION

Progress in the development of short-wave lasers has stimulated interest in the study of high-lying states of atoms and molecules, including autoionization levels and states of the continuous spectrum (continuum), which were investigated mainly with the aid of noncoherent radiation sources. Recently the high-lying states of atoms and molecules have assumed particular importance in connection with problems of laser isotope separation and generation of coherent vacuum-ultraviolet and ultrasoft x-radiation by methods of nonlinear frequency mixing. The solution of this problem is usually connected with transitions in the continuum and is determined by its characteristics.

It is known that the largest fraction of the sum of the oscillator strengths in atoms (with the exception of hydrogen) goes to the continuous spectrum (see, e.g., Ref. 1), but the differential oscillator strengths, as a rule, are small, and this imposes definite limitations, for example, on the effectiveness of laser isotope separation and on the value of the nonlinear susceptibility. The possibility of inducing in gaseous media autoionization-like resonances in the continuum by strong laser radiation was demonstrated in Refs. 2-6. These resonances make it possible to increase significantly the efficiency of the indicated processes in a number of cases. Allowance for the laser-induced perturbation of the continuum states may turn out to be important also in the spectroscopy of solids⁷ and in the study of multiphoton ionization of atoms.⁸ In addition, as shown in Refs. 5 and 9, nonlinear autoionization-like resonances can alter substantially the polarization and the angular distribution of the electrons, and this of interest both from the viewpoint of photoelectron spectroscopy and for the development of sources of polarized electrons.

The experimental investigation of nonlinear resonances in absorption on transitions to the continuum is made difficult mainly by two factors.

First, the small absorption coefficients make it necessary to use extended gas media with relatively high gas pressures. Second, it is quite difficult to ensure a high intensity of the laser radiation over the entire

length of the medium. It is therefore particularly attractive to use the highly sensitive methods of nonlinear polarization spectroscopy to study induced resonances in the continuum.

Methods of polarization spectroscopy have by now been sufficiently developed in detail for discrete atomic transitions, and make it possible to investigate subtle nonlinear spectroscopic phenomena with high degree of accuracy.¹⁰⁻¹² This article considers, for the first time ever, nonlinear polarization spectroscopy phenomena as applied to transitions to the continuum.

2. NONLINEAR RESONANCE OF POLARIZATION ROTATION

We consider first the transition scheme shown in Fig. 1. The field E is strong and is at resonance with the transition between the discrete unpopulated level n and the continuum state ε . The nonlinear autoionization-like resonance produced by such a field in photoabsorption at a frequency ω_0 and in the nonlinear susceptibility was investigated for the process $\omega_0 = 2\omega_1 + \omega_2$ in Refs. 2-6 and 9.

We consider the influence of the field E on the refractive index $n(\omega_0)$ at the frequency ω_0 . If the field E is circularly polarized, then $n(\omega_0)$ is different for right- and left-circularly polarized probing radiation of amplitude E_0 [$n_+(\omega_0) - n_-(\omega_0) \neq 0$]. The action of the field E will rotate the plane of polarization of the linearly polarized radiation E_0 . We shall investigate the dependence of the

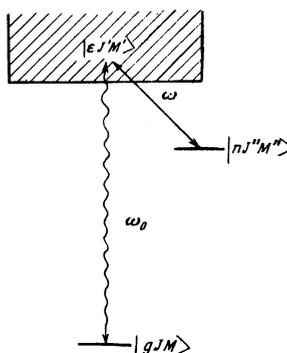


FIG. 1. Transition scheme for laser-induced nonlinear autoionization-like resonances.

rotation angle on the frequency ω_0 at a fixed frequency ω .

In the nonlinear-resonance region $\omega_0 \approx \omega_{ng} + \omega$ the rotation angle θ is described by the expression

$$\theta = \frac{\omega_0 L}{2c} (n_+ - n_-) = \pi \frac{\omega_0}{c} L \operatorname{Re}(\chi_+ - \chi_-), \quad (1)$$

where L is the length of the medium, and χ_{\pm} is the susceptibility of the frequency ω_0 for right- and left-circularly polarized radiation. The quantization axis is chosen along the direction of propagation of the probing radiation. In addition, the difference between the refractive indices for the right- and left-polarized radiation changes the radiation polarization from linear to elliptical. The ratio of the minor and major semi-axes of the ellipse is

$$a = \frac{L}{4} (\alpha_+ - \alpha_-); \quad \alpha_{\pm} = 4\pi \frac{\omega_0}{c} \operatorname{Im} \chi_{\pm}. \quad (2)$$

We determine $\chi_{\pm}(\omega_0)$ for the transition scheme shown in Fig. 1. We put

$$|g\rangle = |gJM\rangle, \quad |n\rangle = |nJ'M''\rangle, \\ |e\rangle = |ej\rangle, \quad j = \{J'M'\}.$$

The susceptibility $\chi(\omega_0)$ at the frequency of the probing field ω_0 in the region of the nonlinear resonance is determined from the formula

$$\chi(\omega_0) = \frac{N}{E_0} \sum_j \int d_{eg}^j r_{eg}^j d\varepsilon, \quad (3)$$

where N is the concentration of the atoms, d_{eg}^j and r_{eg}^j are respectively the matrix element of the dipole moment and the component of the density matrix at the frequency ω_0 . The summation in (3) extends over all the states of the continuum. Integration with respect to ε implies also summation over all the nonresonant discrete states.

To determine the components of the density matrix

$$\rho_{eg}^j = r_{eg}^j \exp\{-i(\omega_0 - \omega_{eg})t\}$$

it is convenient to use the system of equations

$$i\dot{\rho}_{eg}^j(M) = G_{eg}^j(M) \exp\{-i(\omega_0 - \omega_{eg})t\} \rho_{gg}(M) \\ + \sum_{M''} G_{en}^j(M'') \exp\{-i(\omega_0 - \omega_{en})t\} \rho_{ng}(M''M), \quad (4a)$$

$$i\dot{\rho}_{ng}(MM'') + i\Gamma_{ng} \rho_{ng}(M''M) = \sum_j \int G_{ne}^j(M'') \exp\{i(\omega_0 - \omega_{ne})t\} \rho_{eg}^j(M) d\varepsilon. \quad (4b)$$

Here

$$G_{eg}^j(M) = -E_0 \mathbf{e}_0 d_{eg}^j(M) / 2\hbar, \quad G_{en}^j(M'') = -E \mathbf{e} d_{en}^j(M'') / 2\hbar$$

are the matrix elements of the electric-dipole interaction with the radiation fields, Γ_{ng} is the radiative half-width of the discrete transition ng , and \mathbf{e}_0 and \mathbf{e} are the polarizations of the fields E_0 and E . Equations (4) are written in an approximation wherein the field E_0 does not change the population of the level g , and we can assume henceforth

$$\sum_M \rho_{gg}(M) \approx 1.$$

From the quasistationary solution of the system (4), the condition for whose applicability is given below, we obtain by using (3)

$$\frac{\omega_0}{c} (n_+ - n_-) = \frac{1}{2} \sum_M \alpha_0(M) \rho_{gg}(M) \left[q_{gg} + \sum_{M''} \beta(M, M'') D(M'') \right], \quad (5)$$

$$\alpha(\omega_0) = \sum_M \alpha_0(M) \rho_{gg}(M) \left[1 + \sum_{M''} \beta(M''M) F(M'') \right], \quad (6)$$

where $\alpha_0(M) = \sum_j \alpha_j^0(M)$ is the coefficient of absorption from the sub-level M in the absence of the field E , and the dependence of the resonance amplitudes on E is represented by the parameter β :

$$\beta(MM'') = [\gamma_{ng}(MM'')]^2 / [\Gamma_{ng} + \gamma_{nn}(M'')] \gamma_{gg}(M);$$

$$D(M'') = \frac{(q_{ng}^2 - 1)x(M'') - 2q_{ng}}{1 + x^2(M'')},$$

$$F(M'') = \frac{q_{ng}^2 - 1 + 2q_{ng}x(M'')}{1 + x^2(M'')},$$

$$q_{ng} = \delta_{nk'} / \gamma_{nk'}, \quad \gamma_{nk'} = \pi \hbar \sum_j G_{nk'}^j G_{ek'}^j |_{\varepsilon = \omega_0},$$

$$\delta_{nk'} = 2 \sum_j \frac{G_{nk'}^j G_{ek'}^j}{\omega_0^2 - \omega_{jg}^2} \omega_{jg} + \frac{1}{\pi} \int \frac{\gamma_{nk'}(\varepsilon)}{\hbar \omega_0 - \varepsilon} d\varepsilon;$$

the quantity

$$x(M'') = (\omega_0 - \omega - \omega_{ng} - \delta_{nn}(M'')) / (\Gamma_{ng} + \gamma_{nn}(M''))$$

is the deviation of the frequency ω_0 from the quasi-level frequency $\tilde{\omega}(M'') = \omega + \omega_{ng} + \delta_{nn}(M'')$, referred to its half-width $\Gamma(M'') = \Gamma_{ng} + \gamma_{nn}(M'')$. The dependence of the frequency and width of the resonances on M'' reflects the effect of lifting, in the field of the laser radiation, the degeneracy in the projections of the total angular momentum. The quantity $\beta(M, M'')$ is analogous in the present case to the parameter ρ^2 of the theory of autoionization resonances¹ and can be represented in the form of the square of the overlap integral of the two effective wave functions

$$\Psi_g(M) = \frac{1}{[\gamma_{gg}(M)]^{1/2}} \sum_j G_{ge}^j(M) \Psi_{e_0}^j,$$

$$\Psi_n(M'') = \frac{1}{[\Gamma_{ng} + \gamma_{nn}(M'')]^{1/2}} \sum_j G_{ne}^j(M'') \Psi_{e_0}^j,$$

where $\varepsilon_0 = \hbar \omega_0$. In the general case of several different continuums j we have $\beta(MM'') \leq 1$.

The rate of departure from the stage g into the continuum in the absence of the field E is determined by the quantity γ_{gg} , and therefore the quasistationary approximation is valid for times t that satisfy the condition¹³

$$\gamma_{gg}^{-1} \gg t \gg (\gamma_{nn} + \Gamma_{ng})^{-1}.$$

In addition, it is necessary to satisfy the condition of the perturbation theory with respect to two-photon transitions to the level n :

$$\gamma_{nn} + \Gamma_{ng} \gg \delta_{ng}.$$

Neglecting the spin-orbit interaction in the continuum, the quantities $q_{nk'}$ do not depend on the projections of the total angular momentum, so that Eqs. (5) and (6) can be represented in the form

$$\theta = \frac{1}{2} L \sum_{MM''} \rho_{gg}(M) D(M'') [\alpha_0^+ \beta_+(MM'') - \alpha_0^- \beta_-(MM'')], \quad (7)$$

$$a = \frac{1}{2} L \sum_{MM''} \rho_{gg}(M) F(M'') [\alpha_0^+ \beta_+(MM'') - \alpha_0^- \beta_-(MM'')]. \quad (8)$$

Thus, the anisotropy of the medium is determined only

by the difference between the parameters $\alpha_0^\pm \beta_\pm(MM'')$
 $\sim [\gamma_{ng}^\pm(M''M)]^2$.

Let us determine the quantities $\gamma_{ng}(M''M)$, $\gamma_{nn}(M'')$, and $\gamma_{gg}(M)$ contained in (7) and (8) as functions of the characteristics of the medium and of the radiation polarizations. Using the formulas of Ref. 14, it is convenient to represent these quantities in the form

$$\gamma_{ng}(M''M) = \frac{(-1)^{J''+J}}{(2J''+1)^{1/2}} \sum_{KQ} \{(-1)^K (2K+1)^{1/2} C_{JM''KQ}^{J''M''} \bar{\gamma}_{ng}(K) (\mathbf{e}' \times \mathbf{e}_0)_{-q^K}\}, \quad (9a)$$

$$\gamma_{nn}(M'') = \frac{(-1)^{2J''}}{(2J''+1)^{1/2}} \sum_K \{(-1)^K (2K+1)^{1/2} C_{JM''K0}^{J''M''} \bar{\gamma}_{nn}(K) (\mathbf{e}' \times \mathbf{e})_0^K\}, \quad (9b)$$

$$\gamma_{gg}(M) = \frac{(-1)^{2J}}{(2J+1)^{1/2}} \sum_K \{(-1)^K (2K+1)^{1/2} C_{JM K0}^{JM} \bar{\gamma}_{gg}(K) (\mathbf{e}_0 \times \mathbf{e})_0^K\}, \quad (9c)$$

where $\bar{\gamma}_{ng}(K)$, $\bar{\gamma}_{nn}(K)$ and $\bar{\gamma}_{gg}(K)$ are expressed in terms of a $6j$ -symbol and of the reduced matrix elements $G_{n\epsilon_0}(J''J')$ and $G_{g\epsilon_0}(J, J')$. For example, $\bar{\gamma}_{ng}(K)$ is defined by the formula

$$\bar{\gamma}_{ng}(K) = \sum_{J'} \left\{ \begin{matrix} 1 & 1 & K \\ J & J' & J' \end{matrix} \right\} G_{n\epsilon_0}(J''J') G_{g\epsilon_0}(J'J).$$

The quantities $\bar{\gamma}_{nn}(K)$ and $\bar{\gamma}_{gg}(K)$ are similar in form. The components of the tensor product of the polarizations of the radiation \mathbf{e} and \mathbf{e}_0 are determined by the Clebsch-Gordan coefficients:

$$(\mathbf{e}' \times \mathbf{e}_0)_{-q^K} = C_{-q, -q}^{KQ} (-1)^{q'}.$$

In the case of right-polarized radiation at the frequency ω we have $q' = 1$ and $q = \pm 1$ respectively for right- and left-polarized component of the probing radiation. In the electric-dipolar approximation, K ranges from 0 to 2, and Q from $-K$ to K ; in the summation over Q only the terms with $Q = q - q'$ differ from zero.

If the wave vectors of the radiations are not collinear, then the nonlinear resonances can lead to birefringence of the medium. Choosing the quantization axis along the direction \mathbf{k}_0 , we can write the components of the tensor product of the polarizations in (9) in the form

$$(\mathbf{e}' \times \mathbf{e}_0)_{-q^K} = \sum_{q'q_0} (-1)^{q'} C_{-q, -q_0}^{Kq} d_{q'q_0}^{(1)}(\varphi),$$

where φ is the angle between the directions of propagation of the radiation, and the value of q'_0 is defined in the old coordinate system. For right-polarized radiation at the frequency ω we have $q'_0 = 1$. If the strong radiation is linearly polarized, then q'_0 takes on values ± 1 ; $d_{q'q_0}^{(1)}(\varphi)$ is the finite-rotation matrix.¹⁴

In the case of the transition scheme of Fig. 2 the non-collinearity of the radiation leads to a deviation of β_- from 0, which can lead to a decrease in the angle of rotation of the polarization plane. If $\varphi \ll 1$, however, the angle θ can decrease only by small amount $\sim \theta\varphi$.

3. POSSIBLE EXPERIMENTAL REALIZATIONS

Let us analyze the possibilities of an experimental investigation, by the methods of polarization spectroscopy,

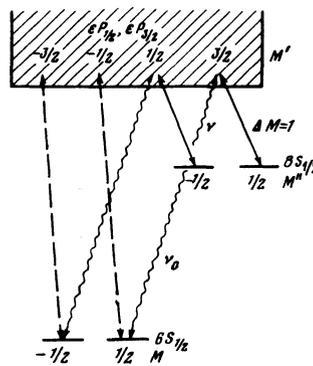


FIG. 2. Transition scheme for experimental observation of polarization resonances of cesium vapor in the continuum.

copy, of the nonlinear resonances in alkali-metal vapor in the continuum, using as an example the transition scheme shown in Fig. 2. In this case $J = J'' = 1/2$; $J' = 1/2, 3/2$; $q' = 1, q = \pm 1$, and consequently $M'' = M$ by virtue of the selection rules. The field E alters the characteristics of the medium only for the right-polarized component of the probing wave, i.e., $\beta_-(M) = 0$.

Without allowance for the spin-orbit interaction in the continuum, the only terms in (4) and (5) that differ from zero are those with $K = Q = 0$. The quantities $x(M)$ and $\beta_+(M)$ in (7) and (8) do not depend here on M . In a sufficiently strong field E , when the condition $\gamma_{nm} \gg \Gamma_{ng}$ is satisfied, we obtain $\beta_+ \approx 1$. The quantity $|D(M)|$ of (5) reaches a maximum equal to $1/2(1 + q_{ng})^2$ at $x = (1 - q_{ng})/(1 + q_{ng})$. At this frequency ω_0 the rotation of the plane of polarization is given by

$$\theta_{\max} = 1/8 \alpha_0 L (1 + q_{ng})^2 = 1/8 \sigma_0 N L (1 + g_{ng})^2, \quad (10)$$

where σ_0 is the cross section of photoionization from the states g in the absence of the field E , and N is the density of the atoms.

Absorption of the probing radiation at the same value of ω_0 , according to (6), is determined by the quantity $\sigma_0 N L (1 + q_{ng}^2)$, which should not exceed unity significantly. Under these conditions the maximum rotation is limited to a value of the order of 1 rad.

The required intensity of the strong laser radiation is determined only by the homogeneous or Doppler broadening of the discrete transition $6S_{1/2} - 8S_{1/2}$. In the case of cesium atoms, the homogeneous width of this transition is determined in the main by the probability of the transition from the level $8S_{1/2}$ to the nearest P states, and amounts to $\sim 10^6 \text{ sec}^{-1}$, while the Doppler width of this transition is $\sim 10^8 \text{ sec}^{-1}$.¹⁵ If the cross section for ionization from the state $8S_{1/2}$, is $\sigma \sim 10^{-19} \text{ cm}^2$ estimates yield for the intensity of the strong laser radiation a value $I \sim 10^6 - 10^8 \text{ W/cm}^2$. This intensity is readily attained by focusing the radiation of medium-power lasers. In alkali-metal vapor we have $\sigma_0 \sim 10^{-18} - 10^{-20} \text{ cm}^2$, and the vapor pressure is limited to $\sim 1 \text{ Torr}$, at which the fraction of the molecular components is still small ($\sim 1\%$).¹⁶ It is therefore natural to turn to the case $q_{ng}^2 \gg 1$. From the form of q_{ng}^2 it follows that the denominator in this expression is proportional to the product of the cross sections of the ionization from the states g and n , whereas the numerator is determined by a quan-

tity that is integral over the continuum (by the Hilbert transform of the denominator). One should therefore expect values $q_{ng}^2 \gg 1$ in the region of the photoabsorption maxima in the continuum.¹ At these energy values, however, it is important to take into account the spin-orbit interaction in the continuum.¹⁷ Let us see how this interaction alters (7) and (8).

4. INFLUENCE OF SPIN-ORBIT INTERACTION ON THE NONLINEAR-RESONANCE PARAMETERS

As shown in Refs. 17, the spin-orbit interaction in the continuum leads to a difference between the radial matrix elements for different total angular momenta J' in the continuum, and makes a noticeable contribution to the absorption spectra of alkali metals. Calculations by the Hartree-Fock method,¹⁷ and in the Coulomb approximation with different parameters of the quantum defect, predict a zero photoabsorption cross section in the first line of the series $nS - \epsilon P$ in a certain region of values of the energy ϵ in the continuum in the alkali atoms Na, K, Rb, Cs. Experiments on photoabsorption,¹⁸ however, yield a nonzero value of the cross section. In Ref. 17 this phenomenon was attributed to the fact that the radial matrix elements $R_{J'}$ for different total angular momenta J' in the continuum vanish at somewhat different values of the energy, and as a result the photoabsorption cross section for the transition $S_{1/2} - \epsilon P_{1/2}, \epsilon P_{3/2}$

$$\sigma \sim 2|R_{3/2}|^2 + |R_{1/2}|^2$$

does not vanish anywhere. This effect turned out to be important for the analysis of the polarization of the photoelectrons in alkali-metal vapors.

When account is taken of the difference between the radial matrix elements for different J' in the continuum the nonzero terms in the expressions (9) are those with $k=0$ and 1. Calculation yields in this case

$$\gamma_{nn}(M) = \frac{4\pi^2 e^2}{9\hbar^2} E^2 [(1-2M)R_{n1}^2 + 2(1+M)R_{n3}^2],$$

$$\beta_+(M) = \frac{[R_{n1}R_{g1}(1-2M) + 2R_{n3}R_{g3}(1+M)]^2}{[R_{g1}^2(1-2M) + 2R_{g3}^2(1+M)][R_{n1}^2(1-2M) + 2R_{n3}^2(1+M)]}$$

where R_{i1} and R_{i3} are the radial matrix elements for transitions from the corresponding discrete state into continuum states with $J' = 1/2$ and $J' = 3/2$. We shall henceforth assume for simplicity that they are real.¹⁾

At $M = 1/2$, the limiting value is $\beta_+(1/2) = 1$ in accordance with the fact that transitions are possible into one continuum with $J' = 3/2$. On the other hand if $M = -1/2$, then the limiting value of β_+ at high intensities of the field E amounts to

$$\beta_+(-1/2) = [2R_{n1}R_{g1} + R_{n3}R_{g3}]^2 / [2R_{g1}^2 + R_{g3}^2][2R_{n1}^2 + R_{n3}^2].$$

Thus, allowance for the spin-orbit interaction in the continuum leads to a difference between the limiting value $\beta_+(-1/2)$ and unity, and also to $x(1/2) \neq x(-1/2)$ and to the fact that the parameter q_{ng} becomes a function of M . By an identity transformation we can represent $\beta_+(-1/2)$ in the form

$$\beta_+(-1/2) = (1 + 2\eta_n\eta_g)^2 / (1 + 2\eta_g^2)(1 + 2\eta_n^2), \quad (11)$$

where

$$\eta_n = (R_{i3} - R_{i1}) / 3R_{i0}, \quad R_{i0} = 1/3(2R_{i1} + R_{i3}).$$

It follows from (11) that the value $\beta_+(-1/2) = 1$ is reached

in the region of the continuum far from the minima of the photoabsorption from the level g and the level n ($|\eta_n|, |\eta_g| \ll 1$), and also when the positions of these minima coincide ($\eta_n = \eta_g$).

Assuming that the minima of the photoabsorption from the states n and g do not coincide, and that the frequency ω_0 corresponds to the minimum of the photoabsorption from the state g , i.e., $|\eta_n| \ll 1, |\eta_g| \gg 1$, and $|\eta_n\eta_g| \gg 1$, we find that x does not depend on M . The main contribution to (7) and (8) can be made in this case only by terms proportional to $q_{ng}^2(M)$, i.e.,

$$\theta_{max} = 1/3\sigma_0NL(q^2(1/2) + q^2(-1/2)).$$

Here $q^2(\pm 1/2) \sim \delta_{ng}^2(\pm 1/2) / \sigma_0\sigma_n$; σ_0 and σ_n are the unperturbed total cross sections for ionization from the states g and n :

$$\delta_{ng}(M) \sim \sum_k \frac{R_{nk}R_{kg}}{\omega_0^2 - \omega_k^2} 2\omega_k + \frac{\hbar}{3} \int \frac{R_{g1}(1-2M) + 2R_{g3}(1+M)}{\epsilon_0 - \epsilon} R_n d\epsilon.$$

We estimate now the contribution of the discrete spectrum for the scheme shown in Fig. 2 for the transitions in Cs.

Calculation yields for Cs a value $q^2 \approx 10^3$ ($\sigma_{0(\min)} = 8 \times 10^{-20} \text{ cm}^2$ and $\sigma_{8S} \sim 10^{-19} \text{ cm}^2$).

For a rough estimate of the contribution of the continuum states we used the formula for the difference of the radial matrix elements^{16,20}

$$\Delta R = R_{g3} - R_{g1} = \Delta\tau \int \frac{R_{g0}(\epsilon)}{\epsilon_0 - \epsilon} d\epsilon, \quad (12)$$

where $\Delta\tau$ is the change of the quantum defect and $R_{g0} = 1/3(2R_{g3} + R_{g1})$. If $M = 1/2$, then

$$\int \frac{R_n R_{g3}}{\epsilon_0 - \epsilon} d\epsilon = \int \frac{R_n R_{g0}}{\epsilon_0 - \epsilon} d\epsilon + \frac{1}{3} \int \frac{R_n \Delta R}{\epsilon_0 - \epsilon} d\epsilon,$$

and if $M = -1/2$,

$$\frac{1}{3} \int \frac{2R_n R_{g1} + R_n R_{g3}}{\epsilon_0 - \epsilon} d\epsilon = \int \frac{R_n R_{g0}}{\epsilon_0 - \epsilon} d\epsilon - \frac{1}{3} \int \frac{R_n \Delta R}{\epsilon_0 - \epsilon} d\epsilon.$$

We take R_n outside the integral sign at $\epsilon = \epsilon_0$. Since $\sigma_{n(\max)} / \sigma_{n(\min)} \approx 4$ for Cs,¹⁸ the resultant error does not exceed 2. The integral

$$\frac{1}{3} \int \frac{\Delta R}{\epsilon_0 - \epsilon} d\epsilon$$

is the inverse Hilbert transform of (12) and is therefore approximately equal to $1/3\pi\Delta\tau R_{g0}$. It can be neglected compared with the first integral, inasmuch as for alkali-metal vapors $\pi\Delta\tau \lesssim 0.1$ rad. Recognizing that $\sigma_{g(\min)} \sim (\Delta R)^2$ (Ref. 20) we find that $q^2 \approx (\pi\Delta\tau)^{-2}$. For the atoms K, Rb, and Cs, the values of $\Delta\tau$ are respectively 0.0029, 0.013, and 0.032.¹⁶ Thus, we have $q^2 \sim 10^2$ for Cs, and consequently the main contribution to q is made by summation over the discrete spectrum. For the angle of rotation we obtain $\theta = 10^{-17}NL$. At a pressure ~ 1 Torr ($N \sim 10^{16} \text{ cm}^{-3}$) and $L = 3 \text{ cm}$, we have $\theta \approx 0.3$ rad and the degree of ellipticity is of the same order.

In conclusion, we discuss the spectral manifestations of the polarization resonances. According to (5) and (6),

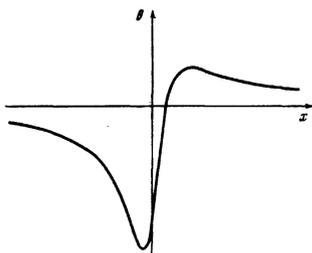


FIG. 3. Qualitative dependence of the polarization-plane rotation angle on the relative detuning.

the absorption and refraction indices as functions of the frequency ω_0 are superpositions of asymmetrical auto-ionization-like resonances. The form of each resonance is shown qualitatively in Fig. 3. The concrete shape of the resulting contour is essentially determined both by their weight $\beta(M, M'')$ and by the ratio of the field shifts and the ionization broadening, i.e., by the quantity $q_{mn} = \delta_{mn}(M'')/\gamma_{mn}(M'')$. This ratio does not depend on the intensity of the radiation and is determined by the energy region of the continuum, with which the interaction of the radiation takes place. Far from the minima of the ionization from the state n , when the relative contribution of the spin-orbit interaction in the continuum is small, the quantity q_{mn} is independent also of M'' . In a strong field E , the individual resonances do not overlap if $|q_{mn}|$ is large. Thus, the form of the spectrum turns out to be sensitive to a number of important characteristics of the continuum.

We note that the quantities q_{bb} do not arise in linear spectroscopy and are peculiar only to nonlinear phenomena. This can explain why there are no theoretical and experimental data on these parameters at present. It appears that nonlinear resonances are the only method of determining these parameters and hence of obtaining information on the integral characteristics of the continuum.

Note added in proof (24 December 1979). When this paper was being readied for press, new experimental observations were reported of nonlinear polarization resonances in continuum in cesium vapor: A. A. Barkhamov, E. E. Kalikin, M. R. Mirzaev, G. Kh. Tartakovskii, S. T. Khudaiberganov, Nonlinear Resonant Conversion of the Frequency of Laser Radiation (Ab-

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¹⁾In fact, the quantities R_{11} and R_{13} are complex.¹⁹ The reason is that the spin-orbit interaction leads to a phase difference between the wave functions of the continuum with different J' . However, allowance for this effect in our case results in a correction $\sim 1\%$ and does not introduce any fundamental changes.

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