$A(\lambda_e)$ detected by two different methods make it possible to establish at least one of the reasons for the nonexponential decay of the Nd^{3+} luminescence in LGS-28 glass and for the λ_e dependence of τ_{av}—the reason associated with the variations of the radiative transition probabilities A for different optical centers in the glass. Just the fact that a λ_e dependence of τ has been observed, however, is not enough to permit a correct conclusion to be reached concerning the mechanism responsible for the changes in the τ values for the different centers.

Thus, our results show that by using nonresonant selective excitation one can obtain new information concerning the Stark structure, the variations of the decay kinetics, and the variations of the radiative transition probabilities A for various optical centers in a disordered activated medium, i.e., information that is usually quite hidden by large inhomogeneous broadening when traditional spectroscopic methods are used.

[1)] In an earlier study[7] it was not possible to detect the λ_e dependence of τ because of the higher temperature ($T = 300°K$) and broader-band excitation sources ($\Delta\lambda_{gen} = 1.0$—$1.5$ nm) that were used.

[2)] The choice of a short delay time of 10^{-5} sec in recording the excitation spectra eliminates the effects of slow nonradiative deactivation processes such as intracenter relaxation and $Nd^{3+} \rightleftarrows Nd^{3+}$ cross relaxation, which, as our measurements show, cannot change the population of the $^4F_{3/2}$ level in so short a time as 10^{-5} sec when the Nd^{3+} concentration is as low as it was (0.3 wt.%).

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Translated by E. Brunner

Investigation of the absorption spectrum of a two-level system subjected to intense two-photon excitation

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(Submitted 15 June 1977)
Zh. Eksp. Teor. Fiz. **74**, 67–74 (January 1978)

We investigate the absorption spectrum of weak emission from a two-level system (the Zeeman-splitting levels of ^{113}Cd atoms) under intense two-photon excitation. It is observed that, at sufficiently high probability of two-photon excitation, a change in the sign of the absorption coefficient (i.e., amplification) takes place without population inversion, analogous to the changes of the absorption contour in resonant (single-photon) excitation. A theory is constructed that describes the shape of the absorption line in the case of two-photon excitation of a two-level system; the results of the theory agree well with the experimental data. It is shown that reversal of the sign of the absorption coefficient can be used to convert laser emission frequency (to generate the third optical harmonic). Estimates of the efficiency of the frequency tripling of the 7784 Å emission in sodium vapor show that near the third-harmonic frequency it is possible to attain a gain of ~ 1 cm^{-1} at a sodium atom concentration $\sim 10^{16}$ cm^{-3} and at a pump intensity $\sim 10^7$ W/cm^2.

PACS numbers: 32.80.Kf

1. INTRODUCTION

The changes in the absorption and emission line shapes of a two-level system under intense resonant excitation have by now been investigated in sufficient detail.[1–11] The usual Lorentz shape of the absorption and emission lines is the consequence of the exponential law governing the damping of the upper and lower states of the system. The change in the kinetics in the damping of the states of an atom subjected to intense excitation is the reason for the change in the atomic emission and absorption line shapes.

The most interesting phenomenon, first predicted theoretically,[1] is the essential difference between the changes in the line shapes of the spontaneous emission and absorption of auxiliary weak radiation. In the case of intense resonant excitation, the probability of the transition of an atom to an upper state comes close to the stationary value, oscillating about this value at a frequency $dE/\hbar$ (d is the matrix element of the dipole

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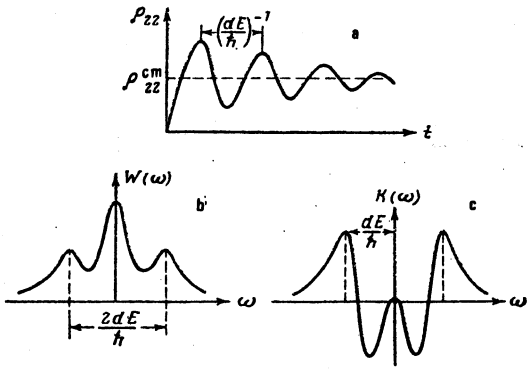

FIG. 1. Population probability ρ_{22} of level 2 as a function of the time (a). Frequency dependence of the probability of spontaneous emission in a strong resonant radiation field (b). Absorption line shape under the same conditions (c).

moment of the transition, E is the amplitude of the exciting-radiation field) (Fig. 1a). A consequence of these oscillations is the splitting of the lines of the spontaneous emission into three components (Fig. 1b), which has been reliably confirmed in experiment.[8,9]

The shape of the absorption line of the sounding radiation is much more complicated (Fig. 1c). The spectral dependence of the absorption coefficient does not coincide with the dependence of the spectral density of the spontaneous emission. The absorption coefficient even changes sign in a frequency interval on the order of $dE/\hbar$ near the frequency of the initial absorption line. The predicted change in the absorption spectrum was also confirmed experimentally.[10] In addition, it was established[11] that a change in the sign of the absorption coefficient takes place also in the case of intense excitation of atoms by noise radiation; such a change is, approximately, the change of multimode lasers without mode locking. The intensity of the exciting radiation should be sufficient to produce short-duration inverted level populations in the course of the establishment of the stationary level values.

In view of the fundamental importance of the change in the relation between the spectral shapes of the emission and absorption lines in intense radiation fields, it is of interest to investigate the analogous lines-shape changes also for nonresonant intense multiphoton excitation. The possibility of splitting of the spontaneous-emission line into three components under intense two-photon excitation of a two-level system was predicted theoretically.[12] The condition for this splitting, as expected, is a sufficiently high probability of two-photon excitation, ensuring periodic inversions of the level populations in the course of establishment of their stationary values.

The present paper is devoted to an experimental and theoretical investigation of the absorption line shape of sounding radiation of a two-level system under intense two-photon excitation. It is established that at sufficiently high excitation intensity the line shape undergoes changes similar to those occuring under resonant (single-photon) excitation. It is shown that the reversal of the sign of the absorption coefficient (i.e., amplification) under multiphoton excitation of atoms can be used to increase effectively the emission frequency of a laser (in particular, for optical third-harmonic generation).

2. EXPERIMENT

The object of the investigation was ^{113}Cd vapor, which has in the ground state only a nuclear spin 1/2. In a constant magnetic field, the ground level splits into two with a transition frequency (ω_{21} = 3 kHz in a field ~3G), which is less by many orders of magnitude than the frequencies of the transitions to the excited levels; this makes it possible to regard such a system as a two-level one. We investigated the spectrum of the absorption of weak sounding radiation $H_y = H\cos\omega t$ by this two-level system when the atoms are acted upon by intense radiation of frequency ω_0 close to $\omega_{21}/2$ (Fig. 2). The weak sounding field H was perpendicular to the constant field H_z directed along the Z axis. The strong field $\mathbf{H}_0(t)$ consisted of two components:

$$\mathbf{H}_0(t) = H_0(\mathbf{e}_x\cos\omega_0 t + \mathbf{e}_y\sin\omega_0 t) + H_1\mathbf{e}_z\cos\omega_0 t\ ,$$

one rotating about the Z axis and one linearly oscillating along the same axis. By virtue of the selection rule for the projection of the magnetic quantum $\Delta m_z = 1$, two-photon excitation of atoms took place only when both strong-field components acted simultaneously. The experimental setup was described in detail previously.[10]

The main features in the change of the absorption spectrum of the sounding radiation turned out to be the same as in intense resonant excitation.[10] The most substantial distinguishing feature of the observed spectrum is the strong dependence of the absorption line shape on the accuracy with which the doubled frequency of the strong field $2\omega_0$ coincides with the frequency of the Zeeman transition between the perturbed levels. The latter does not coincide with the frequency ω_{21} of the transition in the absence of a strong field, but is shifted relative to it by an amount equal to the Bloch-Siegert shift Δ_0. Figure 3 shows the measured absorption spectra of the sounding radiation in the case of exact resonance (Fig. 3a) and in the case of deviation from resonance (Fig. 3b).

3. THEORY

The problem of finding the absorption spectrum reduces to solving an equation for the density matrix, which takes in the interaction representation the form

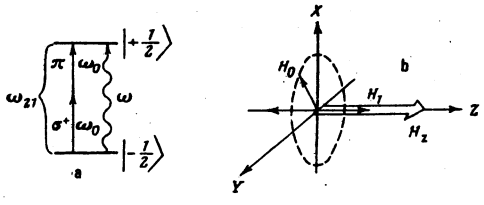

FIG. 2. Level scheme (a) and arrangement of the fields (b) in the experiment.

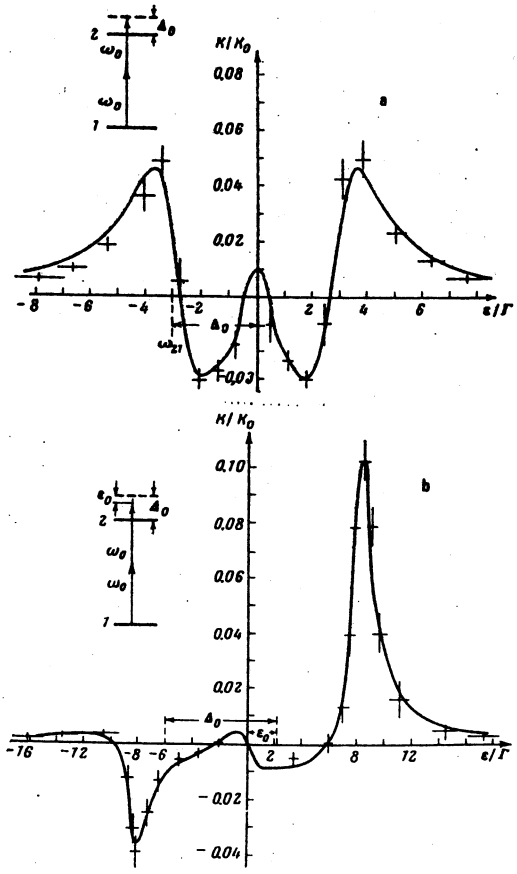

FIG. 3. Dependence of the absorption coefficient on the frequency: a—at $\varepsilon_0=0$ and $\Delta_0=3\Gamma$; b—at $\varepsilon_0=2.07\Gamma$, $\Delta_0=8\Gamma$; $\Gamma=2.7$Hz. Solid curves—theory, crosses—experiment.

$$\begin{aligned} i\dot{f}_{22}&=V_{21}f_{12}-f_{21}V_{12}-i\Gamma_1 f_{22},\\ i\dot{f}_{12}&=-V_{21}^*(f_{11}-f_{22})-i\Gamma_2 f_{12}+f_{12}V_{11}(e^{-i\omega_0 t}+e^{i\omega_0 t}),\\ f_{11}+f_{22}&=1,\quad f_{12}=f_{21}^*. \end{aligned} \tag{1}$$

Here

$$V_{21}=V_0e^{i\omega_1 t}+ve^{i\tilde{\omega}t}=V_{12}^*,\quad V_0=-\pi\gamma H_0,\quad V_{11}=\pi\gamma H_1,\quad v=-\pi\gamma H,$$
$$\gamma=942\ \text{Hz/G},\quad \omega_1=\omega_{21}-\omega_0,\quad \tilde{\omega}=\omega_{21}-\omega,$$

Γ_1 and Γ_2 are the frequencies of the longitudinal and transverse relaxation.

We solve the system (1) subject to satisfaction of the experimentally realized conditions

$$v\ll V_0, V_{11}, \tag{2}$$

$$V_0^2/\omega_1^2\ll 1, \tag{3}$$

$$|2\omega_0-\omega_{21}|\ll\omega_{21}. \tag{4}$$

We seek the solution of (1) in the form

$$f_{ik}=f_{ik}^{(0)}+\varphi_{ik}, \tag{5}$$

where $f_{ik}^{(0)}$ is the solution of the system (1) in the absence of a weak field ($v=0$), and φ_{ik} is a small correction due to the weak field.

The two-photon resonance condition (4) allows us to seek an approximate solution of the system (1) in the absence of a weak field in the form[1)]

$$f_{12}=X_0+X_1e^{i\omega_1 t}+X_{-1}e^{-i\omega_1 t}, \tag{6}$$
$$f_{22}=Z_0+Z_1e^{i\omega_1 t}+Z_{-1}e^{-i\omega_1 t}. \tag{7}$$

Substitution of (6) and (7) in (1) at $v=0$ and retention of only the constant and slowly varying terms with frequency $\delta=\omega_{21}-2\omega_0$ leads to the following system of equations for X_0 and Z_0:

$$\begin{aligned} i\dot{X}_0&=-(\Delta_0+i\Gamma_2)X_0-V_{11}V_0e^{-i\delta t}/\omega_1+2V_0V_{11}e^{-i\delta t}Z_0/\omega_1,\\ i\dot{Z}_0&=-i\Gamma_1Z_0+V_0V_{11}e^{i\delta t}X_0/\omega_1-V_{11}V_0e^{-i\delta t}X_0^*/\omega_1, \end{aligned} \tag{8}$$

where $\Delta_0=2V_0^2/\omega_1$ is the dynamic shift of the transition frequency under the influence of the strong field.

Substitution of $X_0=\rho_{st}e^{-i\delta t}$ transforms the system (8) into a system of linear equations with constant coefficients:

$$\begin{aligned} i\dot{\rho}_{st}+\rho_{st}(\delta+\Delta_0+i\Gamma_2)-2V_0V_{11}Z_0/\omega_1&=V_{11}V_0/\omega_1,\\ i\dot{Z}_0+i\Gamma_1Z_0-V_0V_{11}(\rho_{st}-\rho_{st}^*)/\omega_1&=0. \end{aligned} \tag{9}$$

The stationary solution of the system (9) is easy to obtain by putting $\dot{\rho}_{st}=\dot{Z}_0=0$. This yields the sought changes of the constant population component Z_0 and of the component X_0 that is at resonance with the atom transition frequency:

$$Z_0=\frac{1}{2}\frac{K_2^2\Gamma_2/\Gamma_1}{\varepsilon_0^2+\Gamma_2^2+\Gamma_2K_2^2/\Gamma_1}, \tag{10}$$

$$X_0=\frac{V_{11}V_0}{\omega_1}\frac{\varepsilon_0-i\Gamma_2}{\varepsilon_0^2+\Gamma_2^2+\Gamma_2K_2^2/\Gamma_1}e^{-i\delta t}=\rho_{st}e^{-i\delta t}, \tag{11}$$

$$\rho_{st}=\frac{V_{11}V_0}{\omega_1}\frac{\varepsilon_0-i\Gamma_2}{\varepsilon_0^2+\Gamma_2^2+\Gamma_2K_2^2/\Gamma_1}. \tag{12}$$

Here

$$\varepsilon_0=\delta+\Delta_0,\quad K_2=2V_0V_{11}/\omega_1.$$

It is precisely these values of Z_0 and X_0 which determine the change of the absorption line shape of the sounding radiation under the action of a resonant strong field. Expressions (10) and (11) differ from the corresponding expressions for Z_0 and X_0 under resonant action of a strong field in that the matrix element V_0 of the single-photon transition is replaced by the matrix element K_2 of the two-photon transition (see[3,5]).

Using the obtained solutions

$$f_{22}^{(0)}\approx Z_0,\quad f_{12}^{(0)}\approx X_0=\rho_{st}e^{-i\delta t}$$

and substituting (5) in (1), we obtain, neglecting second-order terms ($v\varphi_{12}$ and $v\varphi_{22}$), the following system of equations for the corrections φ_{22} and φ_{12}:

$$\begin{aligned} i\dot{\varphi}_{22}&=V_0e^{i\omega_1 t}\varphi_{12}+ve^{i\Delta t}f_{12}^{(0)}-V_0e^{-i\omega_1 t}\varphi_{21}-ve^{-i\Delta t}f_{21}^{(0)}-i\Gamma_1\varphi_{22},\\ i\dot{\varphi}_{12}&=2v_0e^{-i\omega_1 t}\varphi_{22}+ve^{-i\Delta t}(2f_{22}^{(0)}-1)+V_{11}(e^{-i\omega_0 t}+e^{i\omega_0 t})\varphi_{12}-i\Gamma_2\varphi_{12}. \end{aligned} \tag{13}$$

Here $\Delta=\omega_{21}-\omega$.

To find the polarization of the system at the frequency ω of the sounding field, the following approximate form of the solution is sufficient:[3,5]

$$\begin{aligned} \varphi_{22}&=A_0+A_1e^{i\omega_1 t}+A_{-1}e^{-i\omega_1 t},\\ \varphi_{12}&=B_0+B_1e^{i\omega_1 t}+B_{-1}e^{-i\omega_1 t}. \end{aligned} \tag{14}$$

Substitution of (14) in (13), after neglecting the rapidly oscillating terms, leads to a system of equations for A_0 and B_0:

$$\begin{aligned} iA_0 &= V_{11}V_0B_0e^{i\delta t}/\omega_1+\rho_{st}ve^{-i\varepsilon t}-V_{11}V_0B_0^*e^{-i\delta t}/\omega_1-\rho_{st}^*ve^{i\varepsilon t}-i\Gamma_1A_0, \\ i\dot{B}_0 &= -(\Delta_0+i\Gamma_2)B_0-vq_{st}e^{-i\Delta t}+2V_{11}V_0A_0e^{-i\delta t}/\omega_1, \end{aligned} \tag{15}$$

where $\epsilon = \omega - 2\omega_0$ and $q_{st} = 1 - 2Z_0$.

Introducing the substitution $B_0 = \bar{\rho}e^{-i\delta t}$ and changing over to the correction for the change of the population difference $\bar{q} = 2A_0$, we can reduce the system (15) to the form

$$\begin{aligned} \dot{\bar{q}} &= -2\mathrm{Re}\{iK_2\bar{\rho}\}-\Gamma_1\bar{q}-4\mathrm{Re}\{ve^{-i\varepsilon t}\rho_{st}\}, \\ \dot{\bar{\rho}} &= -i/_2K_2\bar{q}-(\Gamma_2-i\varepsilon_0)\bar{\rho}+ve^{i\varepsilon t}q_{st}. \end{aligned} \tag{16}$$

The system (16) differs from the corresponding system of equations for the single-photon resonant interaction (Eqs. (3) and (4) of[5]) only in the symbols used for the constant coefficients of the equation. Since a complete analysis of the stationary solution of the system (16) has already been made[5] and has yielded for the nonlinear susceptibility of the atomic system expressions that determine the absorption coefficient and the refractive index of an ensemble of atoms, we present directly the expression for the absorption coefficients of the sounding radiation:

$$K(\omega) = K_0q_{st}\,\mathrm{Re}\frac{(\Gamma+i\varepsilon)(\Gamma+i\varepsilon_0)(\Gamma+i\varepsilon+i\varepsilon_0)-i\varepsilon\Delta_0V_{11}^2/(\omega_{21}-\omega_0)}{(\Gamma+i\varepsilon_0)\mathscr{L}(i\varepsilon)},$$

$$\mathscr{L}(i\varepsilon) = -i\varepsilon^3-3\varepsilon^2\Gamma+i\varepsilon\left(\varepsilon_0^2+\frac{2\Delta_0V_{11}^2}{\omega_{21}-\omega_0}+3\Gamma^2\right)+\Gamma(\varepsilon_0^2+\Gamma^2)+\Gamma^2\frac{2\Delta_0V_{11}}{\omega_{21}-\omega_0}. \tag{17}$$

Here K_0 is the absorption coefficient at the line center in the absence of a strong field, q_0 is the initial population difference of the lower and upper levels, $\Gamma = \Gamma_1 = \Gamma_2$, and $\epsilon_0 = \omega_{21} - 2\omega_0 + \Delta_0$.

4. DISCUSSION OF RESULTS

The absorption line shape calculated from formula (17) agrees within the limits of measurement accuracy with that observed in experiment (Fig. 3). The change in the line shape is similar to that which occurs under resonant or quasiresonant intense excitation. The differences consist of replacement of $\epsilon_0' = \omega_{21} - \omega_0$ by $\epsilon_0 = \omega_{21} - 2\omega_0 + \Delta_0$ and the matrix element of the single-photon transition by the matrix element $K_2 = 2V_0V_{11}/\omega_1$ of the two-photon transition. A spectrum shape that is symmetrical with respect to the doubled frequency $2\omega_0$ of the strong field is obtained when the doubled frequency of the strong field coincides with the frequency of the transition with account taken of the dynamic shift of the levels by the strong field (Fig. 3a).

The reversal of the sign of the absorption coefficient (i.e., that amplification) under multiphoton excitation can be used to develop amplifiers of optical radiation and for the conversion of laser frequencies. Compared with resonant pumping, the use of multiphoton transitions offers two advantages. The first is the possibility of tuning the frequency of the gain (or lasing) line over a wider range. In the case of resonant pumping, this range is severely restricted by the equalization of the level population with increasing pump power and with decreasing gain.[10] In two-photon pumping, the amplification frequency can be tuned by dynamically shifting of the levels by one of the exciting strong fields. The attainable degree of equalization of the populations is ensured by a corresponding decrease of the power of the second field. From the result of the experiment and formula (17) it follows that under optimal conditions the gain can reach 5% of the initial linear-absorption coefficient.

The second advantage of multiphoton pumping is the possibility of obtaining generation in a frequency region greatly differing from the pump frequency or summary or difference frequencies. For atoms, greatest interest attaches to three-photon excitation and lasing at a frequency close to the frequency of the third optical harmonic.

We have estimated the efficiency of frequency tripling of ir radiation in sodium vapor. Three-photon excitation in sodium vapor from the state 3*S* into 7*P* can be produced by radiation of wavelength 7784Å (Fig. 4). As a consequence of the two-photon resonance for the transition 3*S*—4*S*, the required pump power is decreased. The threshold pump power needed to attain gain near the third-harminic frequency ($\lambda = 2595$Å) corresponds to equality of the rate of three-photon excitation and the atom-relaxation rate Γ:

$$\frac{d_{12}E}{2\hbar}\frac{d_{23}E}{2\hbar}\frac{d_{34}E}{2\hbar}\frac{1}{(\omega_{21}-\omega_0)(\omega_{31}-2\omega_0)} = \Gamma. \tag{18}$$

At high pressures, the value of Γ is determined by the impact broadening of the line of the 7*P*—3*S* transition and is of the order of $10^6\,\mathrm{sec}^{-1}$ at a sodium atom concentration on the order of $10^{16}\,\mathrm{cm}^{-3}$.[14] According the data on the oscillator strengths for Na,[15] condition (18) is satisfied at a pump intensity on the order of $10^7\,\mathrm{W/cm}^2$.

The attained gain, under optimal conditions, is equal to $1\,\mathrm{cm}^{-1}$ at an Na atom concentration $\sim 10^{16}\,\mathrm{cm}^{-3}$. It appears that the greatest difficulties for the practical realization of this method of generating optical harmonics are posed by the need for obtaining monochromatic pump radiation. The use of multimode dye lasers for pumping leads to broadening of the gain lines, to an

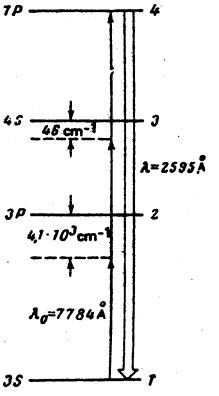

FIG. 4. Scheme of the NaI levels participating in three-photon excitation.

increase of the threshold pump powers, and to a decrease of the gain. We are now planning experiments aimed at determining the requirements that the monochromaticity of the pump radiation must satisfy to obtain optical amplifiers and laser-frequency convertors.

The authors thank S. G. Przhibel'skiĭ for valuable theoretical consultations and A. M. Bonch-Bruevich for interest in the work.

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Translated by G. Adashko

Dissociative recombination of electrons on the molecular ions H_2^+ and D_2^+ with production of strongly excited atoms

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The process of dissociative recombination with production of strongly excited atoms in collisions between electrons of energy 1–13 eV and molecular ions is considered. The recombination cross sections are calculated for the system $e+H_2^+$ (D_2^+) with formation of excited atoms in states with $n \geq 3$ for different vibrational states of H_2^+ (D_2^+). The calculation results are compared with the available experimental measurements. The transition from the quantum description of the motion of the nuclei in dissociative recombination to the classical description is indicated.

PACS numbers: 34.80.Gs

1. INTRODUCTION

One of the important elementary processes that lead to the decay of the electron density in recombining discharges and to the vanishing of electrons and ions from the upper layers of the atmosphere is the dissociative recombination (DR) process

$$e+AB^+ \to AB^* \to A+B^*,$$

in which collision of the electron with the molecular ion produces an autoionization state AB^* with a repulsion term, nuclear motion over which leads to formation of recombination products.

The rate coefficients of DR at thermal energies of the electrons were measured in recombining discharges for many ions (see the reviews[1,2]). For the simplest ions H_2^+ and D_2^+ there was no experimental information on the DR cross sections until recently, because of the rapid formation of the H_3^+ (D_3^+) and H_5^+ (D_5^+) at relatively high pressures of the recombining discharges. Only in recent experiments with crossed beams[3-5] were DR cross sections measured at relative energies on the order of several electron volts, namely, the total DR cross section for collisions with H_2^+ and D_2^+ was measured at energies 0.6—7 eV, in[3] and the DR cross section with production of $D(n=4)$ was measured at energies 1.4—7.5 eV, in[4] and the cross section for recombination with production of $D(n=2)$ was measured at energies 1.4—7.5 eV in[5].

The theoretical papers[6-9] devoted to the description of DR in collisions with H_2^+ and D_2^+ are restricted mainly to slow (up to 1 eV) electrons and to the case when the repulsion term of the autoionization state crosses the ground vibrational state of the molecular ion. The results of these studies cannot be used to describe DR with production of strongly excited ($n \geq 3$) states of hydrogen or deuterium. Thus, the first estimates of the DR cross sections for the system $e+H_2^+$ and energies < 1 eV via production of the autoionization states $H_2^*(2p\sigma_u, 2s\sigma_g, {}^3\Sigma_u)$ and $H_2^*(2p\sigma_u, 2p\pi_u, {}^3\Pi_g)$ were carried out by Bauer and Wu[6] and were repeated with slight modifications by

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