23, 26 (1976) [JETP Lett. 23, 22 (1976)].
[6]L. D. Landau and E. M. Lifshitz, Quantum Mechanics, Pergamon Press, Oxford, 1974.
[7]K. T. Hecht, J. Mol. Spectrosc. 5, 355, 390 (1960).
[8]Ya. B. Zel'dovich, Zh. Eksp. Teor. Fiz. 51, 1492 (1966) [Sov. Phys. JETP 24, 1006 (1967)]; Usp. Fiz. Nauk 110, 139 (1973) [Sov. Phys. Usp. 16, 427 (1973)].
[9]V. Ya. Yakubovich and V. M. Strazhinskiĭ, Lineĭnye differentsial'nye uravneniya s periodicheskimi koeffitsientami (Linear Differential Equations With Periodic Coefficients), Nauka, 1972.
[10]V. M. Akulin, S. S. Alimpiev, N. V. Karlov, V. G. Sartakov, and L. A. Shelepin, Zh. Eksp. Teor. Fiz. 71, 454 (1976) [Sov. Phys. JETP 44, 239 (1976)].
[11]A. O. Gel'fond, Ischislenie konechnykh raznosteĭ (The Calculus of Finite Differences), M., 1959.
[12]I. A. Kupnin, Teoriya uprigikh sred s mikrostrukturnoĭ (Theory of Elastic Media With a Microstructure), Nauka, 1975.
[13]R. Feynman and A. R. Hibbs, Quantum Mechanics and Path Integrals, McGraw-Hill, New York, 1965 (Russ. Transl., Mir, M., 1968).
[14]V. S. Letokhov and A. A. Makarov, Coherent Excitation of Molecular Systems in a Strong Quasiresonance Laser Field, Preprint of The Institute of Spectroscopy, Academy of Sciences of the USSR, 1976.

Translated by S. Chomet

Effect of coherent radiation on the translational motion of atoms

A. Yu. Pusep, A. B. Doktorov, and A. I. Burshteĭn

Institute of Chemical Kinetics and Combustion, Siberian Division, USSR Academy of Sciences
(Submitted July 8, 1976)
Zh. Eksp. Teor. Fiz. 72, 98–105 (January 1977)

We consider the action of a traveling monochromatic wave on freely moving atoms having a transition that is at resonance with the light (between levels 1 and 2). A direct solution is obtained for the self-consistent quantum problem of the simultaneous effect of the field on the internal and external (translational) degrees of freedom. It is established that the effect of motion on the periodic variation of the populations of levels 1 and 2 (nutation) reduces to averaging the known solution (without allowance for the motion) over the momentum distribution, the form of which is the same as the initial one but the center of which is shifted by half the momentum of the traveling-wave photon. It is shown that if the momentum distribution is so narrow that the spread of the Doppler shifts is small in comparison with the nutation frequency, then the initial coordinate wave packet is divided by the light into two packets with different velocities, of which one spreads out more slowly than in the absence of the action of light, while the other spreads more rapidly. The conditions under which the spreading of the first packet is anomalously slow are obtained.

PACS numbers: 32.80.–t

1. INTRODUCTION

Monochromatic radiation that is at resonance with one of the atomic transitions causes periodic changes (nutation) in the populations of the levels that are included in the process. The dynamics of this process has been thoroughly investigated[1–4] neglecting the influence of the translational motion of the atom on the nutation and the effect of the nutation on the motion. The presence of translational motion leads to averaging of the nutation process over the distribution of the frequencies of the atomic transitions, which is the result of the Doppler shift. However, the initial momentum distribution, which is responsible for the Doppler broadening, is altered by the interaction, since the light-induced transitions between the resonant terms change in the momentum of the atom. In fact, the form of this distribution can be determined only by directly solving the self-consistent quantum problem of the simultaneous influence of the field on the internal and external (translational) degrees of freedom. The solution of this problem makes it possible not only to determine the influence of the translational motion on the dynamics of the nutation transitions, but also to describe the reaction of these transitions on the translational motion of the atoms.

In addition to the general physical importance of a rigorous solution of the problem of resonant interaction of light with atoms, this interaction is of interest also for some applications of laser optics. We have in mind, in particular, acceleration of atoms by resonant radiation,[5–10] which makes it possible to separate from atomic beams particles that are at resonance with the light field.

We consider in this paper the effect of a traveling monochromatic wave on freely moving atoms that have a transition resonant with the light. It is established that the effect of the motion on the light-induced transitions between two nondegenerate levels reduces to an averaging of the solution of the known nutation problem over the momentum distribution, the form of which is the same as the initial one, but the center is shifted by $\hbar k_0/2$ ($\hbar k_0$ is the momentum of the traveling-wave photon). We obtain the time evolution of the shape of the wave packet. It is shown that in the case of a sufficiently narrow initial momentum distribution, when the Doppler-shift spread due to the momentum uncertainty is small

in comparison with the nutation frequency, the initial coordinate wave packet is divided by the light into two packets. These packets have different velocities and are characterized by different rates of spreading. It is important that under appropriate conditions one of them spreads even less than in the absence of light: its width $\Delta z(t)$ increases much more slowly than $\Delta v t$ (Δv is the quantum uncertainty of the atom velocity.

2. GENERAL FORMALISM

When considering the action of a monochromatic field on an atom, we shall assume the field to be strong enough for its classical description to remain valid. Since there are precedents demonstrating light-induced intra-atomic transitions have a quantum character that makes it necessary to take the quantum effects into acco· also in the translational motion of the atom,[8,10] we shall use a quantum description not only for the internal coordinates of the atom but also for the translational ones. We carry out the analysis in a coordinate system that rotates at the same frequency ω_0 as the field (see, e.g.,[4]). The atom levels $|1\rangle$ and $|2\rangle$, which are coupled by the light-induced transitions, are separated in coordinate systems by a distance $\hbar\Delta_0$, $\Delta_0 = \omega_{21} - \omega_0$, ω_{21} is the transition frequency (Fig. 1). The Hamiltonian $\hat{H}^{(0)}$ of this two-level system (in the momentum representation and with the kinetic energy of the atom taken into account) is given by

$$H_{ii}^{(0)}(\varkappa,\varkappa') = [(\hbar\varkappa)^2/2M \mp \hbar\Delta_0/2]\delta(\varkappa-\varkappa'), \quad i=1,2;$$

$$H_{12}^{(0)} = H_{21}^{(0)} = 0, \tag{1}$$

where $\varkappa$ and $\varkappa'$ denote the momentum of the atom divided by $\hbar$, while M is the mass of the atom. Neglecting spontaneous photon emission, the total Hamiltonian takes the form

$$\hat{H} = \hat{H}^{(0)} + \hat{H}^{int}; \tag{2}$$

$$H_{ii}^{int} = 0, \quad i=1,2, \quad H_{12}^{int}(\varkappa,\varkappa') = H_{21}^{int}(\varkappa',\varkappa) = -\hbar V\delta(\varkappa-\varkappa'+k_0). \tag{3}$$

where $k_0 = \omega_0/c$, and $V = dE_0/\hbar$ (d is the matrix dipole element of the transition, and E_0 is the amplitude of the electric field of the wave).

To solve the time-dependent Schwinger equation we determine first the values and the eigen functions of the Hamiltonian (2):

$$\sum_j \int H_{ij}(\varkappa,\varkappa')c_j(\varkappa')d\varkappa' = \hbar\varepsilon c_i(\varkappa), \tag{4}$$

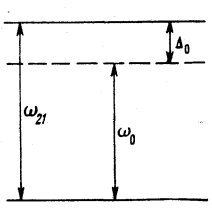

FIG. 1. Energy levels of the atom in a coordinate frame that is at rest and in a frame rotating with frequency ω_0.

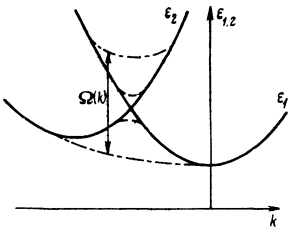

FIG. 2. Dispersion curves $\varepsilon_1(k)$ and $\varepsilon_2(k)$; solid curves—at $V=0$, dashed—at $V<\hbar k_0^2/4M$, dash-dot—at $V>\hbar k_0^2/4M$. Distance $\varepsilon_2(k)-\varepsilon_1(k)=\Omega(k)$.

where $c_i(\varkappa)$ is the probability amplitude of finding the system in the state $|i\rangle$, and with momentum $\hbar\varkappa$ ($\hbar\varepsilon$ is the energy of the stationary state). Taking (1)–(3) into account, Eq. (4) takes the form

$$\begin{aligned} &{}^1/_2(\hbar\varkappa^2/M-\Delta_0)c_1(\varkappa) - Vc_2(\varkappa+k_0) = \varepsilon c_1(\varkappa), \\ &-Vc_1(\varkappa-k_0) + {}^1/_2(\hbar\varkappa^2/M+\Delta_0)c_2(\varkappa) = \varepsilon c_2(\varkappa) \end{aligned}$$

or

$$\begin{aligned} &[\hbar\varkappa^2/2M - \Delta_0/2 - \varepsilon]c_1(\varkappa) - Vc_2(\varkappa+k_0) = 0, \\ &-Vc_1(\varkappa) + [\hbar(\varkappa+k_0)^2/2M + \Delta_0/2 - \varepsilon]c_2(\varkappa+k_0) = 0. \end{aligned} \tag{5}$$

This system has a nonzero solution only at $\varkappa = k$, where k is defined by the dispersion relations

$$\varepsilon = \alpha(k) \mp \Omega(k)/2. \tag{6}$$

Here

$$\alpha(k) = \frac{1}{2}\left[\frac{\hbar k^2}{2M} + \frac{\hbar(k+k_0)^2}{2M}\right], \tag{7}$$

$$\Omega(k) = [4V^2 + \Delta^2(k)]^{1/2}, \tag{8}$$

$$\Delta(k) = \Delta_0 + \frac{\hbar}{M}\left(k + \frac{k_0}{2}\right)k_0. \tag{9}$$

The different signs in (6) pertain to the energy-spectrum branches $\varepsilon_1(k)$ and $\varepsilon_2(k)$ shown in Fig. 2 and corresponding to the lower and upper states of the two-level system. The label $j = 1, 2$ of the state and its wave vector k will henceforth number all the energy eigenvalues and the corresponding eigenfunctions:

$$\psi_{j,k}(\varkappa) = \begin{pmatrix} c_1^{j,k}(\varkappa) \\ c_2^{j,k}(\varkappa) \end{pmatrix} = \begin{pmatrix} [\Omega(k) \pm \Delta(k)]\delta(\varkappa-k) \\ \pm 2V\delta(\varkappa-k-k_0) \end{pmatrix} \tag{10}$$

($j = 1, 2$). Having obtained a stationary basis, we can express in its terms the sought solution of the time-dependent problem in the following manner:

$$\psi(t,\varkappa) = \begin{pmatrix} c_1(t,\varkappa) \\ c_2(t,\varkappa) \end{pmatrix} = \sum_j \int dk\, a_j(k)\psi_{j,k}(\varkappa)\exp\{-i\varepsilon_j(k)t\}, \tag{11}$$

where the expansion coefficients $a_j(k)$ determine the wave-packet form specified by the initial conditions. Substituting the stationary functions (10) in (11) and integrating with respect to k, we can verify that the nutation process does not deform the wave packet in k-space, and connects pairwise the temporal variation of the amplitudes c_1 and c_2, which are displaced relative to each other by k_0:

$$\begin{pmatrix} c_1(t,\varkappa) \\ c_2(t,\varkappa+k_0) \end{pmatrix} = a_1(\varkappa)\begin{pmatrix} \Omega(\varkappa)+\Delta(\varkappa) \\ 2V \end{pmatrix} e^{-i\varepsilon_1(\varkappa)t} + a_2(\varkappa)\begin{pmatrix} \Omega(\varkappa)-\Delta(\varkappa) \\ -2V \end{pmatrix} e^{-i\varepsilon_2(\varkappa)t}. \quad (12)$$

The natural explanation is that when the atom is excited it acquires the momentum of the photon absorbed by it.

The dyad defined in (12) is not a wave function of the system (in view of the relative difference k_0 between the amplitudes, but it is precisely this dyad which should be regarded as the vector of that space in which the energy spin[11] or the representative vector[2,4] is defined with allowance for the motion. Using this important analogy, we can regard the spinors in the right-hand side of (12) as stationary solutions of the traditional Rabi problem of nutation in a system of two levels separated by the kinetic energy of the atom ($T_1 = (\hbar\varkappa)^2/2M$ in the lower state and $T_2 = \hbar^2(\varkappa + k_0)^2/2M$) in the upper state). In the gyroscopic model, such a notation constitutes precession around an effective field[2,3] whose vertical component is corrected for the Doppler frequency shift $\Delta(\varkappa) - \Delta_0 = (T_2 - T_1)/\hbar$ (Fig. 3).

If the atom is on the lower energy level at the initial instant of time, i.e., if it is described by the function

$$\psi(0,k) = \begin{pmatrix} \varphi(k) \\ 0 \end{pmatrix}$$

then we obtain with the aid of (12) the solution

$$c_1(t,k) = \left\{\cos\frac{\Omega(k)t}{2} + i\frac{\Delta(k)}{\Omega(k)}\sin\frac{\Omega(k)t}{2}\right\}\varphi(k)e^{-i\alpha(k)t},$$
$$c_2(t,k) = \frac{2iV}{\Omega(k)}\sin\frac{\Omega(k)t}{2}\varphi(k)e^{-i\alpha(k)t}. \quad (13)$$

We see therefore that nutation transitions take place between the states $|1\rangle$ and $|2\rangle$ of the atom, and, as follows from physical considerations, the transitions are accompanied by changes of the momentum of the atom by an amount equal to the photon momentum $\hbar k_0$. The latter circumstance affects the nutation frequency (8), which is adjusted by an amount equal to the Doppler shift $k_0\bar{v} = k_0(k + k_0/2)\hbar/M$, where $\bar{v}$, obviously, is simply the arithmetic mean of the velocities of the atom in the lower state $\hbar k/M$ and in the upper state $\hbar(k + k_0)/M$.

Using this result, we calculate the probability of finding the atom in the excited state:

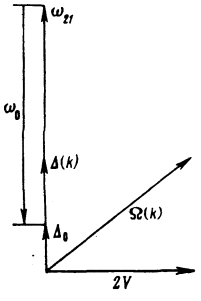

FIG. 3. Gyroscopic model. Introduction of the effective field with account taken of the motion of the atom. $\Delta(k) - \Delta_0 = \hbar k_0(k_0/2)M^{-1}$—Doppler shift.

$$P_2(t) = \int_{-\infty}^{+\infty} |c_2(k)|^2\,dk = 4V^2\int_{-\infty}^{+\infty}\frac{\sin^2[\Omega(k)t/2]}{\Omega^2(k)}|\varphi(k)|^2\,dk$$

or

$$P_2(t) = 4V^2\int_{-\infty}^{+\infty}\frac{\sin^2[\Omega_k t/2]}{\Omega_k^2}\left|\varphi\left(k - \frac{k_0}{2}\right)\right|^2 dk, \quad (14)$$

where $\Omega_k = \Omega(k - k_0/2)$ is the nutation frequency with the Doppler shift corresponding to the atom momentum $\hbar k$. Thus, the result (14) allows us to state that the influence of the translational motion on the nutation transitions reduces to averaging over the Doppler shifts (in analogy with the averaging in[12]), it being necessary to average over a momentum distribution whose shape is the same as the initial one but whose center is shifted by $\hbar k_0/2$.

3. TRANSFORMATION OF THE SHAPE OF A WAVE PACKET

We consider now the influence of the nutation transitions on the translational motion of atoms. We investigate the evolution of the shape of a coordinate wave packet. To this end we rewrite the wave function (13) in the coordinate representation:

$$c_i(t,z) = (2\pi)^{-1/2}\int_{-\infty}^{+\infty} e^{ikz}c_i(t,k)\,dk. \quad (15)$$

Expressions (15) and (13) constitute a solution in quadratures. We analyze the obtained solution, assuming that the initial function $\varphi(k)$ has a width Δk small enough for the Doppler-shift scatter to be much smaller than the average nutation frequency $\Omega = \Omega(\bar{k})$ ($\bar{k}$ is the center of the distribution $|\varphi(k)|^2$):

$$\hbar\Delta k k_0/M \ll \Omega. \quad (16)$$

According to (8), the frequency Ω is a function of the electric field intensity, and consequently the condition (16) is easy to satisfy in sufficiently strong fields. When this condition is satisfied, the nutation frequency $\Omega(k)$ can be expanded in powers of $q = k - \bar{k}$:

$$\Omega(k) = \Omega(\bar{k}+q) = \Omega + \frac{\hbar k_0\Delta}{M\Omega}q + \frac{2V^2}{\Omega^3}\left(\frac{\hbar k_0 q}{M}\right)^2 - \frac{2V^2\Delta}{\Omega^5}\left(\frac{\hbar k_0 q}{M}\right)^3; \quad (17)$$

and is necessary here to retain for $\Omega(k)$, which enters in the arguments of the periodic functions, the terms up to second order of smallness inclusive (if the second term is equal to zero, the third term must be included), whereas in the pre-exponential factors it suffices to replace $\Omega(k)$ and $\Delta(k)$ by Ω and $\Delta = \Delta(\bar{k})$. Using (17), we obtain from the exact solution (15) and (13) the distribution function of the atom coordinate.

$$\Phi(t,z) = |c_1(t,z)|^2 + |c_2(t,z)|^2 = p_1\Phi_1(t,z) + p_2\Phi_2(t,z); \quad (18)$$
$$p_{1,2} = \tfrac{1}{2}(1 \pm \Delta/\Omega), \quad p_1 + p_2 = 1, \quad (19)$$
$$\Phi_{1,2}(t,z) = \frac{1}{2\pi}\left|\int_{-\infty}^{+\infty}\exp\{iq(z - v_{1,2}t) - i\varepsilon_{1,2}^{(2)}(q)t\}\varphi(\bar{k}+q)\,dq\right|^2, \quad (20)$$
$$v_{1,2} = \hbar(\bar{k} + k_0 p_{2,1})/M, \quad (21)$$
$$\varepsilon_{1,2}^{(2)}(q) = \frac{\hbar q^2}{2M}\left(1 \mp \frac{2V^2\hbar k_0^2}{M\Omega^3}\right). \quad (22)$$

The solution (18)–(22) shows that $\Phi(t, z)$ is the sum of two packets, Φ_1 and Φ_2, which move with different velocities v_1 and v_2, and in addition spread at different rates. The evolution of these packets is determined by the dispersion curves $\varepsilon_{1,2}(k)$ (Fig. 2). The slopes of the curves at the point $k=\bar{k}$ yield the motion velocities (21), and the next term of the expansion (22) determines the rate of spreading, Attention is called to the substantially different behavior of the $\varepsilon_1(k)$ and $\varepsilon_2(k)$ curves. The upper curve $\varepsilon_2(k)$ has a nonzero second derivative at all points (see formula (22) and Fig. 2), and therefore the dispersion law for the packet Φ_2 is always quadratic in q, just as for a free particle, and its width is therefore

$$\Delta z(t)=\frac{\hbar\Delta k}{M}\left(1+\frac{2V^2\hbar k_0^2}{M\Omega^3}\right)t. \tag{23}$$

In contrast to $\varepsilon_2(k)$, the lower curve $\varepsilon_1(k)$ can have inflection points (the case shown dashed in Fig. 2). We note that according to (22), the condition

$$\Omega^3=2V^2\hbar k_0^2/M, \tag{24}$$

which connects the values of the effective field and the average Doppler shift Δ, should be satisfied at the inflection points. If the wave packet is localized in momentum space (in the sense of condition (16)) near the inflection point, then the spreading of the coordinate packet Φ_1 is described by the dispersion law

$$\varepsilon_1^{(3)}(q)=\frac{V^2\Delta}{\Omega^5}\left(\frac{\hbar k_0 q}{M}\right)^3 \tag{25}$$

and is anomalously slow—in this case

$$\Delta z(t)=\frac{\hbar\Delta k}{M}\left(\frac{\hbar k_0\Delta k}{4\Omega^2}\Delta\right)t\ll\frac{\hbar\Delta k}{M}t. \tag{26}$$

Let us determine the field intensities acting on the atom at which an anomalously slow spreading is possible. From the condition (24) we easily find that the $\varepsilon_1(k)$ can have inflection points only if

$$2V\leqslant\hbar k_0^2/2M \tag{27}$$

(one point in the case of the equality and two points in the case of the inequality). However, since an arbitrarily weak field cannot hinder the normal spreading of a wave packet, it becomes obvious that there is also a lower bound on the field intensity. We obtain this bound,

$$2V\gg\hbar\Delta k(k_0\Delta k)^{1/2}/M, \tag{28}$$

by using (16) and (24). It means that the energy of the "binding" of the atom by the field dE_0 should exceed the kinetic energy due to the uncertainty in the momentum of the atom $\hbar\Delta k$.

Let us discuss the results in terms of a two-level model. The packets ε_1 and ε_2 describe the motion of atoms that are in fundamental states with respective energies $\varepsilon_{1,2}(\bar{k})$, the factors p_1 and p_2 (18) being the probabilities for the expansion of the initial (unexcited) state of the atom in a stationary basis. An equally simple interpretation can be offered for the velocities of the packets (formulas (21)). Since the momenta of the atom is $\hbar\bar{k}$ in the lower state and $\hbar(\bar{k}+k_0)$ in the upper state, the velocity is

$$v_i=\hbar\bar{k}w_1(i)/M+\hbar(\bar{k}+k_0)w_2(i)/M, \tag{29}$$

where $w_1(i)$ and $w_2(i)$ are respectively the probabilities of finding the atom excited and unexcited in the i-th stationary state, respectively. Since

$$w_1(1)=w_2(2)=p_1,\quad w_2(1)=w_1(2)=p_2,$$

we obtain from (29) the velocities (21).

Thus, when the condition (16) that the Doppler shifts have a small scatter is satisfied, the initial coordinate wave packet splits up into two packets Φ_1 and Φ_2, which evolve in different manners: they move with different velocities v_1 and v_2, and also spread out at different rates. It is interesting to note that the velocity $(\bar{k}+k_0/2)\times\hbar/M$, which enters in definite values of the Doppler shift $\Delta=\Delta(k)$, does not coincide generally speaking with either v_1 or v_2, and is equal to $(v_1+v_2)/2$. The splitting into two packets is the consequence of the fact that the initial state was a superposition both of states $\psi_{1,k}(\varkappa)$ (with energy ε_1) and of states $\psi_{2,k}(\varkappa)$ (with energy ε_2). By a special formulation of the initial state, i.e., in terms of either only the functions $\psi_{1,k}$ or only the functions $\psi_{2,k}$ we obtain one packet—either Φ_1 or Φ_2. If the condition (24) is satisfied in addition, we can obtain only one anomalously slowly spreading packet.

According to the solution (18)–(22), the packet velocities v_1 and v_2 are constant, i.e., the atom moves uniformly. This result may seem strange. Indeed, radiation produces between levels transitions accompanied by a change in the velocity of the atom by an amount $\hbar k_0/M$. It seems thus that these transitions should lead to alternating acceleration and deceleration of the motion of the atom. The reason for the absence of such pulsations in the motion of the atom is that they cannot be discerned against a background of the large uncertainty in the coordinate of the atom. In fact, when (16) is satisfied, this uncertainty is given by

$$\Delta z\geqslant 1/\Delta k\gg\hbar k_0/M\Omega,$$

and since the oscillations of the velocity with amplitude $\hbar k_0/M$ and frequency Ω lead to oscillations of the coordinate $\Delta z_\Omega\sim\hbar k_0/M\Omega$, we obtain the inequality $\Delta z\gg\Delta z_\Omega$. In the solution (18), the terms corresponding to the oscillations Δz_Ω, have been discarded because they are of the order of smallness $\hbar k_0\Delta k/M\Omega\ll 1$ (these are interference terms containing the products of integrals of exponentials contained in (20)).

We thank A. G. Kofman for a useful discussion.

[1]N. Torrey, Phys. Rev. 76, 1059 (1949).
[2]I. I. Rabi, N. F. Ramsey, and J. C. Schwinger, Rev. Mod. Phys. 26, 167 (1954).
[3]L. D. Landau and E. M. Lifshitz, Kvantovaya mekhanika (Quantum Mechanics), Fizmatgiz, 1963, p. 173 [Pergamon,

1965].
[4]A. I. Burshteĭn, Lektsii po kursu Kvantovaya kinetika (Quantum Kinetics Lecture Course), Pt. 1, Novosibirsk State Univ., 1968, p. 23.
[5]A. Ashkin, Phys. Rev. Lett. 25, 1321 (1970).
[6]A. P. Kazantsev, Zh. Eksp. Teor. Fiz. 63, 1628 (1972); 66, 1599 (1974) [Sov. Phys. JETP 36, 861 (1973); 39, 784 (1974)]; Pis'ma Zh. Eksp. Teor. Fiz. 17, 212 (1973) [JETP Lett. 17, 150 (1973)].
[7]A. P. Kazantsev and G. I. Surdutovich, Pis'ma Zh. Eksp. Teor. Fiz. 21, 346 (1975) [JETP Lett. 21, 158 (1975)].
[8]A. P. Kazantsev, Zh. Eksp. Teor. Fiz. 67, 1660 (1974) [Sov. Phys. JETP 40, 825 (1975)].
[9]A. P. Botin and A. P. Kazantsev, Zh. Eksp. Teor. Fiz. 68, 2075 (1975) [Sov. Phys. JETP 41, 1038 (1975)].
[10]A. Yu. Pusep, Zh. Eksp. Teor. Fiz. 70, 851 (1976) [Sov. Phys. JETP 43, 441 (1976)].
[11]V. M. Faĭn, Fotony i nelineĭnye sredy (Photons and Nonlinear Media), Sov. Radio, 1972, p. 113.
[12]A. Abragam, Principles of Nuclear Magnetism, Oxford, 1961, (Russ. Transl. IIL, 1963, p. 32).

Translated by J. G. Adashko

New bands in low-temperature fluorescence of anthracene crystals at high exciton concentrations

V. A. Benderskiĭ, V. M. Beskrovnyĭ, V. Kh. Brikenshteĭn, V. L. Broude,[1)] A. G. Lavrushko, and A. A. Ovchinnikov[2)]

Division of Institute of Chemical Physics, USSR Academy of Sciences
(Submitted July 14, 1976)
Zh. Eksp. Teor. Fiz. 72, 106–118 (January 1977)

Changes in the fluorescence spectra of the anthracene crystals were investigated at a pulsed pump intensity 10^{20}–2×10^{23} cm^{-2} sec^{-1} and at crystal temperatures (at the instant of measurement) 10–16° K. It is observed that at exciton concentrations $(2–5)\times10^{17}$ cm^{-3} each of the vibronic bands of the fluorescence spectrum acquires, above a certain threshold, an additional component with half-width 100–150 cm^{-1}. At maximum pump intensities the additional emission can reach half the total integrated emission of the crystal. When the new bands are produced, a decrease is observed in the quantum yield of the total radiation from the crystal. It is shown that these bands are due to interaction between the excitons when their concentration is high. This interaction is interpreted in terms of a phase transition with formation of a new phase having an increased exciton concentration. Experimental and theoretical estimates are obtained for the exciton concentration in the phase, and yield values that are one or two orders of magnitude smaller than the concentration of the molecules in the crystal.

PACS numbers: 78.55.Kz, 71.35.+z

1. INTRODUCTION

An investigation of germanium crystals subjected to intense optical excitation has led to observation of exciton condensation, as a result of which electron-hole drops (EHD) are produced at a definite temperature and concentration of the excitons[1–3]; this process has a threshold. Data on the formation of EHD in crystals of other semiconductors were also obtained.[4,5] For a gas of Frenkel excitons, the binding energies of which are much larger than their interaction energies, one can expect condensation to result not in EHD but in drops of a dielectric exciton liquid. Because of the short-lived interaction (compared with the Coulomb interaction) between small-radius excitons, and also because of their short lifetimes, the exciton concentrations needed for this purpose must be several orders of magnitude higher than in semiconducting crystals, and the measurement times correspondingly shorter. It was observed earlier[6] that in a molecular anthracene crystal a strong broadening of the exciton-fluorescence bands sets in at exciton concentrations $\sim10^{7}$ cm^{-3}. This phenomenon was tentatively explained as being due to formation of a dielectric exciton phase of increased density, the emission of which consists of broad bands that are superimposed on the narrower fluorescence bands of the free excitons.

An investigation of the fluorescence of anthracene crystals at low temperatures and high optical-pumping levels[7–9] has shown that, depending on the experimental conditions (sample thickness, dimensions of the pumping region, the quality of the end-face faceting) the result is either lasing at the most intense vibronic transition in the fluorescent spectrum, or a decrease of the fluorescence quantum yield as a result of bimolecular recombination of the excitons (nonlinear quenching). Since lasing is accompanied by stabilization of the exciton concentration, high concentrations can be obtained only by suppressing the lasing, in particular, by decreasing the linear dimensions of the pumped volume. Even in this case, however, the exciton concentration increases sublinearly with the pump, owing to nonlinear quenching, making it necessary to use very high pump levels (up to 10^{23} cm^{-2} sec^{-1}) in order to exceed the threshold exciton concentrations at which the aforementioned band broadening takes place. Under these conditions, heating of the crystals during the pulse is unavoidable because of the nonradiative conversion of the excitation quanta into excitons of the lowest band and the release