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Nonresonant transitions and ionization of atoms in slow collisions in a laser field

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We consider inelastic atomic collisions that take place in the field of intense electromagnetic radiation and are accompanied by absorption of nonresonant photons. The cross sections are calculated for the excitation $[\mathrm{H}(1S)+\mathrm{H}(1S)+\omega_1\rightarrow\mathrm{H}(1S)+\mathrm{H}(2S)]$ and charge exchange $[\mathrm{H}(1S)+\mathrm{H}(1S)+\omega_2\rightarrow\mathrm{H}^+ + H^-]$ of hydrogen atoms in close collisions. The transition cross sections turn out to depend substantially on the form of the potential curves of the quasimolecule H_2. We calculate also the two-photon ionization of hydrogen atoms irradiated by an intense laser pulse in close collisions. The cross section depends little on the form of the potential curves of the quasimolecule, and an estimate of its value shows that collision ionization makes a contribution comparable with that of direct ionization even at medium densities. In the case of remote collisions, an analytic expression is obtained for the cross section of nonresonant transitions following adiabatic turning on the field, with account taken of the Stark shift of the atomic levels.

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1. INTRODUCTION

Much attention has been paid in recent years to the study of the influence of intense electromagnetic radiation on the dynamics of atomic collisions. Interest in this group of problems is due to the possibility of using laser radiation to stimulate various processes that occur in atomic collisions. Most theoretical and experimental studies were devoted to the so-called optical and radiative collisions (see the review by Yakovlenko[1]), which occur at large interatomic distances, and the frequency of the absorbed (emitted) photon is close to the natural frequencies of a system of two non-interacting atoms. These processes have a clearly pronounced resonant character. Single-photon ionization in the case of remote collisions, accompanied by excitation transfer from one atom to another, was considered in Refs. 2 and 3, while two-photon ionization was considered in Ref. 4.

In this paper we consider processes that occur in atomic collisions and are accompanied by absorption of photons whose frequency differs from the natural atomic frequencies by an amount comparable with the frequencies itself. We shall call these processes nonresonant.

We note that nonresonant excitation (detuning ~1000 cm^{-1}) was observed in recent experiments,[5] as well as binding of atoms of alkali-metal vapor into a molecule following collisions in a laser-radiation field. If the detuning reaches a value on the order of the atomic frequency, then the nonresonant processes occur at close collisions, since the close approach can lead to cancellation of the detuning on account of the strong distortion of the energy levels of the interacting atoms (quasimolecule). For a consistent calculation of the cross sections in the adiabatic approximation it is therefore necessary, first, to know the exact parameters of the quasimolecule; second, to take into account the motion of the nuclei along the real rather than a straight-line trajectory. On the other hand, by measuring the cross sections of nonresonant processes in close collisions it is possible to obtain information on the parameters of the quasimolecule (the angular momenta of the transitions, the shapes of the potential curves).

In the next section of this paper we reduce the problem in the adiabatic approximation to a description, in an external field, of a two-level quasimolecular system the upper level of which is shifted and broadened on account of the interaction with the electronic continuum. In Sec. 3 we investigate the case of nonresonant transitions, when the ionization of the atoms during the collision time can be neglected. In Sec. 4 we consider two-photon ionization of atoms in close collisions. The results of the calculation of the nonresonant excitation and of the charge exchange of the hydrogen atoms with photon absorption, as well as of two-photon ionization in close collisions, are discussed in Sec. 5. In Sec. 6 we take into account the influence of adiabatic turning on the field on the nonresonant transition in the case of remote collisions and relatively small detuning.

2. SOLUTION METHOD

The motion of the atomic nuclei will be described classically. In the case of close collisions, when the transitions occur at distances on the order of several atomic units between the nuclei, this approximation is not obvious. Thus, for the lightest hydrogenlike atoms moving with thermal velocities, the deBroglie wavelength is close to the atomic unit. We consider only the case when the potential curve of the relative nuclear motion is not repulsive. Then, for example, when the hydrogen atoms come close together their relative velocity increases by at least one order of magnitude. Consequently, in this case, when describing the motion of hydrogen as well as of heavier atoms, we can use the concept of classical trajectory.

The Schrödinger equation for two colliding atoms interacting with electromagnetic field can be written in the form¹⁾

$$i\partial\Psi(\mathbf{r},t)/\partial t=\{H(\mathbf{r},R(t))+V(t)\}\Psi(\mathbf{r},t). \tag{1}$$

Here $\mathbf{r}$ is the aggregate of the electron coordinates, R is the distance between the nuclei, and H is the Hamiltonian that determines the set of eigenfunctions $\Phi_n(\mathbf{r},R)$ and terms $U_n(R)$ of the quasimolecule:

$$H(\mathbf{r},R)\Phi_n(\mathbf{r},R)=U_n(R)\Phi_n(\mathbf{r},R),$$

$V(t)=-\mathbf{F}_0\mathbf{d}[\mathbf{r},\mathbf{R}(t)]e^{i\omega t}/2+\text{H.c.}$ is the operator of interaction with the electromagnetic wave in the dipole approximation, and $\mathbf{F}_0$ and ω are the amplitude and frequency of the wave. The velocity of the nuclei is assumed to be small compared with the velocities of the electron motion, so that the function $R(t)$ varies adiabatically slowly.

It is convenient to analyze the problem qualitatively by using the formalism of quasienergy states (QES).⁶ When the interaction $V(t)$ is turned off, the set of QES and of the quasienergies U_{nk} of Eq. (1) takes at fixed R the form

$$\Phi_n(\mathbf{r},R)\exp(-iU_n(R)),\ U_{nk}(R)=U_n(R)+k\omega,\quad k=0,\pm1,\ldots$$

The perturbation $V(t)$ will obviously lead to transitions between the QES of the discrete spectrum, for which there exists a point of intersection of the quasienergies R_ω, and to a decay of the QES that land in the electronic continuum. We specify a definite arrangement of the continuum and of the two discrete terms of interest to us, as functions of R. To derive the system of equations it is more convenient to make the continuum boundary $U_0(R)$ independent of R and reckon the energy from it. It is easy to see that this can be done by replacing the operator H by $H-U_0(R)$. The corresponding arrangement of the discrete terms and of the continuum is indicated in Fig. 1. We thus arrive at the problem of transitions between two quasistationary QES following an adiabatic change of the parameter $R(t)$. Starting with the assumptions indicated above, we seek the solution of Eq. (1) in the form

$$\begin{aligned}\Psi(\mathbf{r},t)=&\sum_{n=1}^{2}a_n(t)\Phi_n(\mathbf{r},R(t))\exp\left\{-i\int^t U_n(R(t))dt\right\}\\&+\int d\nu b_\nu(t)\Phi_\nu(\mathbf{r},R(t))\exp\left\{-i\int^t U_\nu(R(t))dt\right\}.\end{aligned}\tag{2}$$

We note that a model that makes use of a coupling of two discrete states and the electronic continuum can be justified only in the resonance approximation.

Substituting the function (2) in the Schrödinger equation (1), using the adiabaticity condition (we neglect terms containing $\partial\Phi_i/\partial t$), and omitting, in addition, the rapidly oscillating terms, we obtain in the resonance approximation the following system of equations:

$$\begin{aligned}i\dot a_1&=V_{12}\exp\left\{i\int^t(U_1-U_2+\omega)dt\right\}a_2,\\i\dot a_2&=V_{12}^*\exp\left\{i\int^t(U_2-U_1-\omega)dt\right\}a_1+\int b_\nu V_{2\nu}\exp\left\{i\int^t(U_2-U_\nu+\omega)dt\right\},\\i\dot b_\nu&=V_{2\nu}^*\exp\left\{i\int^t(U_\nu-U_2-\omega)dt\right\}a_2,\end{aligned}\tag{3}$$

where

$$V_{12}=\langle\Phi_1|\mathbf{d}(\mathbf{r},\mathbf{R}(t))|\Phi_2\rangle F_0/2,\quad V_{2\nu}=\langle\Phi_2|\mathbf{d}(\mathbf{r},\mathbf{R}(t))|\Phi_\nu\rangle F_0/2.$$

In the derivation of the system (3) it was assumed, just as in Refs. 4 and 7, that there is no coupling between the states of the continuum, i.e., the matrix elements $V_{\nu\nu'}$ are small.

We assume that prior to the collision the atoms were in their unperturbed ground states. This corresponds to choosing the initial condition for the integration of Eqs. (3) in the form

$$a_1(-\infty)=1,\quad a_2(-\infty)=b_\nu(-\infty)=0. \tag{4}$$

The condition (4) corresponds, generally speaking, to instantaneous turning on the electromagnetic field. But if the action of the field on the isolated atoms can be neglected, then the cross sections of the nonresonant processes should not depend on the manner in which the field is turned on prior to the collision.

The system (3) can be further simplified by assuming that the second term lies sufficiently close to the con-

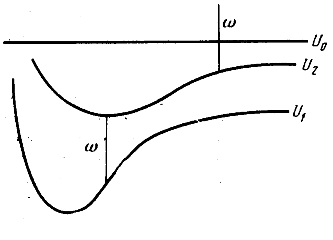

FIG. 1.

tinuum:

$$\max|U_2(R)-U_2(R')|\ll\min(U_2(R)+\omega). \tag{5}$$

In this case we can approximately assume that $U_2(R)-U_\nu(R)+\omega$ is constant. We integrate by parts the right-hand side of the third equation of the system (3). The functions $V_{2\nu}$ and a_2 vary slowly with time, so that we can neglect their derivatives. Substituting the resultant expression for $b_\nu(t)$, with account taken of the initial condition (4), in the second equation of (3), we arrive at a system of two equations:

$$\begin{gathered}i\dot{a}_1=V_{12}\exp\left\{i\int^t(U_1-U_2+\omega)\,dt\right\}a_2,\\ i\dot{a}_2=V_{12}^*\exp\left\{-i\int^t(U_1-U_2+\omega)\,dt\right\}a_1+a_2\Delta-ia_2\Gamma/2,\end{gathered} \tag{6}$$

where the shift of the second level on account of the interaction with the continuum is

$$\Delta(t)=P\int_0^\infty dU_\nu\frac{\int|V_{2\nu}|^2d\nu/dU_\nu}{U_2-U_\nu+\omega},$$

and the broadening of the second level on account of the interaction with the continuum is

$$\Gamma(t)=2\pi\int|V_{2\nu}|^2d\nu/dU_\nu,\quad U_\nu=U_2+\omega;$$

$d\nu=dU_\nu d\nu/dU_\nu$ takes into account the degeneracy of the continuum.

In the case of terms that do not depend on the time, the obtained system of equations coincides with the equations of Ref. 7. We note that the system (6), without indication of the condition (5) for its validity, was used in Ref. 4 in an investigation of remote collisions between atoms.

The total ionization probability is given by

$$W_i(t)=\int d\nu|b_\nu|^2=\int_{-\infty}^t|a_2(t)|^2\Gamma(t)\,dt; \tag{7}$$

where the amplitudes satisfy the normalization condition

$$|a_1(t)|^2+|a_2(t)|^2+W_i(t)=1. \tag{8}$$

Following the phase transformation

$$a_2=b_2\exp\left\{-i\int^t\Delta dt\right\}$$

the system (6) reduces to

$$\begin{gathered}i\dot{a}_1=V_{12}b_2\exp\left\{-i\int^t\delta(t)\,dt\right\},\\ i\dot{b}_2=V_{12}^*a_1\exp\left\{i\int^t\delta(t)\,dt\right\}-ib_2\Gamma/2,\end{gathered} \tag{9}$$

where

$$\delta(t)=U_2(R(t))+\Delta(t)-U_1(R(t))-\omega.$$

In the vicinity of the intersection point t_ω, $\delta(t_\omega)=0$, we solve the system (9) in an approximation similar to that of Landau and Zener, i.e., we assume V_{12} and Γ to be constant, and $\delta(t)=(t-t_\omega)d\delta(t_\omega)/dt$. After eliminating the amplitude a_1, the system (9) reduces to the Weber equation[8]

$$d^2y/dz^2+(n+{}^1/_2+z^2/4)y=0,$$

where

$$\begin{gathered}y=b_2\exp\left\{\int_{t_\omega}^t(\Gamma-i\delta(t))\,dt/2\right\},\\ z=\left[t-t_\omega-\frac{i\Gamma}{d\delta(t_\omega)/dt}\right]\left[\frac{d\delta(t_\omega)}{dt}\right]^{1/2}\exp\left(\frac{i\pi}{4}\right),\\ n=-\frac{i|V_{12}|^2}{d\delta(t_\omega)/dt}.\end{gathered}$$

The general solution of this equation can be represented in the form of a linear combination of two parabolic-cylinder functions:

$$y=C_1D_n(-z)+C_2D_{-n-1}(iz).$$

Using the initial condition (4) we get

$$C_1=0,\quad|C_2|^2=in\exp(-i\pi n/2).$$

If we use the asymptotic form of the function $D_{-n-1}(iz)$ as $t\to\infty$, then we can obtain the following expression for the probability of exciting the second state after passing through the intersection point:

$$|a_2(t)|^2=(1-P)\exp\{-\Gamma(t-t_\omega)\}, \tag{10}$$

where

$$P=\exp\left(-\frac{2\pi|V_{12}|^2}{|d\delta/dt|}\right),\quad t=t_\omega. \tag{11}$$

Joining together the function (10) to the solution of the system (6) far from the intersection point, where there are no transitions between the states, we get

$$|a_2(t)|^2=\begin{cases}(1-P)f(t), & t>t_\omega\\ 0, & t<t_\omega\end{cases},\quad f(t)=\exp\left(-\int_{t_\omega}^t\Gamma dt\right). \tag{12}$$

Substituting (12) in (7), we obtain the probability of ionization after passage through the intersection point:

$$W_i(t)=(1-P)(1-f(t)). \tag{13}$$

The probability of remaining in the first state can be determined for example, by using the normalization condition (8), from which it follows that

$$|a_1|^2=P. \tag{14}$$

We note that the shift Δ is relatively small. Therefore, with sufficient accuracy, the intersection point, R_ω is determined from the condition $U_2(R_\omega)=U_1(R_\omega)+\omega$, and we can put in the denominator of the exponential (11)

$$d\delta(t_\omega)/dt\approx v(R)\,d(U_2-U_1)/dR,\quad R=R_\omega, \tag{15}$$

where $v(R)$ is the radial velocity of the relative motion of the nuclei.

3. NONRESONANT TRANSITION IN CLOSE COLLISIONS

We assume that the ionization during the time of motion in the transition region can be neglected, i.e., $\Gamma\tau\ll1$ (τ is the characteristic time of motion between the term-intersection points). Putting $\Gamma=0$ in (10) we obtain the transition probability after passage through the intersection point. The probability of remaining in the initial state is determined as before by (14).

The electromagnetic field is assumed to be linearly polarized, so that the matrix element V_{12} in (11) can be represented in the form

$$V_{12}=F_0d_{12}\cos\theta/2, \tag{16}$$

where d_{12} as the matrix element of the operator of the electron dipole moment between the states 1 and 2 of the quasimolecule, and θ is the angle between the quasimolecule axis and the field intensity F_0.

The probability of remaining on the i-th term after the j-th passes through the intersection point $R_\omega (i, j = 1, 2)$ is determined, according to (11), (14), and (15), by the relation

$$P_j^{(i)} = \exp(-S_j^{(i)}), \tag{17}$$

where

$$S_j^{(i)} = \frac{\pi F_0^2 |d_{12}(R)|^2}{2v(R)\,|d(U_2-U_1)/dR|} \cos^2\theta_j^{(i)}(R), \quad R=R_\omega. \tag{18}$$

The interaction (16) in the numerator of expression (18) leads to repulsion of the adiabatic quasienergy terms at the point of their intersection.

The angle $\theta_j^{(i)}$ is connected with the angle $\varphi_j^{(i)}$, which is determined from the classical equations of motion of a particle having a reduced mass M in a central field:

$$\varphi_1^{(i)} = \int\limits_{R_\omega}^{\infty} K_{ii}\,dR - \pi/2, \quad \varphi_2^{(i)} = \varphi_1^{(i)} + 2\int\limits_{R_b^i}^{R_\omega} K_{ii}\,dR,$$

$$\varphi_2^{(j)} = \varphi_1^{(i)} + 2\int\limits_{R_b^j}^{R_\omega} K_{ij}\,dR,$$

where

$$K_{ij} = Mv_0 b[2M(Mv_0^2/2 + U_i(\infty) + \operatorname{sign}(U_j - U_i)_{R\omega\infty} - U_j(R) - Mv_0^2 b^2/2R^2)]^{-1/2}/R^2,$$

R_b^i is the classical turning point. The angles $\theta_j^{(i)}$ and $\varphi_j^{(i)}$ are connected by the relation

$$\cos\theta_j^{(i)} = \cos\alpha\sin\varphi_j^{(i)} + \sin\alpha\cos\varphi_j^{(i)}\sin\beta,$$

which follows from Fig. 2. The radial velocity $v(R_\omega)$ at the intersection point is given by

$$v(R_\omega) = [2(Mv_0^2/2 + U_1(\infty) - U_1(R_\omega) - Mv_0^2 b^2/2R_\omega^2)/M]^{1/2},$$

where v_0 is the initial velocity of the relative motion of the nuclei and b is the impact parameter.

The probability of the transition after double passage through the intersection point can be obtained from the formula

$$W = P_1^{(1)}(1 - P_2^{(1)}) + (1 - P_1^{(1)})P_2^{(2)}. \tag{19}$$

The cross section for inelastic collision with absorption (emission) of a photon is determined by multiplying the probability (19) by the impact parameter and integrating with respect to the value of this parameter up to $b_{\max}$ and over all the directions in the plane perpendicular to v_0 (see Fig. 2):

$$\sigma = \int\limits_0^{2\pi} d\beta \int\limits_0^{b_{\max}} Wb\,db. \tag{20}$$

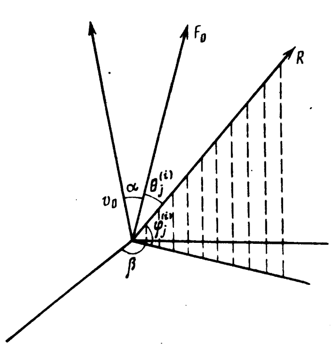

FIG. 2.

The upper limit $b_{\max}$ is obtained from the condition that a turning point appear in the region $R \geqslant R_\omega$ in the case of motion in the field $U_1(R)$, i.e., from the condition that the radial velocity $v(R)$ vanish. We note here that we are considering the case when no bound or quasibound motion states are produced when going over to the potential curve $U_2(R)$.

If the angle α between the directions of the initial velocity of the relative motion of the nuclei and the field $\mathbf{F}_0$ is not fixed, the cross section (20) must be averaged over all the angles:

$$\bar{\sigma} = \int\limits_0^{\pi} \sigma\,d\alpha/\pi. \tag{21}$$

The rate constant of the process is obtained by averaging over the Maxwellian distribution of the velocities:

$$K = \langle v_0 \bar{\sigma} \rangle.$$

A regime that is linear in the field intensity is obtained if $S_j^{(i)} \ll 1$ in the exponential of (18). In this case the expression for the transition probability (19) simplifies to

$$W = S_1^{(1)} + S_2^{(1)}. \tag{22}$$

The cross section (20) is given, after integration with respect to the angle β, by

$$\sigma = \frac{\pi^2 F_0^2 |d_{12}|^2}{2|d\Delta U/dR|} \int\limits_0^{b_{max}} \frac{b\,db}{v(R)} [\sin^2\alpha(\cos^2\varphi_1^{(1)} + \cos^2\varphi_2^{(1)}) + 2\cos^2\alpha(\sin^2\varphi_1^{(1)} + \sin^2\varphi_2^{(1)})], \quad R=R_\omega,$$

from which, in particular, it follows that $\sigma(\alpha) = \sigma(\pi - \alpha)$.

We note that if single-photon ionization from the upper state of the quasimolecule is possible, as proposed in Sec. 2, then $\Gamma\tau \sim S_j^i$ and the ionization of the quasimolecule during the collision time cannot be neglected in the nonlinear regime.

4. COLLISIONAL IONIZATION

We consider the case when the collisional ionization is significant, i.e., $\Gamma\tau \gtrsim 1$. Assuming a double passage through the intersection point, we represent the expression for the ionization probability in the form

$$W_i(t) = (1 - P_1^{(1)})[1 - f(t_\omega + \tau_2)] + P_1^{(1)}(1 - P_2^{(1)})[1 - f(t)/f(t_\omega + \tau_1)] + (1 - P_1^{(1)})P_2^{(2)}[f(t_\omega + \tau_2) - f(t)],$$

where $\tau_{1,2}$ is the time of motion between the intersection points over the first and second terms. The ionization cross section σ_i during the time T of the action of the laser pulse is determined in the same manner as the cross section of the nonresonant transition (20), in which W (19) was replaced by $W_i(T)$:

$$\sigma_i = \int\limits_0^{2\pi} d\beta \int\limits_0^{b_{max}} W_i(T)\,b\,db. \tag{23}$$

We consider the regime of ionization saturation in collision, i.e., we assume that $\Gamma\tau \gg 1$ for τ intervals of the order of the time of motion between the intersection point and the time of action of the laser pulse. As follows from (12) and (13), in this case the ionization probability is equal to the probability of excitation of the second level in the absence of broadening of the latter. After double passage through the intersection point, the ionization probability is determined by the formula

$$W_i=1-P_1^{(1)}+P_1^{(1)}(1-P_2^{(1)})=1-P_1^{(1)}P_2^{(1)}.$$

In Sec. 5 we shall be interested in the case of ionization of the quasimolecule through two intermediate states 2 and 2′, when there exists also an intersection point $R'_\omega > R_\omega$ of the ground term and the term 2′.

We denote by $Q_j^{(1)}$ the probability of remaining in the ground state after passing through the point R'_ω ($j=1, 2$). We then obtain for the probability of ionization in the saturation regime, after double passage through each of the intersection points, the following expression:

$$W_i=1-Q_1^{(1)}+Q_1^{(1)}(1-P_1^{(1)})+Q_1^{(1)}P_1^{(1)}(1-P_2^{(1)}) +Q_1^{(1)}P_1^{(1)}P_2^{(1)}(1-Q_2^{(1)})=1-Q_1^{(1)}Q_2^{(1)}P_1^{(1)}P_2^{(1)}, \tag{24}$$

where $Q_j^{(1)}$ is determined by Eqs. (17) and (18), in which the parameters of the term 2 must be replaced by the parameters of the term 2′ of the quasimolecule.

5. CALCULATION OF THE EXCITATION, CHARGE EXCHANGE, AND IONIZATION OF HYDROGEN ATOMS

By way of example we consider close collisions of hydrogen atoms. The numerical values of the terms of the quasimolecule H_2 and of the dipole moments of the transition were taken from Ref. 9, in which the wave functions of the quasimolecule were constructed in the form of expansions in the complete system of the functions of the molecular ion H_2^+. The infinite system of homogeneous algebraic equations for the expansion coefficients was terminated in a self-consistent manner to suit the specified calculation accuracy. As a result it was possible to obtain the wave functions and the terms of both the ground of several excited states of the quasimolecule, by diagonalizing a matrix of finite order. Figures 3 and 4 show the terms and dipole moments of the electronic transitions of the quasimolecule H_2, needed for our calculations and obtained by this method, as functions of the internuclear distance R.

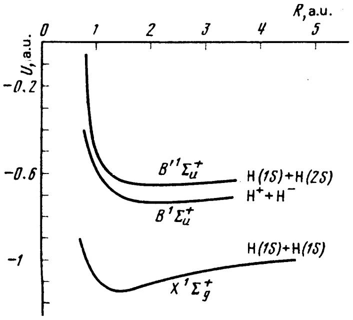

FIG. 3. Terms of the quasimolecule H_2, with account taken of the repulsion between the nuclei, as functions of the internuclear distance. On the right is indicated the free-atom state into which the given quasimolecular term goes over.

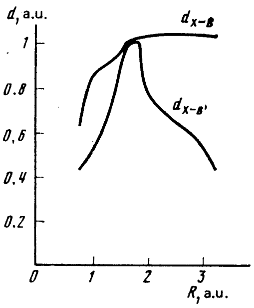

FIG. 4. Dipole moments of the transitions of the quasimolecule H_2, calculated for the terms indicated in Fig. 2.

A. Excitation and charge exchange

Using the results of section 3, we obtain the cross sections of the nonresonant excitation and charge exchange of hydrogen atoms with photon absorption:

1). $H(1S)+H(1S)+\omega_1 \rightarrow H(1S)+H(2S)$,
2). $H(1S)+H(1S)+\omega_2 \rightarrow H^+ + H^-$.

For the excitation process, the states 1 and 2 are the ground state $X^1\Sigma_g^+$ and $B'^1\Sigma_u^+$ terms of the quasimolecule, while the charge exchange process these states are the ground and $B^1\Sigma_u^+$ terms (see Fig. 3).

Estimates of the parameter $S_j^{(i)}$ (18) show that a regime linear in the field intensity is observed for hydrogen atoms that collide at thermal velocities up to fields ~10^7 V/cm, and the cross sections reach at these intensities ~10 a.u. (~3×10^{-16} cm^2). In the considered transitions, the energy of the absorbed photon is ~10 eV, so that single-photon ionization from the states of the quasimolecule is possible. As noted in Sec. 3 in this case the ionization can be neglected only in the linear regime. Calculation of the probability by formula (19) does not make it possible therefore to determine the transition cross section in the nonlinear regime, but shows that a correct allowance for the angular dependence leads to a vanishing of the exponential decrease of the cross section. In fact, the vanishing of $\cos\theta_j^{(i)}$ in (18) at a certain value of the impact parameter keeps the cross section (20) from decreasing exponentially with increasing intensity of the laser radiation. In particular, at an intensity $F_0 = 5\times10^8$ V/cm the cross section levels off and begins to decrease weakly only in fields ~10^9 V/cm. We indicate that in Ref. 5 no decrease of the cross section of nonresonant excitation of rubidium and cesium atoms was observed up to the maximum intensity attained in the experiment. It follows from our estimates that a similar effect will be observed also in collisions of hydrogen atoms, but at laser-field intensities higher than in the experiment of Ref. 5.

Figure 5 shows the dependence of the excitation and charge-exchange cross sections of hydrogen atoms in a regime linear in the field intensity on the internuclear distance at which intersection of the terms takes place. No averaging over the Maxwellian distribution was carried out, and the velocity v_0 was assumed equal to the thermal velocity. In our case the cross section has at any rate a weak dependence on the initial velocity, for when the atoms come close together the change of the

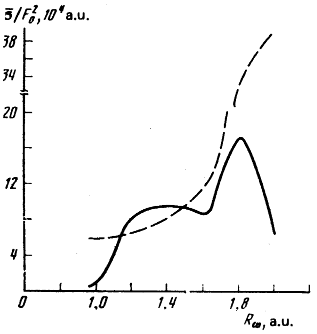

FIG. 5. Cross sections for the excitation (solid line) and charge exchange (dashed) of the colliding hydrogen atoms, as functions of the term intersection point R in the regime linear in the field intensity.

quasimolecule term energy greatly exceeds the kinetic energy of their relative motion at room temperature.

The dependence of the cross sections on the frequency can be obtained if one knows the connection between the intersection point and the frequencies ω_1 and ω_2 of the excitation and charge exchange, respectively; this connection is given in Table I for the R_ω interval represented in Fig. 5.

Let us dwell briefly on an analysis of the obtained curves (see Fig. 5). The maximum near the point 1.4 a.u. for the excitation cross section is connected with the minimum, at this point, of the X term of the H_2 molecule, while the maximum at the point 1.8 a.u. is connected with the minimum of the B' term in its vicinity. For the charge-exchange cross section the first maximum vanishes and is offset by the abrupt decrease of the slope of the term of the excited state and by the increase of the dipole moment of the transition (see Figs. 3 and 4).

Analyzing the experimental data on the frequency dependence of the cross sections of the nonresonant transitions we can thus obtain information on the structure constants of the quasimolecule.

B. Ionization in the saturation regime

We calculate now the cross section of two-photon ionization of the hydrogen atom from the ground state in the saturation regime, with the atoms colliding at thermal velocity v_0. The frequency ω is chosen such that the intersection point R'_ω of the ground term $X^1\Sigma_g^+$ and the term $B'^1\Sigma_u^+$ of the quasimolecule H_2 be of the order of several atomic units. At such a frequency there exists also an intersection point $R<R'_\omega$ of the ground term and the term $B^1\Sigma_u^+$ (see Fig. 3). Consequently, in our case the ionization proceeds via two intermediate states of the quasimolecule.

TABLE I. Dependence of the term intersection point on the frequency of the absorbed photon for excitation and charge exchange of colliding hydrogen atoms.

R_ω, a.u.	ω_1, eV	ω_2, eV	R_ω, a.u.	ω_1, eV	ω_2, eV
1.0	16.09	13.91	1.6	13.67	11.38
1.2	14.65	12.96	1.8	12.97	10.72
1.4	14.33	12.07	2.0	12.58	10.25

Using formulas (23) and (24), and assuming the terms of X, B, and B' to be respectively the states, 1, 2, and 2′ (see Sec. 4), we can calculate the ionization cross section. To realize the saturation regime, the field intensity F_0 is assumed to be 10^{-1} a.u. (5×10^8 V/cm).

Table II lists the cross section σ_i averaged over the angle α between the field intensity and the initial velocity of the relative motion of the nuclei, for several values of the frequency ω. As seen from the table, the cross section depends little on the frequency, and consequently on the position of the term into section points (on their form), in contrast to the excitation and the charge-exchange cross sections calculated in the regime linear in the field intensity for the same frequency interval. The weak frequency dependence of the cross section is due both to the nonlinear regime and to the interference of the contributions made to the ionization by the two intermediate levels of the quasimolecule H_2. Naturally, this conclusion need not necessarily be drawn for collisions of other atoms.

From the known value of the cross section we can estimate the number of ions produced per unit volume and per unit time, $N_i = n^2 v_0 \bar{\sigma}_i$ (n is the density of the atoms). If $\bar{\sigma}_i \sim 100$ a.u., then at a density $n \sim 10^{20}$ cm^{-3} (pressure $\gtrsim 1$ Torr) the yield of ions due to the collisions becomes comparable with the yield of the ions due to direct two-photon ionization of the isolated hydrogen atoms. With decreasing field intensity the collisional polarization will make a contribution comparable with the direct ionization even at lower particle densities. The effect will be observed at least up to intensities such that the single-photon ionization of the excited state of the quasimolecule manages to take place completely within the collision time. The reason is that the probability of the direct two-photon ionization decreases like F_0^4, whereas the probability of the transition of a quasimolecule into an excited state increases like F_0^2 in the linear regime and depends little on the field intensity and the saturation regime.

Thus, collisional ionization in an intense electromagnetic field makes a substantial contribution to the total number of produced ions even at medium densities of the colliding atoms.

6. REMOTE COLLISIONS. ADIABATIC TURNING ON THE FIELD

In the case of remote collisions, the terms of the quasimolecules can be constructed analytically, and the

TABLE II. Dependence of the cross section of two-photon ionization of colliding hydrogen atoms on the laser-radiation frequency in the saturation regime. The table lists also the internuclear distances at which excitation of the quasimolecule takes place (term intersection points).

ω, eV	$\bar{\sigma}_i$, a.u.	R'_ω, a.u.	R_ω, a.u.	ω, eV	$\bar{\sigma}_i$, a.u.	R'_ω, a.u.	R_ω, a.u.
13.67	157.1	1.6	1.016	12.58	158.3	2.0	1.104
12.97	165.3	1.8	1.070	11.01	166.0	3.2	1.295

motion of the atoms can be regarded as linear, $\mathbf{R}=\mathbf{b}+\mathbf{v}_0 t$. The ensuing simplifications make it possible to take into account the influence exerted on the noninteracting atoms (prior to the collision) of an intense electromagnetic field that is turned on adiabatically. It suffices for this purpose to consider transitions between the QES of the atoms.

Choosing the atomic QES constructed in Ref. 10 as the basis, we take into account the dipole-dipole interaction in the Hamiltonian (1) by perturbation theory. As a result we obtain the following expression for the quasienergy term in second order of the interatomic interaction and in F_0

$$U=\varepsilon_a+\varepsilon_b+U_d+U_V,$$

where

$$U_d=\alpha_a(\omega)\alpha_b(\omega)(1-3\cos^2\theta)F_0^2/2R^3$$

is the energy of the induced dipole-dipole interaction, $U_v=C_6/R^6$ is the energy of the van der Waals interaction, $\varepsilon_{a,b}=E_{a,b}-\alpha_{a,b}(\omega)F_0^2/4$ is the quasienergy for different atoms a and b with account taken of the Stark shift of the energy levels $E_{a,b}$, and $\alpha_{a,b}(\omega)$ is the dynamic polarizability of the atoms as calculated in Ref. 10.

Assume that the collision causes the atom b to be excited and to go from state 1 into state 2, and the system absorbs at the same time k photons. We introduce the resonance detuning $\Delta_k=E_b^{(2)}-E_b^{(1)}-k\omega$. We consider the case of a weak laser field, when the Stark shift is much less than the detuning:

$$|\alpha_b(\omega)F_0^2/4|\ll|\Delta_k|, \tag{25a}$$

$$|U_d|\ll|U_V|, \text{ но } |U_V|\sim|\Delta_k|. \tag{25b}$$

If we put $\Delta_k\sim10^2-10^3$ cm^{-1}, then the condition (25a) is satisfied for alkali-metal atoms up to fields ~10^5 V/cm, and condition (25b) is satisfied up to fields ~10^7 V/cm, when $R>10$ a.u.

A formula similar to (18) can be obtained for the parameter S. The matrix element of the operator of the configuration interaction, which leads to repulsion of the quasienergy levels, is calculated in the perturbation-theory technique for degenerate QES,[11] i.e., it is taken over the configuration space and is averaged over the period of the field with a weight $\exp(ik\omega t)$. As a result, the first term that does not vanish as a function of the field turns out to be proportional to F_0^k. Using Eqs. (20)-(22) and recognizing that in the case of straight-line motion we have $b_{\max}=R_\omega$, and that

$$v(R_\omega)=(R_\omega^2-b^2)^{1/2}v_0R_\omega^{-1},$$

we get

$$\bar\sigma=\sigma=8\pi^2R_\omega^9|V^k|^2F_0^{2k}/7|C_6^{(2)}-C_6^{(1)}|v_0,$$

where V^k is a composite matrix element of order k of the configuration-interaction operator and of the dipole moment, $C_6^{(1)}$ and $C_6^{(2)}$ are the Van der Waals constants for atoms interacting in the ground and excited states.

The term intersection point R_ω is defined by

$$R_\omega=\left(\frac{C_6^{(2)}-C_6^{(1)}}{\Delta_k-(\alpha_b^{(2)}(\omega)-\alpha_b^{(1)}(\omega))F_0^2/4}\right)^{1/6},$$

from which it is seen that when both conditions (25) are satisfied the intersection point does not depend on the field intensity and the cross section is proportional to the k-th power of the field intensity. When the Stark shift of the atomic levels it is taken into account [the condition (25a) is violated] the intersection point turns out to depend on the field intensity. This leads to a nonlinear dependence of the cross section on the intensity even at $k=1$, on account of the adiabatic turning on of the field.

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1) We use in this paper the atomic system of units.

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