

temperatures, the situation is different. The nonlinear damping of the sound increases the role of the volume in the process of light scattering (see Fig. 6); the small duration of the giant laser pulse leads to non-stationarity of the SMBS, which limits the length of interaction to a quantity of the order of 10^{-1} – 10^{-2} cm. Whereas this suffices for the development of SMBS at room temperatures, at low temperatures such a length does not assure the effective transformation of the light.

All that has been said above enables us to suggest the following scheme for the generation of intense hypersound. Two laser beams of equal intensity I_0 and a frequency difference corresponding to the backward SMBS are directed against one another on a crystal of thickness $L \approx \kappa_{n1}/\sqrt{\gamma}$ placed in a liquid helium cryostat. Here the maximum intensity of the hypersound amounts to $c_0 Y I_0 / n c \epsilon$. For example, for quartz and at an intensity $I_0 = 100 \text{ MW/cm}^2$ ($\tau_L \sim 10^{-7}$ sec), we get $L = 5 \times 10^{-2}$ cm, $I_{ac} = 300 \text{ W/cm}^2$ (a similar calculation from linear theory yields an estimate that is larger by a factor of 20). We note that even for crystal lengths of the order of a centimeter, intense hypersound cannot be extracted from the crystal, because of its nonlinear absorption.

Retuning the frequency of one of the laser beams and directing the light beams on the crystal at the corresponding angle, we can change the frequency of the generated hypersound.

The authors thank R. V. Khokhlov for interest in the work, and S. A. Akhmanov, N. S. Bakhvalov, Ya. M. Zhileikin, A. L. Polyakova, V. S. Starunov and I. L. Fabelinskii for useful discussions.

- ¹I. L. Fabelinskii, *Molekulyarnoe rasseyanie sveta (Molecular Scattering of Light)* Nauka, 1965. English Translation, Plenum, 1968.
- ²V. S. Starunov and I. L. Fabelinskii, *Usp. Fiz. Nauk* 98, 441 (1969) [*Sov. Phys. Usp.* 12, 463 (1970)].
- ³A. L. Polyakova, *Pisma Zh. Eksp. Teor. Fiz.* 4, 132 (1966); 7, 76 (1968) [*JETP Lett.* 4, 90 (1966); 7, 57 (1968)].
- ⁴I. L. Fabelinskii, *Izv. Akad. Nauk SSSR Ser. Fiz.* 35, 874 (1971).
- ⁵N. N. Lavrinovich, *Zh. Eksp. Teor. Fiz.* 60, 69 (1971) [*Sov. Phys. JETP* 33, 39 (1971)].
- ⁶S. B. Krivokhizha, *Trudy (Works) of the Phys. Inst., Acad. Sci. USSR* 72, 3 (1974).
- ⁷O. V. Rudenko and S. L. Soluyan, *Teoreticheskie osnovy nelineinoi akustiki (Theoretical Bases of Nonlinear Acoustics)* Nauka, 1975.
- ⁸P. Klemens, in: *Physical Acoustics*, v. IIB (Warren Mason, Ed.) New York.
- ⁹R. G. Brever, *Appl. Phys. Lett.* 6, 165 (1965).
- ¹⁰O. V. Rudenko, *Pisma Zh. Eksp. Teor. Fiz.* 20, 445 (1974) [*JETP Lett.* 20, 203 (1974)].
- ¹¹A. A. Karabutov, E. A. Lapishin and O. V. Rudenko, *Prod. Sixth Internat. Symp on Nonlinear Acoustics*, Moscow State Univ. Press, 1975, p. 28.
- ¹²A. A. Karabutov and O. V. Rudenko, *Zh. Tekh. Fiz.* 45, 1457 (1975) [*Sov. Phys. Tech. Phys.* 20, 920 (1975)].
- ¹³S. A. Akhmanov and R. V. Khokhlov, *Problemy nelineinoi optiki (Problems of Nonlinear Optics)* (VINITI, 1964).
- ¹⁴M. J. O. Strutt, *Lame, Mathieu and Related Functions in Physics and Technology*, (Russ. transl. ONTI, 1935). [German ed. reprinted by Edwards Brothers, 1944].
- ¹⁵N. S. Bakhvalov, *Chislennye metody (Numerical Methods)* Nauka, 1974.
- ¹⁶K. Grob, *Z. Physik* 201, 59 (1965).
- ¹⁷M. J. Brienza and A. S. De Maria, *Appl. Phys. Lett.* 11, 44 (1967).

Translated by R. T. Beyer

Quantized motion of atoms and molecules in electromagnetic fields

A. P. Botin, A. P. Kazantsev, and V. S. Smirnov

L. D. Landau Institute of Theoretical Physics, USSR Academy of Sciences
(Submitted December 30, 1975)
Zh. Eksp. Teor. Fiz. 71, 122–134 (July 1976)

Atoms and molecules may exist in coupled states in the strong field of a standing electromagnetic wave. The absorption spectrum peaks (against the background of the Doppler contour) acquire a fine structure when the distance between levels begins to exceed the line width. This occurs for atoms in fields $\sim 10 \text{ W/cm}^2$ and for molecules in fields $\sim 0.1 \text{ W/cm}^2$. The peak width is investigated as a function of the frequency detuning of the strong field for broad and narrow molecular resonances. Discontinuities arise in the atomic spectra when the condition $\epsilon(\hbar k) > \hbar \gamma$ is satisfied and they may produce dips of the absorption coefficient. The case of a strong field ($\geq 1 \text{ kw/cm}^2$) is considered when the general shape of the absorption contour changes, viz., the Doppler contour is replaced by a band whose width is proportional to the field amplitude.

PACS numbers: 31.30.Gs

1. INTRODUCTION

A strong inhomogeneous electromagnetic field acts on atoms and molecules in two ways: the Stark shift alters the energy levels and the particle velocities. The ef-

fective modulation of the levels (the dynamic Stark effect) has been investigated in detail, principally in connection with the theory of gas lasers.^[1–3] In this theory it is very important that the atoms move with constant unperturbed velocities. We consider in this paper

a situation wherein both modulations—of the levels and of the velocities—are substantial. A strong correlation is then produced between the motion of the atom, its response to an external field, and the absorption (emission) spectrum. This modulation does not reduce to allowance for the Doppler effect. Not knowing the trajectories of the atom, it is in general impossible to determine its response to the external field, and vice versa. In other words, the mechanical and optical phenomena become intermixed in a strong resonant inhomogeneous field.

In weak fields, as is well known, the change of motion of the atom in optical transitions reduces to a recoil effect, which can become noticeable only for narrow resonances

$$\varepsilon(\hbar k) > \hbar\gamma, \quad (1)$$

where $\varepsilon(\hbar k) = (\hbar k)^2/2M$, $\hbar k$ is the photon momentum, M is the atom mass, and γ is the resonance width. In optics, condition (1) is satisfied at $\gamma < 10^5$ Hz. The singularities of the recoil effect in stimulated transitions in a weak field were investigated by perturbation theory in^[4,5] in connection with the theory of the Lamb dip.

In the field of a strong standing light wave, the criterion for the need of taking the recoil effect into account in induced transitions is of the form^[6,1]:

$$kdE/p > \gamma, \quad (2)$$

where d is the dipole moment of the transition, E is the amplitude of the field, and p is the momentum of the atom relative to the standing wave. The left-hand side of this inequality is the Doppler shift of the frequency due to the velocity modulation in the inhomogeneous field. Some singularities of the recoil effect in a strong field were discussed earlier.^[7,8]

The criterion (2) depends significantly on the atom velocity. For atoms that are trapped in the potential wells of a standing wave and for which $\varepsilon(p) \sim dE$, we have in place of (2) the condition

$$\Omega = k(dE/M)^{1/2} > \gamma, \quad (3)$$

where Ω is the characteristic frequency of the oscillations of the atoms in the potential well. The distance between the energy levels is then larger than $\hbar\gamma$. The critical field intensity $I_0 = c\gamma^4 E^2/4\pi d^2 k^4$, which corresponds to the condition $\Omega = \gamma$, depends strongly on the resonance width. For atoms with $\gamma \sim 10^7$ Hz we have $I_0 \sim 10$ W/cm². For molecules with $\gamma \sim 10^5$ Hz we have $I_0 \sim 0.1$ W/cm².

At $I > I_0$ a fine structure due to the bound states appears against the background of the Doppler absorption contour, near the center of the line. The number of bound states of the atoms at the threshold is $dE/\hbar\Omega \sim 30$. Under the conditions of the inequality (3), the effects due to quantization of the atom motion must be taken into account.

We note that the influence of the trapped atoms on the

spectral characteristics under the conditions $\varepsilon(\hbar k) \ll \hbar U \ll \hbar\gamma$ (U is the depth of the nonresonant potential of the atom) was considered classically by Letokhov.^[14] It was possible in this case to disregard the field-induced shift of the levels.

In addition to condition (2), there is also the criterion $kdE/p > \varepsilon(\hbar k)/\hbar$ or

$$dE/\hbar > kp/M, \quad (4)$$

which means that the motion of the atoms and the change of the atomic states takes place in quasiclassical fashion. If the condition (4) is satisfied up to the thermal momenta p_T of the atoms (the critical intensity corresponding to the condition $dE/\hbar = kp_T/M$ will be designated I_1), then the absorption contour differs substantially from the Doppler contour. For alkali-metal atoms we have $I_1 \sim 1$ kW/cm². Thus, the change of the atom motion in an inhomogeneous field manifests itself either in the form of a certain fine structure in the absorption coefficient, due to the bound states at $I > I_0$, or in a change of the general form of the absorption coefficient due to the change of the continuous states at $I > I_1$.

We have calculated the weak-signal absorption coefficient in the presence of a strong standing wave acting on an adjacent transition. To find the absorption line contour it is necessary to know the spectrum and the wave functions of the atoms in the discrete and continuous states. This problem can be solved with the aid of the system of wave equations for a many-level atom.^[9]

2. FUNDAMENTAL EQUATIONS

We considered the line shape for the absorption of a weak field $V_0(x, t)$ by a three-level system with states 0, 1, and 2. The frequency of the signal V_0 is close to the frequency ω_{20} of the 0–2 transition. The adjacent 1–2 transition is acted upon by a strong field $V(x, t)$ with frequency close to ω_{21} . All the quantities pertaining to the weak field and to the transition 0–2 are labeled with the index zero, while the quantities for the strong field and the 1–2 transition are written without an index:

$$\begin{aligned} V(x) \exp[-i(\omega_{21} + 2\Delta)t], \quad V(x) = V \cos kx, \quad V = dE/\hbar, \\ V_0 \exp[-i(\omega_{20} + \Delta_0)t + ik_0 x], \quad V_0 = d_0 E_0/\hbar \ll V. \end{aligned}$$

The three-level system was investigated theoretically and experimentally in many studies (see, e.g.,^[10–12]). These, however, dealt only with traveling waves, in which the recoil effect is merely the same as in spontaneous transitions. We start from a system of Schrödinger equations having in the resonant approximation the form

$$\begin{aligned} i \left(\frac{\partial}{\partial t} + \gamma \right) \begin{pmatrix} \psi_1 \\ \psi_2 \end{pmatrix} = \begin{pmatrix} H_0/\hbar + \Delta, & V(x) \\ V(x), & H_0/\hbar - \Delta \end{pmatrix} \begin{pmatrix} \psi_1 \\ \psi_2 \end{pmatrix} + \nabla_0 \psi_0 \begin{pmatrix} 0 \\ 1 \end{pmatrix}, \quad (5) \\ H_0 = -\frac{1}{2M} \left(\hbar \frac{d}{dx} \right)^2, \quad \nabla_0 = V_0 e^{i\Delta_0 t}. \end{aligned}$$

For simplicity, the relaxation constants of the excited levels 1 and 2 are assumed to be equal, and the lower level is assumed to be the ground state. At the initial

instant of time, only the ground state is populated with an atom density n_0 , and its change due to the weak field will be neglected.

The wave function of the ground state is of the form

$$\psi_0(x, t) = \frac{1}{\sqrt{L}} \sum_p \psi_p \exp\{ipx/\hbar - i\varepsilon(p)t/\hbar\}, \quad (6)$$

$$\langle \psi_p, \psi_{p'} \rangle = \delta_{pp'} 2\pi\hbar n_0 F(p), \quad F(p) = \pi^{-1/2} p_r^{-1} \exp\{- (p/p_r)^2\},$$

where the angle brackets denote averaging over the thermal motion of the atoms. The weak-field absorption coefficient $q_0(\Delta_0)$ is determined by the relation

$$q = \frac{2\pi\hbar k_0}{L|E_0|^2} \left\langle \int_0^L dx \mathcal{V}_0^*(x, t) \psi_0^*(x, t) \psi_2(x, t) \right\rangle. \quad (7)$$

It is assumed that the averaging region L is large in comparison with the wavelengths $\lambda = 2\pi/k$ and $\lambda_0 = 2\pi/k_0$, but small in comparison with q_0^{-1} .

The effects due to quantization of the atom motion, which will be considered below, manifest themselves only in sufficiently strong fields

$$V \gg \gamma. \quad (8)$$

As to the ratio of the thermal energy of the atoms to the depth of the potential wells, we have $\hbar V \lesssim T$ up to fields close to the critical ionization field. We therefore put

$$\hbar V \ll T. \quad (9)$$

In this range of fields, the most characteristic are two limiting cases, the quasiclassical case $\hbar V \gg \varepsilon(\hbar k)$, when there are many bound states, and the quantum case $\hbar V \ll \varepsilon(\hbar k)$, when there are no bound states. It is convenient to consider these cases separately.

3. THE QUASICLASSICAL LIMIT

We shall deal henceforth (except in Sec. 8) mainly with the quasiclassical limit, when the condition (4) is satisfied with a large margin for all the atoms:

$$V \gg \Delta_T = k p_T / M, \quad (10)$$

where Δ_T is the Doppler width. In this case the condition $\hbar V \gg \varepsilon(\hbar k)$ is all the more satisfied, since $\hbar k \ll p_T$. The wave functions of the atoms can then be obtained in the quasiclassical approximation.

The behavior of the free and bound atoms turns out to be essentially different for small and large detunings of the strong field. At small Δ , it is more natural to represent Eq. (5) in the form

$$i \left(\frac{\partial}{\partial t} + \gamma \right) \begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix} = \begin{pmatrix} H_0/\hbar - V(x), & 0 \\ 0 & H_0/\hbar + V(x) \end{pmatrix} \begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix} \\ - \Delta \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix} + \frac{V_0 \psi_0}{\sqrt{2}} \begin{pmatrix} 1 \\ 1 \end{pmatrix}, \quad \varphi_{1,2} = (\psi_1 \pm \psi_2) / \sqrt{2}, \quad (11)$$

so that the off-diagonal term is a small perturbation.

4. RESONANT CASE

At $\Delta = 0$, the homogeneous equation (11) describes two groups of independent particles in potentials $\pm \hbar V(x)$, and their wave functions are ($\alpha = 1, 2$)

$$\varphi_\alpha = \sum_{m,n} A_{\alpha n}(m, t) \varphi_n(x - m\lambda - x_\alpha) + \sum_{\varepsilon} A_{\alpha \varepsilon}(t) \varphi_\varepsilon(x - x_\alpha),$$

where the functions $\varphi_n(x)$ and $\varphi_\varepsilon(x)$ of the discrete and continuous spectra satisfy the equation

$$(H_0 + \hbar V(x)) \varphi_n = \hbar \omega_n \varphi_n, \quad (H_0 + \hbar V(x)) \varphi_\varepsilon = \varepsilon \varphi_\varepsilon,$$

With $-V < \omega_n < V$ and $\varepsilon > \hbar V$. The eigenfunctions with equal energies and different indices α differ in a shift of the argument by $\lambda/2$. We represent the absorption coefficient in the form of a sum of contribution from discrete and continuous states: $q_0 = q^{\text{cont}} + q^{\text{dis}}$,

$$q^{\text{cont}} = \frac{Q_0 M}{\pi^2 \hbar L} \int_{-\infty}^{+\infty} dp F(p) \sum_{\varepsilon > V} |\varphi_\varepsilon(p_+)|^2 \text{Re}[\gamma + i(\varepsilon - \varepsilon(p) - \hbar \Delta_0)/\hbar]^{-1}, \quad (12)$$

$$q^{\text{dis}} = \sum_n q_n, \quad q_n = \frac{Q_0 k}{\pi p_r} \int_{-\infty}^{+\infty} dp F(p) |\varphi_n(p_+)|^2 \text{Re}[\gamma + i(\omega_n - \Delta_0 - \varepsilon(p)/\hbar)]^{-1}, \quad (13)$$

where $\varphi_{\varepsilon, n}(p)$ is the Fourier transform of the wave function $\varphi_{\varepsilon, n}(x)$, and $p_+ = p + \hbar k_0$.

The integral absorption coefficient

$$Q_0 = \int d\Delta_0 q_0(\Delta_0) = 2\pi^2 d_0^2 n_0 k_0 / \hbar \quad (14)$$

has, by the virtue of the completeness of the system of eigenfunctions $\varphi_\varepsilon(x)$ and $\varphi_n(x)$, the same form as in the absence of an external field (sum rule). Inside the absorption band $-V < \Delta_0 < V$, the resonant denominator can be replaced by a δ -function (energy conservation law). The momentum conservation law is obtained by calculating the Fourier transforms of the eigenfunctions by the saddle-point method.

Contribution from continuous spectrum

Using the conservation laws, we obtain the following absorption coefficient for frequencies that are not close to the absorption band:

$$q^{\text{cont}} = \frac{Q_0}{\pi} (V^2 - \Delta_0^2)^{-1/2}, \quad |\Delta_0| < V. \quad (15)$$

The integral absorption coefficient (15) coincides with Q_0 . This circumstance is a consequence of the inequality (9): the fraction of the atoms in bound states is a small quantity of order $(\hbar V/T)^{1/2}$.

The atoms observed a weak signal at those points of space x_0 at which the term of the excited atom intersects the detuning, $V(x_0) = \Delta_0$. The absorption coefficient is proportional to the density of states of the atoms, which is inversely proportional to the slope of the term $dV(x_0)/dx$ at the intersection point. This leads to formula (15). As Δ_0 approaches the boundary of the absorption band, the density of states increases and q_0 increases. At $|\Delta_0| = V$ we have not a crossing but a second-order term tangency at the points where $\cos kx = \pm 1$.

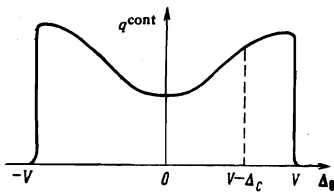


FIG. 1.

Taking this circumstance into account, we have at the boundary of the absorption band the following formula:

$$q_0 = aQ_0 V^{-1/3} \Delta_T^{-2/3}, \quad |\Delta_0| = V, \quad (16)$$

where $a = 3^{-1/3} \cdot 2^{-2/3} \Gamma^2(\frac{1}{3}) \pi^{-5/2} \approx 0.5$ and $\Gamma(x)$ is the Euler Gamma function.

At the boundary, the absorption coefficient increases in comparison with the center of the band by an approximate factor $(V/\Delta_T)^{1/3} \gg 1$. The transition from (15) and (16) takes place in a relatively narrow frequency interval $\Delta_c \sim V^{1/3} \Delta_T^{2/3} \ll V$. At $|\Delta_0| > V$, the absorption coefficient becomes small by virtue of the conditions (8) and (10). The boundary of the absorption n is smeared out by an amount of the order of $(\Delta_T^0)^2/V$. The dependence of q^{cont} on Δ_0 is shown schematically in Fig. 1. We note for comparison that in a homogeneous field (traveling-wave field) under the conditions of the inequality (10) the absorption contour constitutes narrow peaks at the peaks $\Delta_0 = \pm V$. Thus, in the field of a strong resonant standing wave the absorption line spreads to form a band whose width is proportional to the amplitude of the external field.

Contribution from discrete spectrum

For the absorption coefficient q^{dis} inside the absorption band we have the expression

$$q^{\text{dis}} = Q_0 [\hbar/\pi T (V + \Delta_0)]^{1/2}. \quad (17)$$

In this case there is no singularity at $\Delta_0 = V$, since the number of the bound states contributing to the absorption vanishes here. At $\Delta_0 = -V$ expression (17) goes over into

$$q^{\text{dis}} = a' Q_0 V^{-1/3} T^{-1/3} \hbar^{-1/2} e^{-1/2} (\hbar k), \quad (18)$$

where $a' = 3^{2/3} \cdot 2^{-7/3} \Gamma^2(\frac{1}{3}) \pi^{-5/2} \approx 0.2$. The frequency region of the transition from (17) to (18) is of the order of $\Delta_c' = V^{2/3} (\epsilon(\hbar k)/\hbar)^{1/3} \ll V$.

Formula (17) yields inside only a certain averaged

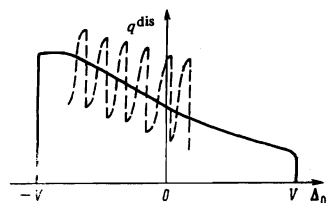


FIG. 2.

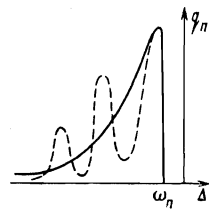


FIG. 3.

dependence of the absorption on the frequency; in Fig. 2 it is represented by the solid line. The exact behavior of $q^{\text{dis}}(\Delta_0)$ has a more complicated structure, owing to the resonances (they are shown dashed in Fig. 2) at $\Delta_0 = \omega_n$, where $\hbar\omega_n$ are the energies of the bound states. The authors contribution to the absorption from the discrete state is of the order of $(\hbar V/T)^{1/2} \ll 1$, so that $q_0 \approx q^{\text{cont}}$.

5. STRUCTURE OF BOUND STATES

Inasmuch as in a strong field at $\Omega \gg \gamma$ the peaks due to the bound states are far enough from one another, it is of interest to consider their structure in greater detail. At frequencies satisfying the condition $\Omega \ll V + \Delta_0 \ll V$ we can use the quasiclassical oscillator approximation and attain for the contribution made to the absorption by the n -th bound state the expression

$$q_n = \frac{\hbar k Q_0 \sin^2 \alpha_n}{\pi^{1/2} p_T [(V + \Delta_0) (\omega_n - \Delta_0)]^{1/2}}, \quad (19)$$

where

$$\alpha_n = (n+1)\pi/2 + k_0 [2\hbar(n+1)/M\Omega]^{1/2} + 2[(n+1/2)(\omega_n - \Delta_0)/\Omega]^{1/2}$$

is the quasiclassical phase.

The absorption intensity q_n is subject to oscillations with a frequency that increases with the level number n . However, at $n > \Omega/\gamma$ the oscillations vanish because of the averaging, in the exact formula (13), over a frequency interval on the order of γ . Therefore at $n > \Omega/\gamma$ it is necessary to put $\sin^2 \alpha_n \approx \frac{1}{2}$ in formula (19). The structure of an individual peak is shown schematically in Fig. 3. Near the resonance the peak no longer has a square-root form, and at $\Delta_0 = \omega_n$ the factor $\gamma^{1/2}$ appears in the denominator (19). The case of oscillations at $n < \Omega/\gamma$ is shown schematically by the dashed curve. The characteristic root structure of the periods is due to the fact that the weak signal V_0 mixes the discrete states of the excited atoms with the continuous states of the unexcited atoms. If the weak signal were to act on the same transition as the strong field, then transitions would be produced also between discrete-discrete states and the shape of the peak would be Lorentzian. The height of the peak would increase by an approximate factor $(\Omega/\gamma)^{1/2}$.

We estimate now the relative height of the peak a , defined by the relation

$$a = [q_0(\omega_n) - q_0(\omega_n - \Omega)] / q_0(\omega_n) \approx \left[q_n(\omega_n) + \frac{dq^{\text{cont}}(\omega_n)}{d\Delta_0} \Omega + \frac{1}{2} \frac{d^2 q^{\text{cont}}(\omega_n)}{d\Delta_0^2} \Omega^2 \right] / q^{\text{cont}}(\omega_n). \quad (20)$$

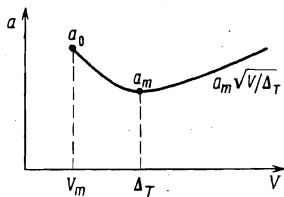


FIG. 4.

In the calculation of a , the smooth part of the discrete spectrum can be disregarded at $\Omega \gg \gamma$. At the center of the band, where the first derivative of $q^{\text{cont}}(\Delta_0)$ vanishes, we have

$$q_n(\omega_n)/\Omega^2 \frac{d^2 q^{\text{cont}}}{d\Delta_0^2} \gg (\Delta_T/\gamma)^{1/2} \gg 1.$$

Thus, for resonances located near the center of the band (their approximate number is $(\Delta_T/\gamma)^{1/2}$), the curvature of the absorption band can be neglected. The exact formulas for a are quite cumbersome, and we confine ourselves therefore to very rough estimates. The qualitative dependence of a on the field amplitude is shown in Fig. 4, $a_m \sim 10^{-2} A^{-3/4}$ and $a_0 \sim A^{-3/2}$, where A is the atomic weight. The minimum field amplitude V_m corresponds to the condition $\Omega = \gamma$. This dependence is suitable approximately for atoms (we used for estimates the parameter values $k \sim k_0 \sim 10^5 \text{ cm}^{-1}$ and $\gamma \sim 10^7 \text{ Hz}$) and for molecules ($k \sim 2 \times 10^4 \text{ cm}^{-2}$, $\gamma \sim 10^5 \text{ Hz}$). For $A \sim 10$, the relative height of the peak fluctuates from tenths of a percent to several percent.

6. DYNAMIC BROADENING OF RESONANCES

We investigate now the structure of the bound states at small detunings of the strong field. The case $\Delta = 0$ of the resonance of the strong field is special, for only in this case can the Schrödinger equations be diagonalized exactly. The quasiparticles moving in potentials $\pm \hbar V(x)$ do not interact with one another in this case and make equal contributions to the absorption. At finite detunings, the off-diagonal term in (11) leads to a mixing of the quasiparticles and to a change of the resonance widths.

Indeed, as seen from Fig. 5, the wave functions of the particles of different sorts can spatially overlap. One potential in the figure is shown by the solid line and the other dashed. Under the influence of the perturbing potential proportional to Δ , a particle of one sort with energy $\hbar\omega_n$ can go over virtually into a state of particles of another sort with energy $\hbar\omega_{\bar{n}}$ and "drop" to a level with energy $\hbar\omega_n$ in the neighboring well. A transition from one well to another is produced in this manner. The probability of the transition is in this case not an exponentially small quantity relative to the quasi-classical parameter.

The amplitudes of the states whose energy of the negative satisfy the following system of equations:

$$i \left(\frac{\partial}{\partial t} + \gamma \right) A_{an}(m) = \omega_n' A_{an}(m) + \frac{1}{2} \Gamma_n (A_{an}(m-1) + A_{an}(m+1)) + \int dx \varphi_{an}(x - m\lambda - x_a) \hat{V}_0(x, t) \psi_0(x, t). \quad (21)$$

ω_n' denotes the resonant frequency with allowance for the shift due to the virtual transitions to discrete and continuous states. In order of magnitude we have $\omega_n' - \omega_n \sim \Delta^2/V$. The frequency of the hopping to the neighboring levels is given by

$$\Gamma_n = \frac{\Delta^2 N}{\omega_n - \omega_{\bar{n}}} \left[\int dx \varphi_n(x - x_a) \varphi_{\bar{n}}(x - x_{\bar{a}}) \right]^2. \quad (22)$$

To estimate the overlap integrals we can use the Franck-Condon principle for electronic transitions in molecules. These integrals have the largest value when the turning points n and \bar{n} of the states coincide in the vicinity of $\Delta x = \lambda [\epsilon(\hbar k)/\hbar V]^{1/3} \sim \hbar^{2/3}$. The distance between the turning points of neighboring levels is of the order of \hbar , so that \bar{n} levels fall in the vicinity of Δx in the quasi-classical limit $N \gg 1$. As a result we have the following estimate for the hopping frequency: $\Gamma_n \sim \Delta^2(\omega_n - \omega_{\bar{n}})^{-1}$.

Solving (21), we obtain for q_n the expression

$$q_n(\Delta_0) = \frac{Q_0 k}{\pi^{1/2} p_T} \int_{-\infty}^{+\infty} dp |\varphi_n(p_+)|^2 \text{Re} [\gamma + i(\omega_n' + \Gamma_n \cos(p_+ \lambda/\hbar) - \Delta_0 - \epsilon(p)/\hbar)]^{-1}. \quad (23)$$

The level smears out into a band of width $2\Gamma_n$, while the period in momentum space is equal to $\hbar k$.

In the case of positive energies, different states with identical energy become mixed in first order in Δ , as shown in Fig. 5 for states with energy $\hbar\omega_e$. The formula for q_n then becomes

$$q_n(\Delta_0) = \frac{k Q_0}{\pi^{1/2} p_T} \int_{-\infty}^{+\infty} dp |\varphi_n(p_+)|^2 \left\{ \cos^2 \left(\frac{p_+ \lambda}{4\hbar} \right) \times \text{Re} [\gamma + i(\omega_n' + \Gamma_n \cos(p_+ \lambda/\hbar) - \Delta_0 - \epsilon(p)/\hbar)]^{-1} + \sin^2 \left(\frac{p_+ \lambda}{4\hbar} \right) \text{Re} [\gamma + i(\omega_n' - \Gamma_n \cos(p_+ \lambda/\hbar) - \Delta_0 - \epsilon(p)/\hbar)]^{-1} \right\}. \quad (24)$$

This expression differs from (23) in that the level now smears out into two bands that interfere with each other. This gives rise to modulation factors $\cos^2(p_+ \lambda/4\hbar)$ and $\sin^2(p_+ \lambda/4\hbar)$ for positive energies we have $\Gamma_n \sim \Delta [\epsilon(\hbar k)/\hbar V]^{1/4}$. Further analysis of the structure of the resonances depends essentially on the relation between the width $\hbar\gamma$ of the resonance and the recoil energy $\epsilon(\hbar k)$. We consider two characteristic limiting cases.

Broad resonances

Atomic dipole-allowed transitions usually satisfy the condition

$$\epsilon(\hbar k) \ll \hbar\gamma. \quad (25)$$

When Γ_n is large enough ($\gamma \ll \Gamma_n \ll \Omega$), the main contribu-

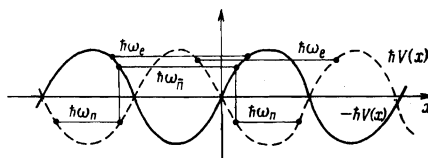


FIG. 5.

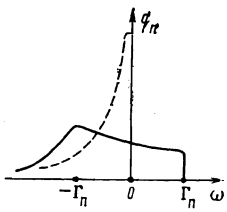


FIG. 6.

tion to the integrals (23) and (24) is made by a rather large momentum region in which $\cos(p_+ \lambda / \hbar)$ is a rapidly oscillating function. Averaging over these oscillations we obtain

$$q_n(\omega) = \frac{kQ_0 |\varphi_n(p=0)|^2}{2\pi^{3/2} p_T} \int dp [\Gamma_n^2 - (\varepsilon(p)/\hbar + \omega)^2]^{-1/2}, \quad (26)$$

where $\omega = \Delta_0 - \omega'_n$. The integration is carried out over a momentum region in which the radicand is positive. We have assumed for simplicity $n > \Omega/\gamma$, so that the oscillations of the matrix element of the transition can be disregarded. The function $q_n(\omega)$ is illustrated in Fig. 6 by the solid line, while the dashed line shows the same dependence at $\Gamma_n = 0$. The effective resonance width turns out to be of the order of the tunneling frequency Γ_n . The vertex of the peak flattens out in this case in such a way that the integrated intensity remains unchanged. When the condition (25) is satisfied, the shape of the peaks for the positive and negative energies turns out to be the same, differing only in the resonance width Γ_n .

Narrow resonances

For certain narrow molecular resonances and weakly-resolved atomic transitions, the condition (1) may be satisfied.¹⁾ It is of interest to consider the shape of the peaks in this case. To simplify the calculations we assume

$$\varepsilon(\hbar k) \gg \hbar\gamma. \quad (27)$$

The contribution to the absorption is determined by the small vicinity of momenta in which the energy conservation law is satisfied

$$\mathcal{E}(p) = \varepsilon(p) + \hbar\omega + \hbar\Gamma_n \cos(p_+ \lambda / \hbar) = 0. \quad (28)$$

Let us examine the character of the absorption singularities that arise when Γ_n is varied. The singularities manifest themselves in the case when the function $d\mathcal{E}/dp$ vanishes at the point where $\mathcal{E}(p) = 0$. At small values of Γ_n there is only one value of the frequency ω' at which the equation $d\mathcal{E}/dp = 0$ has a solution and which corresponds to the right-hand boundary of the absorption peak.

At $\Gamma_n = \Gamma_c$, a second frequency ω'' appears, at which the equation $d\mathcal{E}/dp = 0$ also can have a solution. The exact value of Γ_c depends on the ratio k/k_0 , and in order of magnitude we have $\Gamma_c \sim \varepsilon(\hbar k)/\hbar$. As seen from a graphic analysis of the equation $d\mathcal{E}/dp = 0$, the second

root appears in the case when the linear function is tangent to a sinusoid. This means that $d^2\mathcal{E}/dp^2 = 0$ at this point and the singularity becomes stronger than a root singularity.

Thus, at $\Gamma_n = \Gamma_c$ there appears (see Fig. 7a) an additional symmetrical peak

$$q_n(\omega) = \frac{kQ_0 (2\hbar/9)^{1/2} |\varphi_n(p=0)|^2}{\pi^{3/2} p_T (d^2\mathcal{E}/dp^2)^{1/2} (\omega - \omega'')^{1/2}}. \quad (29)$$

At the maximum, the symmetrical peak exceeds the square-root peak, $q_n(\omega'')/q_n(\omega') \sim (\varepsilon(\hbar k)/\hbar\gamma)^{1/6}$. At $\Gamma_n > \Gamma_c$, the symmetrical peak splits into two asymmetrical root-type peaks, as shown in Fig. 7b. As a result of this splitting, the band of width $\omega' - \omega'' \sim \varepsilon(\hbar k)/\hbar$ becomes detached from the peak. Inside the band, near its boundaries, we have a square-root behavior $q_n \sim (\omega' - \omega)^{-1/2}$ as $\omega \rightarrow \omega'$ and $q_n \sim (\omega - \omega'')^{-1/2}$ as $\omega \rightarrow \omega''$. This behavior of the absorption coefficient is due to the fact that the quasiparticles have a positive effective mass near the bottom of the band and a negative mass near the top of the band.

With increasing Γ_n , the process of detachment of bands of width $\varepsilon(\hbar k)/\hbar$ from the initial peak repeats itself. The heights of the individual root-type peaks decrease like $(M_{\text{eff}}/M)^{1/2}$, where $M_{\text{eff}} \sim M(1 + \hbar\Gamma_n/\varepsilon(\hbar k))^{-1}$ is quasiparticle effective mass. Thus, the broadening of the resonance with increasing Γ_n is via "production" of an ever larger number of bands with decreasing amplitudes. The broadening of the resonances with positive energy takes place qualitatively in the same manner. The difference lies in the fact that the peak splits into two peaks and two systems of bands, the intensities of which are modulated by the factors $\cos^2(p_+ \lambda / 4\hbar)$ and $\sin^2(p_+ \lambda / 4\hbar)$.

So long as the perturbation-theory criterion $\Gamma_n \ll \Omega$ is satisfied, the summary area of the band is equal to the modulation of the peak at $\Gamma_n = 0$. If we disregard the not too significant quasiclassical factor, the criterion of the resonant approximation is

$$\Delta \ll \Omega. \quad (30)$$

7. NONRESONANT CASE

At a large detuning

$$\Delta \gg \Omega \quad (31)$$

the Hamiltonian of the strong field can be diagonalized only approximately. In the quasiclassical approxima-

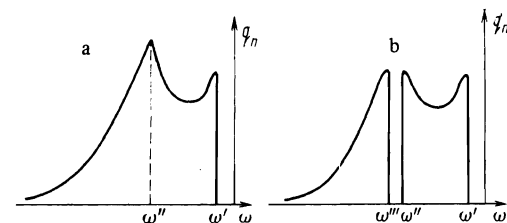


FIG. 7.

tion we have the following system of equations for the upper working levels:

$$i\left(\frac{\partial}{\partial t} + \gamma\right)\begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix} = \begin{pmatrix} H_0/\hbar + U(x), & 0 \\ 0, & H_0/\hbar - U(x) \end{pmatrix}\begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix} + \frac{\hbar}{M}(1+\xi^2)^{-1} \frac{d\xi}{dx} \begin{pmatrix} 0, & -1 \\ 1, & 0 \end{pmatrix} \frac{d}{dx} \begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix} + (1+\xi^2)^{-1/2} V_0 \psi_0 \begin{pmatrix} \xi \\ 1 \end{pmatrix},$$

$$U(x) = V\Delta^2 + V^2(x), \quad \xi(x) = (U(x) - \Delta)/V. \quad (32)$$

The off-diagonal component, which is proportional to $d\xi/dx$, is a small quantity; its order of magnitude relative to $U(x)$ is $kp/MV \ll 1$ in accordance with condition (10). In principle order we have two groups of particles moving in potentials $\pm \hbar U(x)$. We determine from them the contribution made to the absorption, neglecting the off-diagonal component in (32):

$$q^{\text{cont}}(\Delta_0) = \frac{Q_0}{\pi} \left[\frac{\Delta_0 - 2\Delta}{\Delta_0(V^2 + 2\Delta_0\Delta - 4\Delta^2)} \right]^{1/2},$$

$$\Delta - U < \Delta_0 < 0, \quad 2\Delta < \Delta_0 < \Delta + U. \quad (33)$$

Formula (33) is defined in two regions; the absorption band splits into the two bands shown in Fig. 8. We note that the absorption coefficient has a regular normalization. Near the absorption-band boundaries themselves, formula (33) is violated and instead of the square-root singularity we have a finite value, just as in the resonant case. In addition, no account in (33) of the Doppler smearing of the boundaries by an amount on the order of $(\Delta_0^2)^2/V$. With increasing Δ , the area of the right-hand band decreases like $(V/\Delta)^2$. At $\Delta=0$, formula (33) goes over into formula (15). Bound states appear in the potentials $\pm \hbar U(x)$ and make a contribution to the fine structure of the absorption coefficient. Small off-diagonal terms in (32) lead to a mixing of the independent states. The bound states in the potential $+\hbar U(x)$ are against the background of the continuous spectrum of particles that move in a potential $-\hbar U(x)$. However, the decay of these bound states is impossible in the quasiclassical limit, since the momentum conservation law is not satisfied here. All that remains is hopping to the same energy level in a neighboring well. The picture of the splitting of the peaks is of approximately the same form as in the resonant case for bound states with negative energies. The hopping frequency is of the order of

$$\Gamma_n \sim \Delta^{-1} \xi^2 (\epsilon(\hbar k)/\hbar)^{1/2} V^{2/3}. \quad (34)$$

At large detunings, the hopping frequency decreases like Δ^{-3} .

Γ_n increases with decreasing Δ , and at $\Gamma_n \sim \Omega$ per-

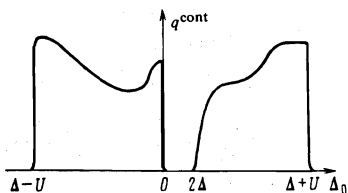


FIG. 8.

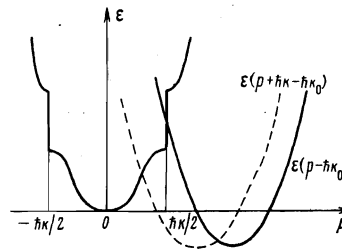


FIG. 9.

turbation theory no longer holds for the bound states. This takes place (leaving out the quasiclassical factor $(\epsilon(\hbar k)/\hbar V)^{1/3}$) at $\Delta \sim \Omega$.

We can thus state that the bound atoms move in potentials $\pm \hbar V(x)$ at $\Delta < \Omega$ and in potentials $\pm \hbar U(x)$ at $\Delta > \Omega$.

8. QUANTUM LIMIT

So far we have considered the quasiclassical case $\epsilon(\hbar k) \ll \hbar V$. Let us discuss the singularities of the absorption coefficients in the quantum limit

$$\epsilon(\hbar k) \gg \hbar V. \quad (35)$$

As before, we assume here that the condition (8) is satisfied. Both conditions (35) and (8) can be satisfied only for very narrow resonances $\gamma \sim 10^5 - 10^3$ Hz. For simplicity we assume the strong field to be resonant ($\Delta=0$). In this case, as is well known, the atom-excitation spectrum has discontinuities at $p = \pm s\hbar k/2$, where s is an integer. Figure 9 shows the discontinuities at $s=1$. When the detuning satisfies the condition

$$\hbar\Delta_0 = \epsilon(\hbar k/2) - \epsilon(\hbar k/2 - \hbar k_0), \quad (36)$$

the parabola corresponding to the spectrum of the atoms in the ground state falls in the forbidden band and the absorption decreases to a relative value either γ/V or $\hbar V/\epsilon(\hbar k)$. The last estimate follows from the fact that the wave function of the excited atoms contain higher harmonics, the momenta of which are shifted by an amount $m\hbar k$, while the amplitude decrease like $(\hbar V/\epsilon(\hbar k))^m$. Scattering by these waves can take place with energy conservation. The case $m=1$ is shown dashed in Fig. 9. Thus, in the quantum limit (35), a number of dips appear in the absorption coefficient. The dip with the largest area corresponds to the first forbidden band. The center of the dip does not depend on the field, and the width of the dip is of the order of V , while the relative amplitude at the minimum is of the order of γ/V or $\hbar V/\epsilon(\hbar k)$.²⁾ The relative depth of the dip can therefore be quite large. The width of the dip increases with increasing field, and the depth decreases. At $\hbar V \geq \epsilon(\hbar k)$ the dip vanishes.

9. CONCLUSION

Our analysis shows that the influence of the mechanical phenomena on the optical phenomena can be very appreciable. There are two characteristic field intensities at which it is necessary to take into account the action of the light pressure on the atoms and molecules.

At $I > I_0$ bound states appear in the standing-wave field and contribute to the fine structure of the absorption coefficient. At $I > I_1$ the general form of the absorption coefficient changes: the Doppler contour gives rise to an absorption band with a width proportional to the field amplitude.

The structure of the absorption peaks due to the bound states depends strongly on the standing-wave field parameters and on the width of the resonance. In the case of atomic resonances the peaks have a relatively simple form and their width is either equal to the resonance width or is of the order of the frequency detuning of the strong field (at small detunings). The narrow molecular resonances can form peaks of more complicated form. In particular, "quantization" of the peaks sets in: when the strong-field frequency is changed by an amount $\epsilon(\hbar k)/\hbar$, a narrow band is detached from the peak.

When the condition (1) is satisfied, discontinuities can appear in the spectrum of the atoms and can lead to formation of dips in the absorption coefficient.

The appearance of narrow peaks or dips in the absorption coefficient is of interest for frequency standards. We note that although the position of the peak depends on the field intensity, the effect of splitting of one peak into two is determined only by the field detuning. The position of the dip does not depend on the field.

The authors thank S. G. Rautian and A. M. Shalagin for a useful discussion.

¹Hall^[13] has reported observation (by an indirect method) of the recoil effect, as deduced from the shape of the Lamb dip

in a laser with methane call. Direct observation is possible under the condition (1), which can be satisfied at low pressure and at large transit times (the diameter of the light beam is ≥ 10 cm). The critical intensity corresponding to this condition is $I_0 \lesssim 10^{-5}$ W/cm². The criterion (1) can be satisfied also for certain weakly-resolved atomic transitions, for example the intercombination transition $4^1S_0 - 4^3P_1$, $\lambda = 6572$ Å, $I_0 \sim 10^{-4}$ W/cm² in Ca.

²If the criterion (1) is satisfied only for slow atoms, then the depth of the dip decreases.

¹S. G. Rautian, Tr. Fiz. Akad. Nauk SSSR **35**, 43 (1968).

²S. Stenholm and W. E. Lamb, Jr., Phys. Rev. **181**, 618 (1969).

³B. J. Feldman and M. S. Feld, Phys. Rev. **A1**, 1375 (1970).

⁴A. P. Kol'chenko, S. G. Rautian, and R. I. Sokolovskii, Zh. Eksp. Teor. Fiz. **55**, 1864 (1968) [Sov. Phys. JETP **28**, 986 (1969)].

⁵S. Stenholm, Preprint N34-73, Helsinki, 1973.

⁶A. P. Kazantsev, Zh. Eksp. Teor. Fiz. **67**, 1660 (1974) [Sov. Phys. JETP **40**, 825 (1975)].

⁷A. P. Kazantsev, Zh. Eksp. Teor. Fiz. **66**, 1599 (1974) [Sov. Phys. JETP **39**, 784 (1974)].

⁸A. P. Botin and A. P. Kazantsev, Zh. Eksp. Teor. Fiz. **68**, 2075 (1975) [Sov. Phys. JETP **41**, 1038 (1975)].

⁹A. P. Kazantsev, Preprint ITF, Chernogolovka, 1975.

¹⁰A. M. Bonch-Bruevich, N. N. Kostin, V. A. Khodovoi, and V. V. Khromov, Zh. Eksp. Teor. Fiz. **56**, 144 (1969) [Sov. Phys. JETP **29**, 82 (1969)].

¹¹Yu. M. Kirin, D. P. Kovalev, S. G. Rautian, and R. I. Sokolovskii, Pis'ma Zh. Eksp. Teor. Fiz. **9**, 7 (1969) [JETP Lett. **9**, 3 (1969)].

¹²T. Yu. Popova, A. K. Popov, S. G. Rautian, and A. A. Feoktistov, Zh. Eksp. Teor. Fiz. **57**, 444 (1969) [Sov. Phys. JETP **30**, 243 (1970)].

¹³D. L. Hall, Dokl. IV Vavilovskoi konferentsii (Paper at 4th Varilov Conf.) Novosibirsk, 1975.

¹⁴V. S. Letokhov, Piz'ma Zh. Eksp. Teor. Fiz. **7**, 348 (1968) [JETP Lett. **7**, 272 (1968)].

Translated by J. G. Adashko

Nonlinear Doppler-free narrow resonances in optical transitions and annihilation radiation of positronium

V. S. Letokhov and V. G. Minogin

Institute of Spectroscopy, USSR Academy of Sciences

(Submitted January 15, 1976)

Zh. Eksp. Teor. Fiz. **71**, 135-147 (July 1976)

The possibility of Doppler-free narrow resonances is considered for transitions between fine-structure levels of the ground and first excited states of the positronium atom. An analysis is given of the conditions necessary for the observation of narrow saturation resonances in the case of single-quantum absorption in 1S-2P transitions, and narrow two-photon absorption resonances in 1S-2S transitions. It is shown that it is possible to obtain 2γ -annihilation lines from the positronium atom with widths much smaller than the Doppler width.

PACS numbers: 36.10.Dr

1. INTRODUCTION. FORMULATION OF THE PROBLEM

Studies of the structure of the energy levels of the bound system consisting of an electron and a positron, i. e., the positronium (Ps) atom, and precise measure-

ments of the transition frequencies of this atom, provide a unique possibility for a test of relativistic quantum theory. It is well known that there are two experimental methods at present for investigating the positronium energy levels, namely, the direct microwave method in which the fine-structure intervals are deter-