Thus, we have demonstrated the possibility of performing experiments in polarized colliding beams of electrons and positrons.

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[1)]This variation is a method for the accurate absolute measurement of the electron energy in a storage ring, since the anomalous magnetic moment of the electron is known extremely accurately, to $\sim 3\times 10^{-6}$. The accuracy achieved by this method in absolute calibration of the energy of the VÉPP-2M storage ring[13] is 10^{-4}.

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Non-adiabatic frequency resonant radiation

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A formula for the probability of non-adiabatic multiphoton excitation of a molecule from the electronic ground level to an excited level in the field of an external electromagnetic wave is derived by the WKB method. Resonant excitation of vibrations by laser radiation and multiphoton transitions due to the interaction between the electron and the "hot" vibrations are investigated separately.

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1. Many recent experimental and theoretical papers are devoted to the excitation and decay of molecules under the influence of laser radiation of frequency close to the frequencies of dipole-active molecular vibrations (see, e.g., [1–4]). Investigations of the pure vibrational mechanism of excitation and breakup of molecules have by now become traditional.[5,6]

The electronic mechanism of multiphoton excitation of molecules was investigated on the basis of perturbation theory.[7,8] The non-adiabatic channel of multiphoton excitation of molecules in a strong electromagnetic field, has insofar as we know, not been considered. (Naturally, the dissociation limit of the molecule with respect to a given vibration should lie higher than the electron-excitation energy.)

The existence of various dipole moments (d_{ii}, $i=1, 2$) for the ground (1) and excited (2) terms of the molecule leads, in principle, to a new situation for multiphoton transitions, in comparison, say, with multiphoton transitions between two nondegenerate electronic states in an atom.[9] In this case the situation is closer to the scheme of multiphonon transitions in a solid, owing to the possibility of a real intersection of the terms and the appearance of effects of the Landau-Zener type.[10]

An electromagnetic (EM) wave can interact either directly with the dipole moment of an electron or with a dipole-active vibration.[11] The buildup of the latter also activates multiphoton transitions of the electron. We consider first the case when the wave frequency ω is not at resonance with the frequency Ω of the active vibra-

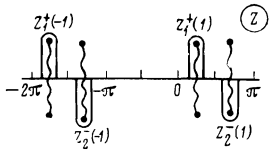

FIG. 1. Complex turning points and contour of the translation of the solution (the contour C circles around the points $z_{1,2}^{\pm}(n)$ in the complex plane).

tion of the molecule and effects of electron-vibrational interaction can be neglected. This problem is of independent interest, as a model problem of multiphoton transition in a generalized two level system (GTS), i.e., in a system with nonzero average dipole moments in each state (or one of two). The case of resonance ($\omega = \Omega$) will be investigated in the Conclusion.

2. We consider multiphoton transitions in a GTS. The equation for the amplitudes $\langle i|\hat{S}|i\rangle$ of a two-level system interacting with a low-frequency alternating electric field of intensity $\bar{F} = F\sin\omega t$ of a classical EM wave can be written in the form ([10], p. 132)

$$\frac{d^2u_i(z)}{dz^2} + u_i(z)\left[\left(\frac{n}{2}\right)^2\chi_0(z) \mp i\frac{n}{2}\chi_1(z)\right] = 0. \tag{1}$$

(The signs $\mp$ pertain respectively to $i=1$ and $i=2$). We used the notation

$$u_i(z) = \exp\left\{-\frac{in}{2}\int_{-\infty}^{z}(1+D\sin z)\,dz\right\}\langle i|\hat{S}(z)|i\rangle,$$
$$z=\omega t, \quad n=(E_2-E_1)/\hbar\omega,$$
$$\chi_0(z) = (1+D\sin z)^2 + V^2\sin^2 z, \quad \chi_1(z) = D\cos z,$$
$$D \equiv \frac{F(d_{22}-d_{11})}{E_2-E_1}, \quad V \equiv \frac{2Fd_{12}}{E_2-E_1},$$

d_{12} is the dipole transition while E_1 and E_2 are the unperturbed GTS energy levels.

The initial conditions for Eq. (1) are governed by the requirements $\langle 1|\hat{S}(-\infty)|1\rangle = 1$ (or respectively $\langle 2|\hat{S}(-\infty)|2\rangle = 0$) and $\langle i|\hat{S}(-\infty)|i\rangle = 0$. We seek the solution of (1) by the WKB method.[12] The points in the vicinity of which the WKB description no longer holds (the turning points) are determined by the condition $\chi_0(z) = 0$. The turning points $z_1^{\pm}$, (n) are distributed in the complex plane with a period 2π (Fig. 1). The solution of equations with pairs of complex-conjugate turning points was investigated in[13]. The translation over N periods ($N = \omega t/2\pi$, $t \to +\infty$) is realized in accordance with[13,14,15]. In contrast to[14], account is taken of the time dependence in the term $V^2\sin^2 z$. The amplitudes of the quasi-classical solutions as $t \to -\infty$ and the corresponding amplitudes as $t \to +\infty$ are connected by a matrix relation. The square of the modulus of the off-diagonal matrix element B of the coupling matrix determines the probability of the transition to the excited level.[13-15]

After making the appropriate calculations, we obtain

$$|B|^2 = |b|^2\frac{\sin^2 N\psi}{\sin^2\psi},$$
$$|b|^2 = 4e^{-2\nu}(1-e^{-2\nu})\sin^2\frac{\mu_+ + \mu_-}{2}, \quad \nu = \frac{n}{2}\oint_C \chi_0^{1/2}(z)\,dz. \tag{2}$$

$$\psi = \arccos\{(1-e^{-2\nu})\cos\mu_1 + e^{-2\nu}\cos\mu_-\}.$$

The contour C circles around the root branch point (see Fig. 1)

$$\mu_{\pm} = \mu_1 \pm \mu_2,$$
$$\mu_1 = \frac{n}{2}\int_{z_1^n}^{z_2^n}\chi_0^{1/2}(z)\,dz, \quad \mu_2 = \frac{n}{2}\int_{z_2^n}^{z_1^{n+1}}\chi_0^{1/2}(z)\,dz;$$

z_1^n and z_2^n are real minimum points of $\chi_0(z)$.[1)] The parameter b in formula (2) corresponds to the off-diagonal element of the coupling matrix for two passes of the complex-conjugate turning points,[13] and its structure reflects the unitarity condition of the matrix. We emphasize that formula (2) is valid only if a "quasi-classical" region exists between two pairs of complex-conjugate turning points ($z_1^{\pm}(n)$ and $z_2^{\pm}(n)$).[2)]

We confine ourselves henceforth to the actual case of not very strong fields ($\nu \gg 1$), for which it is still reasonable to single out the GTS in the system of the terms of the molecule. We write down the formula for the rate of transition as $t(N) \to \infty$:

$$W_{12} \approx \frac{\omega}{2\pi}\tilde{b}^2\delta\left(\frac{\tilde{\psi}}{\pi} - n\right), \quad \tilde{b} = e^{-\nu}\sin\mu_1, \quad \tilde{\psi} = \mu_+. \tag{3}$$

The expression under the δ-function sign has the meaning of the difference between the level energies of the GTS in the EM field, with allowance for the Stark shift (in units of $\hbar\omega$), while $\sin^2\mu$ determines the selection rules. In the particular case $d_{11} = d_{22} = 0$ we obtain from (3) the well-known result of Zaretskiĭ and Krainov.[9] 3)

The expressions for ν and $\mu_{\pm}$ can be expressed in terms of elliptic integrals (of the first, second, and third kind) and elementary functions, but are too cumbersome to be presented here.

Figure 2 shows the universal relation $\tilde{\nu}(F) = \nu(F)/n$ for different values of $\rho = 2d_{12}/|d_{22}-d_{11}|$. The figure shows also the function $\ln|\tilde{b}(F)|$ for the particular case $n=4$ and $\rho=1$. Figure 3 shows the field dependence of the shift $\tilde{\psi}/\pi n$ and of the parameter μ_1.

If $\rho \ll 1$, $D < 1$, formula (3) takes the simple form

$$W_{12} \sim \frac{\omega}{2\pi}\left(\frac{2.718D}{2}\right)^{2n}\sin^2(\mu_1); \tag{4}$$

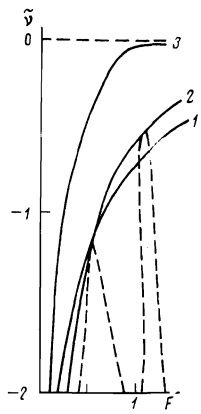

FIG. 2. Dependence of $\nu(\tilde{F})$ (F is in units of E). Curve 1 corresponds to the case $d_{11}-d_{22}=0$, $d_{12}\equiv 1$ (it is analogous to the plot in[9]); 2—to the case $\rho=1$; 3—$\rho=0.1$, where $|d_{11}-d_{22}|/2=1$; the dashed curve corresponds to $\ln|\tilde{b}|/n$, at $\rho=1$ and $n=4$.

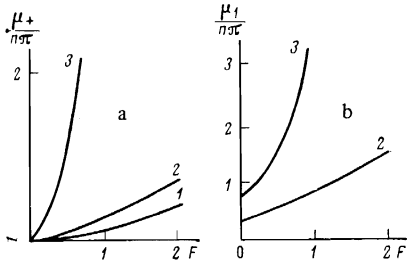

FIG. 3. Plots of $\tilde{\psi}(F)/\pi n \equiv \mu_+(F)/\pi n$ (a) and $\mu_1(F)/\pi n$ (b) (F is in units of E, and $d_{12} \equiv 1$). Curve 1 corresponds to the case $d_{11} - d_{22} = 0$; 2—to the case $\rho = 1$; 3—$\rho = 0.1$.

$\sin^2\mu_1$ "forbids" all the transitions at $d_{12} = 0$, since $\mu_1 \sim n\rho$.

3. We consider the case of resonance between the photon frequency ω and the vibrational quantum frequency Ω. The time dependence of the normal vibration $Q(t)$ under the influence of a field turned on at the instant $t = 0$, under the initial conditions $Q(0) = 0$, $\dot{Q}(0) = 0$, is given by (see, e.g., [17])

$$Q(t) = \bar{Q}(t) + Q_F(t),$$
$$Q_F(t) = Q_0[1 - e^{-\gamma|t|}]\cos\omega t, \quad Q_0 = \left(\frac{M\omega}{\hbar}\right)^{1/2}\frac{2p^2F}{\hbar e_0\gamma}. \tag{5}$$

Here $\bar{Q}(t)$ is the natural damped part of the Q vibration, μ and γ are respectively the reduced mass and the damping constant of an oscillator of frequency Ω, and p is the dipole moment connected with the Q vibration.[18]

Of greatest interest for multiphoton processes is the case of steady-state oscillations corresponding to the instants of time $t > 1/\gamma$; neglecting the small contribution $\bar{Q}(t)$, we obtain for these times

$$Q(t) \approx Q_0 \cos\omega t. \tag{6}$$

Let us consider independently the contributions made to the transition probability by the "direct" electronic mechanism and by the mechanism of the electronic transition induced by the electron-vibrational interaction with the excited mode (we denote the corresponding transition probability by P_{12}).

In the derivation of (2) we used the "translation" of the solutions along the time axis, in which the initial point can be chosen to be $t_0 \approx 1/\gamma$, inasmuch as up to the point t_0 the forced solution is not effective ($Q(t) \approx \bar{Q}(t) + Q_0\gamma t \times \cos\omega t$, the amplitude $Q_0\gamma t$ does not depend on γ, and at the characteristic small values of the time it is small and does not exceed $\bar{Q}$). The turning points corresponding to a growing solution are located at much larger distances from the real axis in the complex plane than the turning points corresponding to steady-state oscillations.

The calculation of P_{12} coincides with the calculation of W_{12}, in which it is necessary only to replace the constants and to shift the phases of the turning points by $\pi/2$ along the real axis. The constants of the electron-vibrational interaction in states 1 and 2 and the mixing matrix element will be designated by V_{11}, V_{22}, and V_{12}, respectively. The minima of the energies of the adiabatic potentials of the electron-vibrational problem in the ground and excited states will be designated I_1 and I_2. In this case $\rho = 2V_{12}/|V_{22} - V_{11}|$ and in the approximation $\rho \ll 1$ we can use formula (4), where

$$D = |V_{11} - V_{22}|Q_0/|I_1 - I_2| < 1.$$

For a real laser source, with allowance for its multimode structure, the probability (4) should be averaged with a Gaussian weight function

$$P_{12}^{(G)} = n!P_{12} \tag{7}$$

(analogously for $W_{12}^{(G)}$).

4. We consider now the influence of a weak anharmonicity of the Q vibration on the probability of the multiphoton excitation. We confine ourselves to the Duffing model[19] (see also[20]). The anharmonicity term in this model is $\alpha\Omega^2Q^3$, where α is the dimensionless anharmonicity constant. The steady-state vibrational solution is of the form

$$Q_F(t) = \frac{f}{\Omega^2}(x_1\cos\omega t + \varepsilon x_3\cos 3\omega t + \ldots),$$
$$f = \left(\frac{M\Omega}{\hbar}\right)^{1/2}\frac{2p^2F\Omega}{\hbar e_0}, \quad \varepsilon = \frac{\alpha f^2}{\Omega^4}, \tag{8}$$
$$x_1 \approx \frac{\Omega}{\gamma\sqrt{2}}\left(1 + \left(1 - \frac{3}{4}\frac{|\varepsilon|}{\gamma^2}\Omega^2\right)^{1/2}\right)^{-1/2}, \quad x_3 \approx x_1^3/36.$$

Since the deviation from the principal resonance $\omega = \Omega$ is negligible, the first term of (8) practically coincides with the law (6) investigated above. The appearance of higher harmonic in (8) imposes a stringent limitation on the anharmonicity parameter α, for which it is correct to confine oneself to the first terms of (8).

In particular, it can be shown that discarding the third harmonic for multiphoton transitions under conditions of formula (4) imposes the following restrictions on the anharmonicity parameter:

$$|\alpha| < 3[V_{11} - V_{22}]^2/[I_1 - I_2]^2.$$

5. Let us obtain numerical estimates. In the case of a pure electronic excitation mechanism, we consider molecules for which the GTS model is applicable. (These may be molecules in which the dipole moment of the ground state exceeds the dipole moment of the excited state, not a rare occurrence. The GTS model is applicable also to molecules in which the highly excited states are poorly mixed with the first excited state.) For the parameters $|d_{11} - d_{22}| = 3D$, $d_{12} = 0.1\ D$, $E_2 - E_1 \sim 10$ eV, $\hbar\omega = 2.34$ eV, according to formulas (4) and (7), if a picosecond laser is used, the gas will be excited if the field is $F \sim 10^6$ V/cm.

Consider the case of a resonance with the vibrational system. The parameter $V_{11} - V_{12}$ is connected with the half-width of the electron-vibrational transition band. Typical ratios are $|V_{11} - V_{22}|/|I_1 - I_2| \sim 0.1$. Consequently, the allowable limit of the anharmonicity parameter is $\alpha < 10^{-2}$. At $\omega \sim 10^{14}$ sec^{-1} the anharmonic shift is $\delta \sim \alpha\hbar\omega < 10$ cm^{-1}.

A transition with a large number of quanta presupposes overcoming the first several vibrational levels as a result of the "anharmonicity overlap" due to rotational, field-induced, etc. broadening. For small anharmonic shifts, on the order of several reciprocal centimeters, this condition is easily realized in a large number of molecules. The highly excited vibrational states are in turn well described by the solution of the classical problem (see (8)). For the parameters written out above and at $V_{12} \sim 0.1 |V_{11} - V_{22}|$, $p \sim 1D$, $M \sim 10^{-25}$ g, $\gamma \sim 10^{11}$ sec^{-1}, and $n = 50$ it is easy to verify with the aid of formulas (4) and (7) that the threshold value of the field intensity, at which complete excitation of the gas takes place, is $F \sim 10^5$ V/cm.

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[1)]We note that the definition of $\mu_{1,2}$ includes a phase φ_0, which can be found by the method of the adjoint equation.[13] We shall consider below, however, the case $\nu \gg 1$, as $\varphi_0 \to 0$.

[2)]See the remark in the paper by Bychkov and Dykhne[15] concerning the applicability of the theory to the case of strong fields.

[3)]In the case of the nonlinear time regime, it is possible to obtain from (2) an equation for the probability $W_2(t)$ of the population of the excited level, analogous to formula (6) of Kraĭnov's paper,[16] in which we must put $R^2 \equiv b^2$.

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Nonlinear resonance in quantum systems

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Both perturbation theory and a quasiclassical method for calculating the quasienergy spectrum have been developed for the problem of a nonlinear quantum oscillator with a resonant force. The results are applied to a description of intermode resonances in autonomous systems. It is shown that quantum effects lead to strong restrictions on the existence of stochastic layers and Arnol'd diffusion and examples of magnetic traps, accelerators, and the solar system are considered. The problem of the decay of an excited mode in molecules and nonlinear chains is discussed. An estimate is made of the limits of transition to a stochastic regime of motion in molecules. A number of the results of the theory is applied to a description of collisionless dissociation of molecules in the field of an intense resonance wave.

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1. INTRODUCTION

In recent years significant progress has been achieved in understanding the overall picture of the motion of an n-dimensional classical system of a general form. It is due to the work of mathematicians, in particular that of Kolmogorov and Arnol'd,[1–3] to the development of the theory and practice of such systems as accelerators of elementary particles, plasma oscillations etc., and particularly to "mathematical experiments" using electronic computers with different model systems.[4–7] This theory is based on a consistent taking into account