INVESTIGATION OF THE STATISTICAL PROPERTIES OF ULTRASHORT LIGHT PULSES BY TWO-PHOTON ABSORPTION IN SEMICONDUCTORS

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The absolute values of the first moments of the radiation from a neodymium mode-locked laser are measured by means of the nonlinear absorption effect which results in a nonexponential decrease of the intensity and is induced by two-photon interband transitions in a semiconductor. The results of the measurements are presented in the form of the effective durations and intensity of the ultrashort pulses. It is shown that the laser output radiation possesses the same moments as those which could be ascribed to a hypothetical pulse with a Gaussian temporal and transverse shape, having a peak intensity 1.5 GW/cm^2 and a duration 8 μsec.

IT has been established by now that pulses from mode-locked lasers (see the reviews[1,2]) have as a rule complicated temporal structures. The main method of measuring the parameters of such lasers is at present the "collision method," based on observing the anti-Stokes luminescence induced by two-photon absorption in organic dyes; there are also methods employing harmonic generation or more complicated nonlinear-optics effects. All these methods make it possible to measure, as a rule, only the relative statistical characteristics of pulses, for example the normalized correlation functions of the intensity, which, as shown by Kuznetsova[2], cannot be used to reconstruct the true shape of the pulse without making additional assumptions[3].

Fortunately, however, in most applications it suffices to know the simplest integral (mean-squared, "macroscopic") parameters of the field. Thus, the energy effectiveness of inertialess n-photon processes, such as harmonic generation in thin layers of matter or multiphoton transitions between sufficiently broad levels, is determined by the n-th moments of the intensity

$$\langle S^n\rangle \equiv \int_{-\infty}^{\infty} S^n(x, y, t)\,dx\,dy\,dt.$$

In the present paper, an attempt is made to measure the absolute values of several first moments of mode-locked neodymium-glass laser emission ($\hbar\omega$ = 1.17 eV) with the aid of the nonlinear absorption effect leading to non-exponential decrease in the intensity and due to two-photon interband transitions in a semiconducting single crystal with suitable forbidden-band width ($CdS_{0.6}Se_{0.4}$, $E_g \sim 2.1$ eV). The use of the nonlinear-absorption effect rather than harmonic generation or the collision method[1] is dictated by the feasibility of simple and sufficiently reliable absolute measurements of $\langle S^2\rangle$ (Sec. 3) and of relative measurements of $\langle S^{3-5}\rangle$ (Sec. 4).

Results of the absolute measurements of the moments are conveniently represented in the form of effective durations[1)]

$$\tau_n \equiv \frac{0.82}{d^2}\left(\frac{\langle S\rangle^n}{n^{3/2}\langle S^n\rangle}\right)^{1/(n-1)} \qquad (1)$$

where n = 2, 3, ..., and d is the beam diameter at the 0.5 level. Thus, τ_n is the duration (at the 0.5 level) of a Gaussian pulse equivalent to the given one in energy $\langle S\rangle$ and in the n-photon effectiveness $\langle S^n\rangle$. The definition (1) presupposes that the investigated beam has a Gaussian cross section[2)]. For the sake of clarity we can introduce also the effective peak intensity

$$S_n \equiv 0.82W/\tau_n d^2. \qquad (2)$$

For example, in the experiments described in Sec. 3

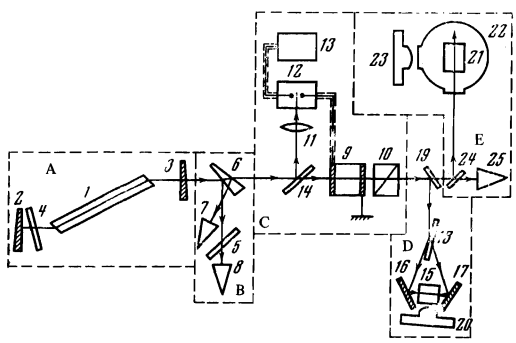

FIG. 1. Experimental setup: A–mode-locked neodymium laser (1–active element, 2, 3–mirrors evaporated on wedge-like substrates, 4–cell with saturating filter); B–scheme for the measurement of the second moment of the radiation (5–$CdS_{0.6}Se_{0.4}$ crystal, 6–glass wedge, 7, 8–calorimeters); C–scheme for separating a single pulse (9–KDP crystal, 10–Glan prism, 11–short-focus lens, 12–nitrogen discharge gap, 13–high-voltage source; 14–glass plate); D–scheme for measuring the correlation time of the radiation intensity (15–$CdS_{0.8}Se_{0.2}$ crystal; 16, 17–mirrors, 18, 19–glass plates, 20–photographic camera); E–scheme for the determination of the higher moments by measuring the luminescence track (21–$Cd_{0.6}Se_{0.4}$ crystal; 22–cryostat, 23–photographic camera, 24–glass plate; 25–calorimeter).

[1)] We note that the definition (1) differs from the effective durations introduced by Kuznetsova [2,4] only in numerical coefficients and in the use of infinite integration limits, so that in our case τ_n characterizes the entire pulse train as a whole.

[2)] In the general case, the cross section d^2 in (1) should be replaced by the corresponding effective value $\langle d^2\rangle_n$; furthermore, if the spatial and temporal coordinates can not be separated, we can determine only the effective volume of the pulse $c\langle\tau d^2\rangle_n$.

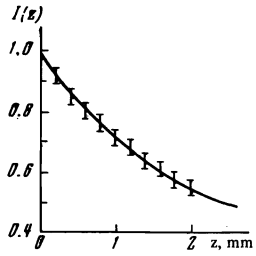

FIG. 2. Decrease of luminescence intensity of a $CdS_{0.6}Se_{0.4}$ crystal along the laser beam following excitation by an ultrashort light pulse.

below, $S_2 = 1.5$ GW/cm^2 at W = 1 mJ, d = 2.8 mm, and $\tau_2 = 8$ psec. These figures were obtained by simultaneously measuring the pulse energy before and after (W and W′) passing through a thin crystal ($l = 0.28$ mm, Fig. 1) mounted at the Brewster angle (see also[5], where a correlation was observed between the difference W − W′ and the second-harmonic intensity).

Additional measurements of the two-photon absorption coefficient β ($\beta = 0.02$ cm/MW) in the employed crystal, using nanosecond single-mode radiation, are described in Sec. 2. The energies W and W′ were also measured in this case, but the input energy W was varied and a thicker crystal was used ($l = 1.6$ mm). In Sec. 1 we consider certain refinements of the formulas used to interpret such experiments.

Finally, in Sec. 4 we describe experiments aimed at determining the higher moments of single picosecond pulses from the rate of decrease of the luminescence brightness I(z) along the laser beam as it passes through the same material. The luminescence track accompany the recombination of two-photon-excited carriers was photographed from the side (Fig. 1)[3)]. The result of these experiments are shown in Fig. 2, from which it follows that the observed I(z) dependence is close to the theoretical one based under the assumption that τ_n = const = 4 psec[4)], S_n = const = 0.2 GW/cm^2, and n = 2—5. Thus, the multiphoton effectiveness (at least for $n \lesssim 5$) of the emission of our laser coincides with that obtained for a single Gaussian pulse having the indicated duration and peak intensity.

1. TRANSFER COEFFICIENT OF A NONLINEAR LAYER—ALLOWANCE FOR PULSE STRUCTURE AND THE REFLECTION FROM THE REAR FACE

When passage of a light pulse through a nonlinear absorbing layer is described, it is customary to disregard[7-10] two essential circumstances, namely that the space-time structure of the pulse is not rectangular, and the presence of Fresnel reflection from the output boundary of the layer. The latter leads to an increase of the energy density and consequently of the rate of the nonlinear absorption.

Under the conditions of the experiments described above it is apparently possible to neglect effects of linear diffraction, self-focusing, dispersion of group velocities, and self-modulation on account of the nonlinear correction to the refractive index. Thus, we assume the response of the medium to be purely active, inertialess, local in time (we exclude stepwise cooperative multiphoton processes, among them absorption by non-equilibrium carriers with large lifetimes) and in space. In these approximations, the only manifestation of nonlinearity of matter is the effect of "self-limitation of the intensity"[7-9] and—in the presence of an opposing wave—the effect of mutual limitation ("cross-limitation"). These effects, with allowance for linear absorption are described, as can be readily shown, by the following system of equations:

$$\begin{aligned} dS_+/d\eta &= \alpha S_+ + \beta S_+^2 + 2\beta S_+ S_-, \\ dS_-/d\xi &= \alpha S_- + \beta S_-^2 + 2\beta S_+ S_-, \end{aligned} \tag{3}$$

where $\eta = -(z + vt)/2$, $\xi = (z - vt)/2$. The coefficient 2 in (3) is connected with the increase of the rate of two-photon absorption in the standing wave formed by opposing waves with identical average frequency.

We consider first the case of radiation with few modes, such that the coherence length $c/\Delta\omega$ is much larger than the layer thickness l. We can then replace ξ and $-\eta$ by z. If, in addition, $\alpha = 0$, then we can write the general solution of (3) in the implicit form:

$$\begin{aligned} S_+ &= \exp[2S_-(c_1 + \beta z)] - 1, \\ S_- &= \exp[2S_+(c_2 - \beta z)] - 1. \end{aligned} \tag{4}$$

We confine ourselves to allowance for single reflection from the exit face of the sample, and then the boundary conditions take the form

$$S_+(0) \equiv S_0, \quad S_-(l)/S_+(l) = R$$

(R is the reflection coefficient) and the integration constants in (4) should be equal to

$$\begin{aligned} c_1 &= \frac{a}{S_+(l)} - l, \quad a \equiv \frac{\ln(1+2R)}{2R}, \\ c_2 &= \frac{b}{S_+(l)} - l, \quad b \equiv \frac{\ln(1+2/R)}{2}. \end{aligned} \tag{5}$$

We denote the transfer coefficient of the layer by $S_+(l)/S_0 \equiv G$ and the normalized thickness of the layer by $\tilde{z} = \beta l S$, and then the dependence of G on $\tilde{z}$ and R is determined implicitly from the transcendental equation

$$\begin{aligned} 1 + f - \exp[f(a/G - \tilde{z})] &= 0, \\ f(G, \tilde{z}, R) &= 4/\{\exp[2(b/G + \tilde{z})] - 1\}. \end{aligned} \tag{6}$$

A plot of this dependence is shown in Fig. 3. In our approximation (narrow spectrum, absence of diffraction, etc.) G depends on x, y, and t parametrically via $S(x, y, 0, t) \equiv S_0$. It follows from Fig. 3 that reflection at $R \gtrsim 0.15$ significantly changes the value of the nonlinear absorption in the layer in comparison with the customarily employed quantity $G_0 = (1 + \tilde{z})^{-1}$.

We now take into account simultaneously the influence of single reflection and linear absorption with the aid of a successive-approximation method. The stationary solution of (3), accurate to terms of order l^2, is

$$\begin{aligned} S_+(l) \equiv S_l &= \sum a_n S_0^n, \\ a_1 &= 1 - al + \alpha^2 l^2/2, \\ a_2 &= -[1 + 2R - 3al(1 - 2R)/2]\beta l, \\ a_3 &= (1 - 5R - 5R^2)\beta^2 l^2. \end{aligned} \tag{7}$$

We take furthermore into account the spatial and slow temporal dependences of the input intensity S_0

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3) A brief description of this method, and also of its three-photon variant, is given in [6].

4) The difference between this figure and that given above may be due to the difference between the values of τ_2 of a single pulse and of a pulse train.

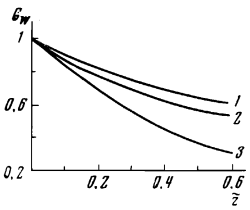

FIG. 3. Dependence of the energy transfer coefficient on the normalized thickness of the layer: curve 1–for $(1 + \tilde{z})^{-1}$, curve 2–for a single-mode Gaussian pulse at R = 0.17 and $\alpha = 0.2$ cm^{-1} according to (8), curve 3–for multimode radiation at R = 0 in accordance with (14).

= TS(x, y, t), where T = 1 − R is the transfer coefficient of the face. We are interested in the output energy

$$W' = T\langle S_l\rangle = \sum a_n T^{n+1}\langle S^n\rangle.$$

We express $\langle S^n\rangle$ in terms of the energy of the initial pulse $W = \langle S\rangle$ with the aid of the "effective volume" $V \equiv 0.82W/\langle S^n\rangle$, where the coefficient $0.82 = (4\pi^{-1}\ln 2)^{3/2}$ is so chosen that in the case of a pulse that is Gaussian in time and in space we have $V_n = \text{const} = d^2\tau$, where d and τ are the diameter and duration at the 0.5 level. On the other hand, if the pulse is Gaussian only in space, then $V^n = d^2\tau_n$.

Thus, in the case of a sufficiently monochromatic radiation with allowance for reflection from the exit face, the usually measured energy transfer coefficient of a layer with linear and quadratic absorption is equal to

$$G_W \equiv \frac{W'}{W} = T^2\sum_{n=1} \frac{a^n}{n^{3/2}}\left(\frac{0.82W}{V_n}\right)^{n-1}. \qquad (8)$$

This formula, with $V_n = \text{const} = d^2\tau$, is used in Sec. 2 to determine the coefficient of two-photon absorption from the measured $G_W(W)$ dependence.

Let us consider further the passage of multimode radiation through a nonlinear layer at R = 0.[5)] Instead of (3) we now have

$$dS/d\eta = \alpha S + \beta S^2, \qquad (9)$$

so that

$$S' = S/(1 + Sz'), \qquad (10)$$

where

$$S' \equiv e^{\alpha z}S(x, y, z, t), \quad S \equiv S(x, y, 0, t - z/v), \qquad z' \equiv \beta(1 - e^{-\alpha z})/\alpha \approx \beta z. \qquad (11)$$

It is expedient to treat multimode radiation statistically, replacing the integration with respect to time by averaging over an ensemble of lasers. The transformation (10) enables us to find the statistics of the output radiation, if we know the distribution law p(S) of the initial radiation. If the modes are not locked, then the distribution can be regarded as normal

$$p(S) = \bar{S}^{-1}\exp(-S/\bar{S}), \qquad (12)$$

so that $\bar{S}^n = n!\bar{S}^n$. Averaging (10), we obtain

$$\bar{S}' = \int_0^\infty S'p(S)dS = \bar{S}G(\tilde{z}), \quad \tilde{z} = z'\bar{S}, \qquad (13)$$

$$G(x) = x^{-1} + x^{-2}e^{1/x}\,\mathrm{Ei}(-x^{-1}) \approx 1 - 2!x + 3!x^2 \ldots .$$

As expected, the normally fluctuating signal attenuates in a thin quadratic layer twice as fast as a single-mode signal with the same average energy, for which $G = 1 - \tilde{z} + \tilde{z}^2 \ldots$ (see Fig. 3).

We now take into account in (13) the slow dependence of S on x, y, and t, which we shall assume Gaussian with maximum value $S_0 = 0.82$ W/d$^2\tau$. Integration yields the following energy transfer coefficient:

$$G_W = 1 - 2!\,2^{-3/2}z'\bar{S}_0 + 3!\,3^{-3/2}(z'\bar{S}_0)^2 - \ldots . \qquad (14)$$

Thus, we have found G_W in the case of a non-synchronized multimode Gaussian pulse at R = 0 (14) and a single-mode Gaussian pulse at R ≠ 0 (Eq. (8) with $V_n = d^2\tau$). A comparison of these expressions with the trivial $G_W = (1 + \beta zS)^{-1}$ (Fig. 3) shows that the use of the latter in the measurement of β (a summary of the measured values of β is contained in the review by Brodin[12]) can result in an overestimate exceeding 100% in the case of a multimode laser at R = 0, and by 1 + 2R times in the case of a single-model laser at R ≠ 0. Additional errors can be introduced by failure to take into account nonlinear absorption, scattering, and the smooth structure of the pulse.

Finally, formula (8) makes it possible to determine from energy measurements the effective durations (volumes, higher moments) of arbitrary radiation if β is known. In practice, however, such measurements are possible only at $\tau_n \lesssim 10^{-10}$ sec[13], since the samples are damaged by longer pulses.

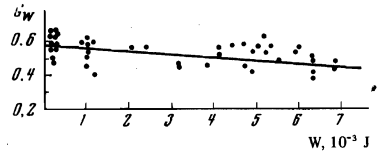

FIG. 4. Experimental dependence of the transmission of the semiconducting single crystal $CdS_{0.6}Se_{0.4}$ on the energy of the incident radiation.

2. MEASUREMENT OF β WITH THE AID OF SINGLE-MODE RADIATION

For an accurate determination of β in $CdS_{0.6}Se_{0.4}$, we used a single-mode Q-switched neodymium laser with controllable time structure of the pulse and intensity distribution over the beam cross section. The pulse structure was well approximated by a product of two Gaussian functions with diameter d = 0.5 mm and duration τ = 30 nsec. The z-cut crystal was 1.6 mm thick and was cooled to 77° K.[6)] The energy W of the pulse incident on the sample was varied with neutral optical filters. The measured functions W(W') are shown in Fig. 4. The straight line was drawn by least squares neglecting the terms quadratic in l. The plot yields an initial sample transmission 0.58 and a linear absorption coefficient $\alpha = 0.8$ cm^{-1}, which agree well with the result of independent measurements made with a helium-neon laser of wavelength 1.15 μ.

The coefficient β, determined from the formula

$$\beta = k2^{3/2}d^2\tau/[1 + 2R - {}^3/_2\alpha l(1 - 2R)]l(1 - R), \qquad (15)$$

which follows from (8) (R = 0.17 and k is the slope of the line in Fig. 3), is equal to 0.02 cm/MW. The error is due mainly to the absolute error of the calorimeters and amounts to ±30%.

[5)] This problem was recently considered by Weber [11], who, in particular, obtained formula (13) at $\alpha = 0$.

[6)] In this case the absorption by the non-equilibrium carriers seems to become negligibly small [9].

3. MEASUREMENT OF THE ABSOLUTE VALUE OF THE SECOND MOMENT OF PICOSECOND LASER EMISSION

The experimental setup with which we determine the absolute value of the second moment is shown in Fig. 1. Part of the mode-locked laser radiation was fed to a thin semiconducting plate (l = 0.28 mm), mounted at the Brewster angle φ to eliminate specular reflection from its faces. Two calorimeters were used to measure the incident and transmitted energies (W and W').

The following formulas for the equivalent duration and intensity can be derived from (1), (2), (7), and (8):

$$\tau_2 = \left(\frac{2\ln 2}{\pi}\right)^{3/2} \frac{\beta l' W G_0^{1/2}}{d^2(1 - W'/G_0 W)}, \qquad S_2 = \left(\frac{8}{G_0}\right)^{1/2} \frac{1 - W'/G_0 W}{\beta l'} \tag{16}$$

($l' = l \sin\varphi$, $\varphi = \tan^{-1} n = 65°$ and $G_0 = T^2(1 - \alpha l)$ is the initial transmission of the plate, with account taken of the linear scattering and absorption); it follows from these formulas that at β = 0.02 cm/MW we get τ_2 = 8 psec and S_2 = 1.5 GW/cm^2.[7] These numbers are averages of approximately 100 measurements made in different days and with different saturable filters. The variation of τ_2 amounted to ~10%, whereas the energy variation was larger by one order of magnitude, thus indicating that τ_2 is a highly universal laser characteristic.

The operation of the described simple correlator was verified by simultaneously observing the second-harmonic energy $W_{2\omega}$ generated in a KDP crystal 4 cm long. Since this length was shorter than the quasistationary length (oo-e interaction), $W_{2\omega}$ should be proportional to $\langle S^2 \rangle \sim 1 - W'/G_0 W$, as was indeed observed in experiment (for details see[5]).

4. DETERMINATION OF HIGHER MOMENTS BY MEASURING THE LUMINESCENCE TRACK

The next series of experiments was aimed at verifying the relative values of the higher moments. The method used for this purpose was photography of the luminescence track produced in a semiconductor by passage of a powerful pulse[6]. The track brightness I(z) decreases rapidly along z as a result of nonlinear absorption of the exciting radiation. The experiments were performed with single pulses (Fig. 1). The pulses were separated by a Pockels cell, to which a voltage shaped by a coaxial line and a discharge gap was applied. The crystal was cooled to 77°K to increase the luminescence quantum yield.

Let us assume that the luminescence is proportional to the mean-squared intensity, $I(z) \sim \langle S^2(z) \rangle$, and then (see[13]) in the case of a known Gaussian pulse

$$I(z) = \sum_{n=0}^{\infty} \frac{n+1}{(n/2+1)^{3/2}} \left(-\beta z T \frac{0{,}82W}{d^2\tau}\right)^n. \tag{17}$$

The experimental data are satisfactorily described by this formula (see Fig. 2), meaning that $\tau_n \sim$ const = τ. Simultaneous measurements of W = 1.2 × 10^{-5} J and d = 1.2 mm yielded at β = 0.02 cm/MW and T = 0.83 an equivalent duration τ = 4 psec and S_n = 0.2 GW/cm^2.[8]

This result agrees with the independent measurements of the intensity correlation time τ_c by the standard collision method[1] (if it is assumed that approximately half of the entire energy is concentrated in its synchronized part), which yielded $\tau_c \approx 4$ psec (contrast 2.3—2.5). In these measurements we used the semiconducting crystal $CdS_{0.8}Se_{0.2}$ in place of the ordinary liquid dyes. This replacement has many advantages: the large cross section and the quantum yield of the two-photon luminescence make it possible to investigate pulses of low energy or to photograph with lenses of smaller aperture, thereby increasing the resolution[14] and making it possible to choose the optimal widths of the forbidden band for lasers with different wavelengths (for example, CdS at λ = 0.69 μ or ZnS at λ = 0.53 μ).

The results of the experiment described here gives grounds for hoping that the use of semiconducting correlators with indication based on I(z) or W'(W) should gain wide use in the study of the properties of laser emission.

The authors are grateful to R. V. Khokhlov for a discussion of the results and to E. A. Muzalevskiĭ for supplying the employed single crystals.

[7] 10% of the radiation intensity was diverted to the simiconducting plate to exclude nonlinear absorption by non-equilibrium carriers.

[8] Only a fraction of the laser radiation is incident on the crystal. The effective peak intensity of the generator is ~2 GW/cm^2.

[1] P. W. Smith, Proc. IEEE, 58, 1943, 1970.
[2] B. Ya. Zel'dovich and T. I. Kuznetsova, Usp. Fiz. Nauk 106, 47 (1972) [Sov. Phys.-Usp. 15, 106 (1972)].
[3] A. S. Chirkin, Zh. Eksp. Teor. Fiz. 60, 1584 (1971) [Sov. Phys.-JETP 33, 856 (1971)]; S. A. Akhmanov and A. S. Chirkin, Opto-Electr. 3, 111 (1971).
[4] T. I. Kuznetsova, Kratkie soobshch. po fizike (FIAN) No. 6, 66 (1970); Izv. VUZov Radiofizika 15, 227 (1972).
[5] V. V. Arsen'ev, V. S. Dneprovskiĭ, D. N. Klyshko, and V. U. Khattatov, Kvantovaya élektronika, No. 7, 33 (1972) [Sov. J. Quant. Electr. 2, 23 (1972)].
[6] V. V. Arsen'ev, V. S. Dneprovskiĭ, D. N. Klyshko, and L. A. Sysoev, Zh. Eksp. Teor. Fiz. 60, 114 (1971) [Sov. Phys.-JETP 33, 64 (1971)].
[7] P. D. Maker and R. W. Terhune, Phys. Rev. 137, A801 (1965).
[8] M. S. Brodin and D. B. Goer, Fiz. Tverd. Tela 5, 256 (1971) [Sov. Phys.-Solid State 5, 186 (1971)].
[9] V. V. Arsen'ev, V. S. Dneprovskiĭ, D. N. Klyshko, and A. N. Penin, Zh. Eksp. Teor. Fiz. 56, 760 (1969) [Sov. Phys.-JETP 29, 413 (1969)].
[10] B. V. Zubov, L. A. Kulevskiĭ, V. P. Makarov, T. M. Murina, and A. M. Prokhorov, ZhETF Pis. Red. 9, 221 (1969) [JETP Lett. 9, 130 (1969)].
[11] H. P. Weber, IEEE, QE-V, 189, 1971.
[12] M. S. Brodin, in: Kvantovaya élektronika (Quantum Electronics), Naukova dumka, 1971, p. 33.
[13] V. V. Arsen'ev, Ya. T. Vasil'ev, V. S. Dneprovskiĭ, and V. U. Khattatov, Fiz. Tekh. Poluprov. 5, 403 (1971) [Sov. Phys.-Semicond. 5, 354 (1971)].
[14] V. V. Korobkin and A. A. Malyutin, Kratkie soobshch. po fizike (FIAN), No. 8, 53 (1970).

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