Experimental observation of Landau-Zener nonlinearity in optical excitation of atoms

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Nonlinear induced transparency of the wing of the atomic line of thallium was observed in the presence of argon. It is shown that at high excitation powers (10^9 W/cm^2) the degree of population of the atomic system decreases with increasing pump intensity (nonlinearity of the Landau-Zener type). The effect was revealed both by the dependence of the integral emission of the vapor as a function of the excitation power and by the change of the kinetics of the atomic fluorescence.

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The investigation of the quasistatic far wings of atomic lines is a traditional problem of practical importance in atomic spectroscopy, in view of the connection of the line shapes with the potentials of the atomic interactions.[1] In a number of cases this connection makes it possible to determine these potentials by measuring the light absorption or emission in collisions between the atoms.[2] With the advent of powerful optical-radiation sources, interest has arisen in the study of the effect of the radiation field on the collision process. Since the resonance between the radiation field and the atomic-transition frequency that varies during the collision is of short duration (~10^{-13} sec), rather high powers are needed to produce a change of any size in the population of the atomic states.

The nonlinear theory of the quasistatic wing of the atomic line was developed only recently.[3-7] The most fruitful approach to this problem was developed in Ref. 3–6 and consists of reducing the problem of absorption (or emission) of a photon of energy $\hbar\omega$ during the collision to the problem of the stationary theory of atomic collisions, i.e., to the problem of non-adiabatic atomic transitions between intersecting quasi-energy levels of the system $U_1(R)+\hbar\omega$ and $U_2(R)$, where the subscripts 1 and 2 correspond to the ground and excited terms of the quasimolecule. The matrix element of the interaction that produces the transition between the quasi-energy levels is the product of the dipole moment of the electronic transition by the amplitude of the electric field intensity of the light wave, $V=\mathbf{d}\cdot\mathbf{E}$.

It should be noted that the problem of optical excitation of a system of colliding atoms has two features that distinguishes it from the problems of ordinary theory of atomic collisions. First, interest attaches here to the dependence of the transition probability on the radiation intensity, whereas in the theory of atomic collisions the corresponding characteristic—the energy of the nonadiabatic coupling of the terms—is a specified quantity, and what is being investigated is the dependence of the transition probability on the relative velocity of the colliding particles. Second, it is assumed that the interaction V does not depend on the distance, and this leads to a great variety in the initial conditions, which are determined by the character of the relaxations, by the law that governs the application of the field, and others.[5]

The probability of observing atoms in an excited state after their flight is given by the Landau-Zener formula[8]

$$w=2e^{-\delta}(1-e^{-\delta}), \tag{1}$$

where $\delta=2\pi V^2/\hbar v\Delta F$, ΔF is the difference between the slopes of the terms at the point R_0 at which the energy difference between the terms $U_2(R_0)-U_1(R_0)=\hbar\omega$, where $\hbar\omega$ is the energy of exciting-field quantum and v is the velocity of the atoms at this point. This form of the dependence is due to the double passage of the atom (on entering and leaving the interaction region) through the term "quasi-intersection" point $R=R_0$.

It is easily seen that the dependence (1) on the excitation intensity is nonmonotonic: the transition probability is small both at low intensities [$V\ll V_{cr}=(\hbar v\Delta F)^{1/2}$], and at high ones ($V\gg V_{cr}$). Just as in the usual theory of atomic collisions, the decrease of the probability of the transition is due to the mutual repulsion of the terms between which the transition takes place. In this case this repulsion is produced by the radiation field itself (the dynamic Stark effect). As a result, at some excitation power $I>I_{cr}$ the medium becomes transparent to the radiation. We note that in ordinary absorption saturation in a two-level system the populations of the corresponding levels become equalized, and the decrease of the absorption coefficient of the medium is due to a trivial decrease of the ratio of the absorbed and incident energies; the absorbed energy itself stays constant and is determined by the rate of the spontaneous relaxation and by the number of atoms in the system. In our case we encounter a new nonlinear effect: when a certain critical excitation power is exceeded the medium not only stops to absorb, but begins to luminesce,[4] so that there are no losses at all.

The physical nature of this phenomenon is nevertheless not clear enough: two passes through the resonance region take place for each collision of the atoms with the broadening particle. In the first pass the atom is excited with near-unity probability, and in the second it goes to the ground state with the same probability.

Although the theoretically predicted[4] field values at which the nonlinear induced transparency of the medium should appear (10^5–10^8W/cm^2) is relatively low and can be produced by modern lasers, there has been to our knowledge no experimental confirmation. This is due, first, to the complexity of experiments of this kind, and second, to the difficulty of choosing a real system for

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its observation. In fact, a more detailed examination of the process of nonresonant excitation of atoms[4,5] leads to the conclusion that induced transparentization of the medium is realized only in a limited power interval of the exciting radiation field $V_{cr} \ll V \ll \hbar(\omega - \omega_0)$, where ω_0 is the transition frequency of the unperturbed atom and ω is the excitation frequency. The inequality on the right side corresponds to the requirement that the atom not be excited in the intervals between the collisions (the process of "three-photon excitation" of the atomic line[9]). This means that the experiment must be carried out at sufficiently large detuning from resonance. To satisfy the inequality on the left side, the system must have slowly varying terms $[V_{cr} \sim (\Delta F)^{1/2}]$. It is readily seen that in practice these requirements are difficult to reconcile.

Another difficulty is the slow decrease of the dependence of the system population on the excitation power at $V > V_{cr}$. In fact, the relation (1) must be averaged over the collision impact parameters, over the relative velocities of the atoms, and over the orientation of the internuclear axis relative to the direction of the electric field intensity of the light-wave field. The last factor is the most important, since it alters drastically the character of the excitation fall-off.[5,6] If the dipole moment of the transition is oriented perpendicular to the internuclear axis, then the probability of excitation of the system decreases in inverse proportion to the intensity of the exciting radiation; on the other hand if it is oriented along the axis, the intensity falls off even more slowly, $\sim I^{-1/2}$.

It must be borne in mind here that the averaging of such a slowly decreasing dependence over the temporal and spatial distributions of the exciting radiation masks the effect completely even when the two distributions are Gaussian. Thus, the effect in question can be observed only by acting on the system with a radiation that is steeper than Gaussian both in time and in space.

The object chosen for the present investigation was the system Tl + Ar, whose term scheme is shown in Fig. 1a.[10] The emission spectrum of the atomic line (transition $7S_{1/2} - 6P_{3/2}$, $\lambda = 535$ nm) of this system (Fig. 1b) contains a narrow satellite ($\lambda_s = 530$ nm), i.e., the point

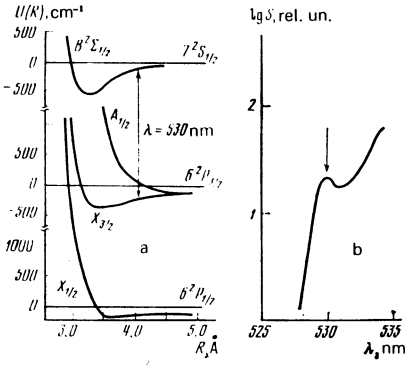


FIG. 1. Term scheme (a) and emission spectrum (b) of the Tl + Ar system in the vicinity of the atomic transition $7S_{1/2} - 6P_{3/2}$ of thallium.[10] The position of the satellite is indicated by the arrows.

where the terms $X_{3/2}$ and $B^2\Sigma_{1/2}$ are parallel.[10] It is known[11,12] that when the terms are parallel the critical saturation power of the transition is minimal and is determined by the spectral width of the satellite Δ_s: $V_{cr} \approx \Delta_s \approx (\hbar^2 v^2 k)^{1/3}$, where k is the coefficient of the quadratic term in the expansion $\hbar\omega + U_1(R) - U_2(R) = \Delta - k(R - R_0)^2$ (Δ is the detuning away from the center of the satellite). The presence of such a singularity in the system expands greatly the interval of the powers at which the induced-transparency effect is observable.

The investigated system was excited by the second harmonic of a mode-locking neodymium laser. This regime was chosen both to increase the peak power of the radiation and to decrease the number of atoms excited during the time between collisions by the mechanism of "three-photon excitation," which is proportional to $\Delta t \gamma_1$, where Δt is the duration of the exciting pulse and γ_1 is the rate of the longitudinal relaxation of the excited state.

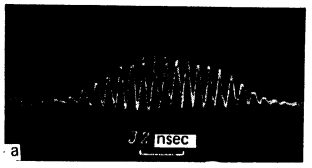
To tune the frequency of the exciting radiation to the center of the satellite, a polarization selector (quartz-crystal plate) was placed in the laser resonator. The laser pulse consisted of 15–18 individual spikes whose duration was measured by the two-photon luminescence method and amounted to $(10–15)\times10^{-12}$ sec. The radiation power was increased with an amplifier, and its frequency was doubled with an $LiNbO_3$ crystal.

The argon pressure was chosen such that during the excitation time ($\sim10^{-10}$ sec) the probability of collision of the thallium and argon atoms was close to unity ($P_{Ar} \sim 1$ atm). For a noticeable population of the initial atomic level $6P_{3/2}$ the cell with the vapor was heated to 830–880° C, in which case the concentration of the atoms on the metastable level $6P_{3/2}$ was $\sim10^{12}$ cm^{-3}.

The population of the excited $7S_{1/2}$ state of the thallium atom was determined by registering the intensity of the luminescence on the transitions $7S_{1/2} - 6P_{1/2}$ ($\lambda = 377$ nm) or $7S_{1/2} - 6P_{3/2}$ ($\lambda = 535$ nm), separated with an MDR-2 monochromator.

Particular attention was paid to the formation of the temporal and spatial distributions of the exciting beam. The oscillogram of the envelope of the pulse and its spatial distribution in the interaction region are shown in Fig. 2. The deviation of the temporal profile of the pulse from Gaussian was due to generation of the second-harmonic of the radiation at a large conversion coefficient.[13] The geometric profile of the beam was shaped by spatial filtering of the radiation[14] and with the aid of a 0.4-mm diaphragm ahead of the cell with the vapor (the diameter of the beam in front of the diaphragm was 0.8 mm). The luminescence was registered in the near field of the beam, where the diffraction phenomena are still insignificant.

The dependence of the atomic emission, measured under the described conditions and integrated over the pulse, on the excitation power is shown in Fig. 3. Each point on this plot is the average of a signals obtained with 12 laser flashes. The maximum experimental scatter is shown in the figure. It is easily seen that the observed decrease of the intensity of the emission at a



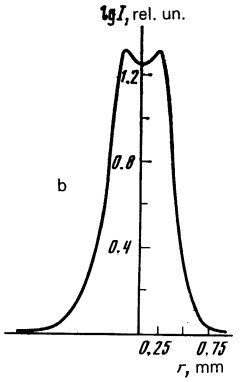


FIG. 2. Oscillogram of radiation pulse (a) and spatial distribution (b) of the beam in the excitation region.

power $I > 10^9$ W/cm^2 exceeds this value. It should be noted here that the insufficient stability of the regime of passive synchronization of the laser mode and of the pulse parameter in the excitation volume has led to a considerable distortion of the obtained relations (Fig. 3 shows the best of them) at any albeit slight readjustment of the setup. Much more reproducible and sensitive was the method of recording this dependence by measuring the kinetics of the emission of the thallium. In fact, the interval between the individual picosecond pulses in the train was 7 nsec, while the lifetime of the excited state (in the absence of dragging) was 7.5 nsec,[15] so that most of the atoms excited during the time of action of one pulse managed to emit in the intervals between them. This makes it possible to regard the luminescence signal produced after each picosecond pulse as the result of a single-pulse excitation, while measurement of the peak intensity of the emission of the atoms corresponding to different train pulses having different laser-pulse amplitudes make it possible to assess the dependence of the population of the system on the excitation power.

Oscillograms of the thallium-vapor fluorescence pulses at three different excitation intensities are shown in Fig. 4. Despite the relatively large resolution time of the recording apparatus (15–20 nsec), the last of the pulses shows clearly a dip at the center, where the intensity of the exciting picosecond pulses is maximal. It is easily seen that qualitatively the shape of this pulse

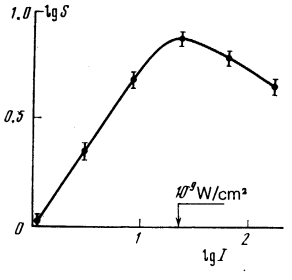


FIG. 3. Dependence of the integral emission of an atomic line of thallium (transition $7S_{1/2} - 6P_{1/2}$) on the excitation power.

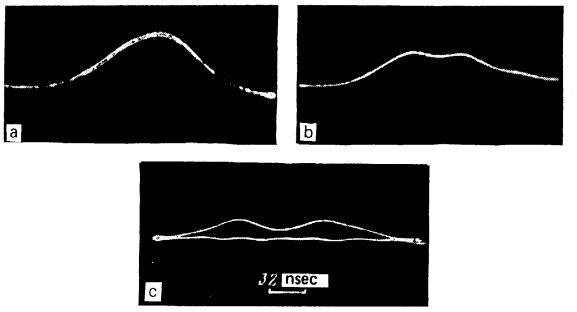


FIG. 4. Oscillograms of thallium-vapor emission pulses at various excitation powers: a) $I = 3 \times 10^8$ W/cm^2, b) $I = 2 \times 10^9$ W/cm^2, c) $I = 8 \times 10^9$ W/cm^2.

agrees with the form of the plot shown in Fig. 3, but unfortunately it is difficult to extract quantitative information from this experiment in view of the almost total lack of information on the temporal envelope of an individual picosecond pulse.[1)] This, as well as the spatial integration that is inevitable in any experiment, does not make it possible to determine with sufficient accuracy the law governing the decrease of the intensity, nor the absolute value of the power at which the transition probability is close to unity. At the same time, this result should be regarded as independent and direct proof of the realization, in the investigated system, of the above-discussed phenomenon of induced transparency in the medium without saturating the latter.[3-6]

We note that the presented estimates and an experimental check have shown that the processes of multiphoton ionization of atoms and of Raman scattering of light on electronic levels of the thallium atom make no noticeable contribution to the recorded emission of the atoms.

To our knowledge, these data represent the first experimental observation of nonlinearity of the Landau-Zener type in optical excitation of atoms. In our opinion, the investigated effect is of independent interest. In a certain sense it is an analog of the self-induced transparency observed under resonance conditions, but in contrast to the latter the condition of radiation propagation do not depend in this case on the ratio of the amplitude of the pulse to its duration, and are determined only by its intensity.

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[1)]According to the data of V. B. Shilov and co-workers (private communication from V. B. Shilov), obtained with the "Agat" high-speed chronograph of VNIIOFI (Res. Inst. for Opt. Phys. Meas.), the decrease of the radiation intensity in each individual laser spike with mode locking is faster (to the 0.1 level) than Gaussian. The fronts of this pulse can become even steeper in the course of generation of the second harmonic of the generation.

[1]S. Chen and M. Takeo, Rev. Mod. Phys. 29, 20 (1957).
[2]G. York, R. Scheps, and A. Gallagher, J. Chem. Phys. 63,

1052 (1975).
[3]L. I. Gudzenko and S. I. Yakovlenko, Zh. Eksp. Teor. Fiz. 62, 1686 (1972) [Sov. Phys. JETP 35, 877 (1972)].
[4]V. S. Lisitsa and S. I. Yakovlenko, Zh. Eksp. Teor. Fiz. 66, 1550 (1974) [Sov. Phys. JETP 39, 759 (1974)].
[5]S. I. Yakovlenko, Kvantovaya Elektron. (Moscow) 5, 259 (1978) [Sov. J. Quantum Electron. 8, 151 (1978)].
[6]R. Z. Vitlina, A. V. Chaplik, and M. V. Entin, Zh. Eksp. Teor. Fiz. 67, 1667 (1973) [Sov. Phys. JETP 40, 829 (1973)].
[7]E. G. Pestov and S. G. Rautian, Zh. Eksp. Teor. Fiz. 64, 2032 (1973) [Sov. Phys. JETP 37, 1025 (1973)].
[8]L. D. Landau and E. M. Lifshitz, Kvantovaya mekhanika (Quantum Mechanics), Fizmatgiz, 1963 [Pergamon, 1968].
[9]P. A. Apanasevich, Osnovy vzaimodeĭstviya sveta s veschestvom (Principles of Interaction of Light with Matter), Minsk, 1977.
[10]B. Cheron, R. Scheps, and A. Gallagher, J. Chem. Phys. 65, 326 (1976).
[11]A. M. Bonch-Bruevich, S. G. Przhibel'skiĭ, and V. V. Khromov, Izv. Akad. Nauk SSSR Ser. Fiz. 43, No. 2, 397 (1979).
[12]T. A. Vartanyan, Yu. N. Maksimov, S. G. Przhibel'skiĭ, and V. V. Khromov, Pis'ma Zh. Eksp. Teor. Fiz. 29, 281 (1979) [JETP Lett. 29, 252 (1979)].
[13]S. B. Arifzhanov, R. A. Ganeev, A. A. Gulamov, V. I. Redkorechev, and T. B. Usmanov. Abstracts of All-Union Conf. on the Problem of Control of Laser-Radiation Parameters, part I, Tashkent, 1978, p. 90.
[14]N. G. Basov, A. R. Zartiskiĭ, S. D. Zakharov, P. G. Kryukov, Yu. A. Matveets, Yu. V. Senatskii, A. I. Fedosimov, and S. V. Chekalin, in: Kvantovaya elektronika (Quantum Electronics), Vol. 6, Sovetskoe radio, 1972, p. 50.
[15]M. D. Havey, L. C. Balling, and G. G. Wright, JOSA 67, 491 (1977).

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Collision anisotropy and impact contour of spectral lines

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An impact theory of spectral-line broadening is constructed for transitions between degenerate states and anisotropic collisions. It is shown that the line contour for the subensemble over atoms with a given velocity consists of a set of Lorentz components that differ in width and in position. The number of components increases with increasing angular momentum of the combining states. The factors that mask the line splitting because of the anisotropy of the collisions, and the results of numerical calculations for Van der Waals and dipole-dipole interactions, are discussed.

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1. INTRODUCTION

It is customarily assumed that the impact contour of the spectral line corresponding to a transition between a pair of isolated levels has a Lorentz shape (see, e.g., Ref. 1). Actually, however, this conclusion of the theory presupposes a spherical symmetry of the average perturbation of the atomic oscillator by the buffer particles, i.e., the latter should be unpolarized (as is practically always the case), have an isotropic velocity distribution, and in addition, their average velocities $\bar{v}_b$ should greatly exceed the average velocities $\bar{v}$ of the radiating particles:

$$\rho_b(\mathbf{v}_b)=\rho_b(|\mathbf{v}_b|);\ \bar{v}_b\gg\bar{v}. \tag{1.1}$$

In other words, when the broadening cross section is averaged the radiating particles should be assumed immobile, while the buffer particles should be assumed to move isotropically. The conditions (1.1) serve as a formulation of the so called isotropic-collision (or isotropic-perturbation) model,[2-6] and if they are violated, the spectral lines have a more complicated structure, which in fact will be dealt with in the present article.

To explain the gist of the matter, we change over to a coordinate system connected with the radiating atom, which moves with velocity $\mathbf{v}$. In this frame a "wind" of buffer particles moving with group velocity $-\mathbf{v}$ blows, as it were, around the radiating atom. It is clear that the perturbation of the wave-emission process will have axial rather than spherical symmetry, and the symmetry axis is collinear with $\mathbf{v}$. For the electronic broadening, the conditions (1.1) are obviously satisfied. In broadening due to collisions with atoms, molecules, and ions, the model of isotropic collisions is adequate for the relatively lighter buffer gas, and its applicability is doubtful in the case of heavy perturbing particles.

The simplest manifestation of the "wind effect" consists in the fact that the impact width and the line shift turn out to depend on the velocity of the radiating atom. This dependence, discussed in the paper of Sobel'man and one of us,[7] and in many succeeding papers,[8-13] is the only consequence of the wind effect in the absence of collisional disorientation. In the opposite case, the line for atoms with a given velocity does not have a Lorentz shape.

The change of the line shape as a result of the anisotropy of the collisions was first established by Kazantsev[14] and was investigated in greater detail by Vdovin and Galitskiĭ.[15] In both papers they considered a tran-

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