Quantum Electronics **51** (10) 915–919 (2021)

ACTIVE MEDIA

https://doi.org/10.1070/QEL17633

Possibility of inversionless amplification and generation of radiation by a two-level system in the 'red' wing of its spectral line under resonant diode pumping

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Abstract. **We report a theoretical study on the possibility of amplification and generation of radiation by an inversionless two-level system in the 'red' wing of its spectral line under resonant diode pumping. The two-level system is used to simulate atoms of an active gas, while it is in the atmosphere of a high-pressure buffer gas. The effect results from the fact that the probability of stimulated emission in the 'red' wing of the spectral line exceeds the probability of absorption if the homogeneous broadening due to the interaction of particles with the buffer gas significantly exceeds the natural one (at high pressures of the buffer gas). It is found that the higher the buffer gas pressure and the higher the pump radiation intensity, the greater the inversionless amplification. It is shown that the gain in a two-level system in the 'red' wing of its spectral line can reach 0.011 cm^{-1} when the radiation intensity and the width of the radiation spectrum of pump diodes are 5 kW cm^{-2} and 4 cm^{-1}, respectively. The use of transverse diode pumping of an active medium placed in an optical cavity will make it possible to obtain lasing with frequency tuning and thereby solve the problem of converting incoherent broadband radiation into a coherent laser light in a gas of two-level active particles.**

Keywords: *inversionless amplification of light, probe field, collisions, Einstein coefficients, level populations, spectral line wing.*

1. Introduction

It was shown in Refs [1–12] that in the wing of the absorption line of active (interacting with radiation) gas particles in the presence of frequent collisions with buffer particles (thermostat), the probabilities of absorption and stimulated emission are not equal to each other. It turned out that the spectral densities of the Einstein coefficients for absorption [$b_{12}(\Omega)$] and stimulated emission [$b_{21}(\Omega)$] are related by the expression [7, 8]

$$b_{21}(\Omega) = b_{12}(\Omega)\exp\left(-\frac{\hbar\Omega}{k_{\rm B}T}\right), \tag{1}$$

where $\Omega = \omega - \omega_{21}$ is the detuning of the radiation frequency ω from the frequency ω_{21} of the 2–1 transition; $\hbar$ is Planck's constant; $k_{\rm B}$ is the Boltzmann constant; and T is the temperature. Relation (1) remains valid for any sign of Ω. When $\hbar|\Omega| \ll k_{\rm B}T$, relation (1) yields the canonical equality for the probabilities of absorption and stimulated emission.

A striking consequence of relation (1) is the formation of a population inversion in a two-level system upon absorption of intense laser radiation in the 'blue' wing of the spectral line and during frequent collisions (at high pressures of the buffer gas). This effect was recorded experimentally in the form of generation of coherent radiation at the resonance frequency of the transition of sodium atoms (D_2 line) upon excitation by pulsed laser radiation in the 'blue' wing of the D_2 line [6, 8–10]. Another interesting consequence of relation (1) is the amplification of radiation by two-level systems without population inversion. If high-power pump radiation, tuned in frequency to resonance with the atomic transition, equalises the populations of the excited and ground levels, then due to an excess of the probability of stimulated emission over the probability of absorption in the 'red' wing of the spectral line for the probe radiation, the amplification regime is realised [7, 13, 14].

Let us ask ourselves a question: Is it possible, due to these effects, to convert incoherent broadband radiation into coherent laser radiation similarly to that described in works on diode-pumped alkali metal vapour lasers [14–19], where population inversion is obtained upon transition to the ground state of atoms under diode pumping at an adjacent transition (three-level configuration)? As for the effect of the formation of population inversion upon pumping in the blue wing of the spectral line, in this case the answer will be negative. Indeed, according to the results of studies [6, 8–10], population inversion is achieved at a very high (up to 10 MW cm^{-2}) pump power density, which is unattainable for modern laser diodes. A significantly more favourable situation for the laser effect in a two-level system is realised in the second variant, where the equalisation of the populations is required under resonant pumping. This has actually already been implemented in diode-pumped alkali metal vapour lasers. Thus, the radiation of existing sources of incoherent radiation (laser diodes) is quite capable of levelling the level populations under resonant excitation and thereby providing lasing in the 'red' wing of the spectral line of a two-level system. In this work, we theoretically investigate the possibility of such lasing with an orientation towards the existing possibilities of pump laser diodes.

2. Initial equations

Consider a gas of two-level active particles (with a ground level 1 and an excited level 2) in a mixture with a buffer gas. We neglect collisions between active particles, assuming that

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Received 30 June 2021; revision received 28 July 2021
Kvantovaya Elektronika **51** (10) 915–919 (2021)
Translated by I.A. Ulitkin

the concentration of the buffer gas, N_b, is much higher than the concentration of the absorbing gas, N. Let the two-level particles be affected by high-power (capable of levelling the populations of levels 1 and 2) pump diode radiation, tuned in frequency to resonance with the atomic transition. We assume that the radiation of the pump diodes has a spectrum of arbitrary width, and the radiation amplified (or generated) in the 'red' wing of the spectral line is monochromatic. The change in the populations N_1, N_2 of levels 1, 2 under the action of pump radiation, frequent collisions with buffer particles, and amplified (generated) radiation is described by the equations:

$$\frac{dN_2}{dt} = -A_{21}N_2 + w_p(N_1 - N_2) + w_{las}(\xi_{las}N_1 - N_2),$$

$$N_1 + N_2 = N, \tag{2}$$

where

$$w_p = \frac{\lambda_{21}^3 A_{21}}{8\pi^2 \hbar c}\int_0^\infty \frac{\Gamma I_{\omega p}(\omega)}{\Gamma^2 + (\omega - \omega_{21})^2}d\omega,$$

$$w_{las} = \frac{\lambda_{21}^3 A_{21} I_{las}}{8\pi^2 \hbar c}\frac{\Gamma_{oc}(\Omega_{las})}{\Gamma^2 + \Omega_{las}^2}, \tag{3}$$

$$\xi_{las} = \exp\left(-\frac{\hbar|\Omega_{las}|}{k_B T}\right), \ \Omega_{las} = \omega_{las} - \omega_{21}.$$

Here w_p and w_{las} are the probabilities of stimulated transitions under the action of pump radiation and amplified (generated) laser radiation, respectively; A_{21} is the rate of spontaneous emission (the first Einstein coefficient) for the 2–1 transition; ω_{21} is the frequency for the 2–1 transition; λ_{21} is the corresponding wavelength; I_{las} and ω_{las} are the intensity and frequency of the amplified (generated) radiation; $I_{\omega p}(\omega)$ is the spectral density of the pump radiation intensity at the frequency ω; and Γ is the collisional half-width of the absorption line (the collisional broadening is assumed to be much larger than the Doppler one). The quantity $\Gamma_{oc}(\Omega_{las})$, which depends on the detuning of the radiation frequency Ω_{las}, characterises the frequency of elastic collisions that 'knock down' the phase of the atomic oscillator [20]. The $\Gamma_{oc}(\Omega_{las})$ is included in the modified Lorentz equation, which describes the entire spectral line profile, including the distant wings [20]. With a small detuning of the radiation frequency ($|\Omega_{las}| \leqslant \Gamma$), the value of $\Gamma_{oc}(\Omega_{las})$ is equal to the impact half-width of the absorption line Γ, and with a large frequency detuning ($|\Omega_{las}| \gg \Gamma$, wing of the absorption line), $\Gamma_{oc}(\Omega_{las})$ can be greater and significantly less than Γ [20].

Under stationary conditions, from equations (2) we easily find the level populations:

$$N_1 = \frac{N}{2}\frac{2 + \varkappa_p + \dfrac{2\varkappa_{las}}{1 + \xi_{las}}}{1 + \varkappa_p + \varkappa_{las}},$$

$$N_2 = \frac{N}{2}\frac{\varkappa_p + \dfrac{2\varkappa_{las}\xi_{las}}{1 + \xi_{las}}}{1 + \varkappa_p + \varkappa_{las}}, \tag{4}$$

as well as the population differences that determine the amplification of the generated radiation and the absorption of the pump:

$$N_2 - \xi_{las}N_1 = N\frac{\varkappa_p\dfrac{1 - \xi_{las}}{2} - \xi_{las}}{1 + \varkappa_p + \varkappa_{las}},$$

$$N_1 - N_2 = N\frac{\varkappa_{las}\dfrac{1 - \xi_{las}}{1 + \xi_{las}} + 1}{1 + \varkappa_p + \varkappa_{las}}. \tag{5}$$

Here $\varkappa_p$ and $\varkappa_{las}$ defined as

$$\varkappa_p = \frac{2w_p}{A_{21}}, \ \varkappa_{las} = \frac{(1 + \xi_{las})w_{las}}{A_{21}} \tag{6}$$

denote the saturation parameters, since each of them characterises the degree of population equalisation at the 2–1 transition in the absence of the second field.

The gain g_{las} of the generated radiation is given by

$$g_{las} = \frac{\hbar\omega_{las}}{I_{las}}w_{las}(N_2 - \xi_{las}N_1)$$

$$= \frac{N\lambda_{21}^3 A_{21}}{4\pi\lambda_{las}}\frac{\Gamma_{oc}(\Omega_{las})}{\Gamma^2 + \Omega_{las}^2}\frac{\varkappa_p\dfrac{1 - \xi_{las}}{2} - \xi_{las}}{1 + \varkappa_p + \varkappa_{las}}, \tag{7}$$

where λ_{las} is the wavelength of the generated radiation, from which it follows that the inversionless amplification of radiation ($g_{las} > 0$) in the region of 'red' frequency detunings (at $\Omega_{las} < 0$) occurs if the condition

$$\varkappa_p > \frac{2\xi_{las}}{1 - \xi_{las}} = \frac{2}{\exp(\hbar|\Omega_{las}|/k_B T) - 1} \tag{8}$$

is met.

According to (8), the larger the frequency detuning of the amplified field, the lower the pump radiation intensity required for the onset of inversionless amplification. In this case, however, the gain g_{las} decreases significantly with increasing $|\Omega_{las}|$, so that in the experiment it is necessary to strive for the maximum possible values of $\varkappa_p$.

One can see from formula (7) that in the wing of the spectral line (at $|\Omega_{las}| \gg \Gamma$), the gain is directly proportional to the collisional phase relaxation rate $\Gamma_{oc}(\Omega_{las})$. Since the value of $\Gamma_{oc}(\Omega_{las})$ is proportional to the buffer gas pressure [20], an increase in the effect should be expected with increasing pressure of the buffer gas.

The gain is the greater, the higher is the concentration of active particles ($g_{las} \propto N$). However, the concentration N should not be too high; otherwise, the pump radiation will be completely absorbed at the entrance to the active medium. For efficient use of the radiation energy, it is necessary that the pump radiation be sufficiently strongly absorbed by the active medium and, at the same time, at the exit from the medium, have an intensity sufficient to maintain the required value of the gain. This requirement can be satisfied if the condition

$$\alpha_p L_p \sim 1 \tag{9}$$

is met, where α_p is the absorption coefficient of pump radiation; and L_p is the size of the active medium in the direction of propagation of the pump radiation. The absorption coefficient of pump radiation is defined as

$$\alpha_p = \frac{\hbar\omega}{I_p} w_p (N_1 - N_2) = \frac{N\pi\hbar c A_{21}}{I_p \lambda_{21}} \frac{\varkappa_p \left[\varkappa_{las} \dfrac{1-\xi_{las}}{1+\xi_{las}} + 1\right]}{1 + \varkappa_p + \varkappa_{las}}, \quad (10)$$

where I_p is the pump radiation intensity. Hence it can be seen that the higher the intensity I_p, the greater the permissible concentration of active particles, and, consequently, the gain.

In order that the requirements for the characteristics of the medium for the generated radiation and for the pump radiation do not conflict with each other, it is expedient to direct the pump radiation orthogonally to the generated radiation (transverse pumping).

3. Gaussian shape of the pump radiation spectrum

To further concretise the calculations using the above formulae, it is necessary to specify the spectral density $I_{\omega p}(\omega)$ of the pump diode radiation. We will assume that the pump radiation spectrum has a Gaussian shape:

$$I_{\omega p}(\omega) = \frac{I_p}{\sqrt{\pi}\,\Delta\omega} \exp\left[-\left(\frac{\omega - \omega_{21}}{\Delta\omega}\right)^2\right],$$

$$I_p = \int_0^\infty I_{\omega p}(\omega)\,d\omega, \quad (11)$$

where $\Delta\omega$ is the half-width (at a height of 1/e) of the pump radiation spectrum. From formulae (6) and (3), taking into account (11), we obtain the expression for the saturation parameter $\varkappa_p$:

$$\varkappa_p = \frac{\lambda_{21}^3 I_p \Phi}{4\pi^2 \hbar c \Gamma}, \quad \Phi = \frac{2}{\sqrt{\pi}} \int_0^\infty \frac{y^2 e^{-t^2}}{y^2 + t^2} dt, \quad y = \frac{\Gamma}{\Delta\omega}. \quad (12)$$

In what follows, for simplicity, we restrict ourselves to considering the case of a weak intensity of the amplified (generated) radiation, when its influence on the population differences, which determine the amplification of radiation and absorption of the pump, can be neglected. From expression (5) it follows that this is possible if the condition

$$\varkappa_{las} \ll 1 + \varkappa_p, \ \frac{1+\xi_{las}}{1-\xi_{las}} \quad (13)$$

is satisfied. Under this condition, the coefficients g_{las} (7) and α_p (10) take the form

$$g_{las} = \frac{N\lambda_{21}^3 A_{21}}{4\pi\lambda_{las}} \frac{\Gamma_{oc}(\Omega_{las})}{\Gamma^2 + \Omega_{las}^2} \frac{\varkappa_p \dfrac{1-\xi_{las}}{2} - \xi_{las}}{1 + \varkappa_p},$$

$$\alpha_p = \frac{N\pi\hbar c A_{21}}{I_p \lambda_{21}} \frac{\varkappa_p}{1 + \varkappa_p}. \quad (14)$$

Using formulae (14) we calculate the gain of a two-level system in the 'red' wing of its spectral line. Let caesium atoms be the active medium in the amplifier cell, and helium be used as a buffer gas. We will assume that the active medium is pumped in a direction transverse to the direction of the amplified laser radiation (transverse diode pumping). We also assume that the cell with active particles and a buffer gas has the shape of a rectangular parallelepiped with a length L in the direction of the amplified laser radiation and with a length L_p in the direction of radiation of the pump diodes (the cell is rather long, $L \gg L_p$).

To calculate the gain in the 'red' wing of the D_1 line of caesium atoms, it is quite possible to use the two-level model of active particles due to the weak collisional coupling between the $6^2P_{1/2}$ and $6^2P_{3/2}$ fine components of the excited state of Cs atoms in He. Indeed, the cross section σ_{FS} for collisional transitions $6^2P_{1/2} \rightarrow 6^2P_{3/2}$ between fine components of the excited state of Cs atoms in He is small: $\sigma_{FS} = 0.57 \times 10^{-20}$ cm^2 [21]. At a helium pressure $p_{He} = 10$ atm and a temperature $T = 470$ K, the frequency of collisional transitions $6^2P_{1/2} \rightarrow 6^2P_{3/2}$ between fine components is $\nu_{FS} = 1.42 \times 10^5$ s^{-1}, which is much lower than the rate of spontaneous decay of the excited $6^2P_{1/2}$ level: $\nu_{FS}/A_{21} = 5 \times 10^{-3}$. On this basis, the 6^2P excited state of Cs atoms can be modelled by a single level. The $6^2S_{1/2}$ ground level of Cs atoms is split into two hyperfine components with a frequency distance between them $\Delta\omega_{HFS} = 5.78 \times 10^{10}$ s^{-1} [$\Delta\omega_{HFS}/(2\pi c) = 0.31$ cm^{-1}] [22]. At a sufficiently high pressure of the buffer gas (several atmospheres and higher), the collisional absorption line width is large in comparison with hyperfine splitting in this state: $2\Gamma \gg \Delta\omega_{HFS}$. Therefore, the ground state can also be modelled with one level.

Let us set the initial data necessary for calculating the radiation gain in the 'red' wing of the D_1 line of caesium atoms ($6^2S_{1/2} - 6^2P_{1/2}$ transition). For caesium atoms, according to the NIST database [23], the rate of spontaneous decay of the excited level $6^2P_{1/2}$ $A_{21} = 2.86 \times 10^7$ s^{-1} and the wavelength of the D_1 line $\lambda_{21} = 894.4$ nm. The collisional broadening for the D_1 line of caesium atoms in the He buffer gas is 10.82 MHz Torr^{-1} at a temperature $T = 470$ K [24]. At a helium pressure $p_{He} = 10$ atm and a temperature $T = 470$ K, the collisional absorption line half-width is $\Gamma/(2\pi c) = 2.74$ cm^{-1}. To calculate the radiation gain in the 'red' wing of the D_1 line of caesium atoms, it is necessary to know the collisional phase relaxation rate $\Gamma_{oc}(\Omega_{las})$. In paper [25], Fig. 13 shows the calculated profile of the D_1 line of Cs atoms in a buffer He gas at a pressure of 13.6 atm and a temperature of $T = 1000$ K, and its comparison with the Lorentzian profile. From this figure it follows that in the 'red' wing of the D_1 line of Cs atoms, $\Gamma_{oc}(\Omega_{las}) \geqslant \Gamma$ up to the frequency detuning $|\Omega_{las}|/(2\pi c) \approx 1100$ cm^{-1}. On this basis, in the calculation below, we will assume that the collisional phase relaxation rate $\Gamma_{oc}(\Omega_{las})$ is equal to the collisional half-width of the line Γ. We assume that $\alpha_p L_p = 1$ (when the radiation of pump diodes passes through the active medium, its intensity decreases by a factor of 2.7). Then, for the length of the active medium in the direction of radiation of the pump diodes $L_p = 1$ cm, the absorption coefficient of the pump radiation is $\alpha_p = 1$ cm^{-1}. Let the intensity of the pump radiation be $I_p = 5$ kW cm^{-2} and the half-width of the pump radiation spectrum be $\Delta\omega/(2\pi c) = 2$ cm^{-1} (the spectral width at half-maximum is $\Delta\lambda = 0.266$ nm). It follows from (14) that the value of $\alpha_p = 1$ cm^{-1} corresponds to the concentration of active particles $N = 1.59 \times 10^{15}$ cm^{-3} (this concentration of vapours of caesium atoms is attained at a temperature of T = 470 K [26]).

With regard to the width of the pump diode emission spectrum adopted in our calculations ($\Delta\lambda$ = 0.266 nm), we note that modern laser diodes used for pumping alkali metal vapour lasers can have an emission spectrum width of less than 0.1 nm [27]. In this case, the radiation power density of individual lines of laser diodes is 1 kW cm^{-2} [27]. Therefore, to achieve the pump radiation intensity I_p = 5 kW cm^{-2}, focusing of the diode radiation is required.

Figure 1 shows the calculated dependence of the gain g_{las} in the 'red' wing of the D_1 line of caesium atoms on the frequency detuning Ω_{las}. With the parameters corresponding to Fig. 1, the maximum gain is achieved at a frequency detuning $\Omega_{las}/(2\pi c)$ = –27 cm^{-1} and is equal to 0.011 cm^{-1}. This means that in one round trip through an active medium with a length of L = 50 cm, the radiation intensity increases by a factor of 1.74.

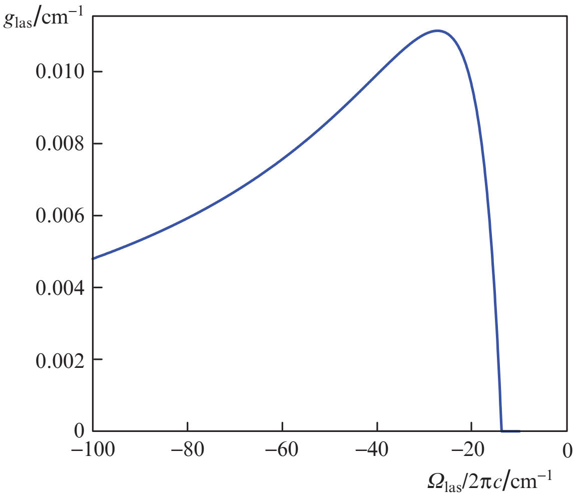

Figure 1. Radiation gain g_{las} in the 'red' wing of the D_1 line of caesium atoms ($6^2S_{1/2} - 6^2P_{1/2}$ transition) as a function of the detuning Ω_{las} of the radiation frequency from the centre transition frequency at a buffer gas pressure of helium p_{He} = 10 atm, λ_{21} = 894.4 nm, $_{21}$ = 2.86×10^7 s^{-1}, T = 470 K (N = 1.59×10^{15} cm^{-3}), I_p = 5 kW cm^{-2}, $\Delta\omega/(2\pi c)$ = 2 cm^{-1}, $\Gamma_{oc}(\Omega_{las}) = \Gamma$, and $\Gamma/(2\pi c)$ = 2.74 cm^{-1}.

This gain is sufficient to excite lasing if the active medium is placed in an optical cavity. Lasing occurs under the condition that the losses in the cavity are compensated for by the gain in the active medium (the threshold condition for the laser operation):

$$R_1 R_2 \exp(2Lg_{th} - \alpha) = 1, \tag{15}$$

where L is the length of the active medium; g_{th} is the threshold value of the gain; R_1 and R_2 are the reflection coefficients of the resonator mirrors; and α are the losses in the cavity upon a double round trip in the cavity (they consist of diffraction losses, losses at the cell windows, and losses associated with the geometric imperfection of the cavity). From condition (15) we find the threshold value of the gain:

$$g_{th} = \frac{1}{2L}\left(\alpha + \ln\frac{1}{R_1 R_2}\right). \tag{16}$$

To ensure lasing, it is necessary that the gain g_{las} exceeds the threshold value: $g_{las} > g_{th}$. The lower the threshold value of the gain (and the easier it is to achieve lasing), the longer the length of the active medium and the reflectivity of the mirrors. At L = 50 cm, α = 0.1, R_1 = 0.6, and R_2 = 1, from (16) we obtain g_{th} = 0.0061 cm^{-1}. At this value of the threshold gain, as can be seen from Fig. 1, the inequality $g_{las} > g_{th}$ (the condition for laser self-excitation) is satisfied in the frequency detuning range from –16 cm^{-1} to –77 cm^{-1}. The wavelength of the generated radiation can be tuned from 895.6 to 900.5 nm. The value of ξ_{las} in this frequency detuning range differs markedly from unity and varies from 0.95 (with a frequency detuning of –16 cm^{-1}) to 0.79 (with a detuning of –77 cm^{-1}).

4. Conclusions

We have studied theoretically the possibility of inversionless amplification and generation of radiation in the 'red' wing of the spectral line of the transition of a two-level system during resonant absorption of broadband radiation from laser pump diodes by active particles residing in a buffer gas atmosphere at high pressure. The reason for the appearance of this effect is the inequality of the spectral densities of the Einstein coefficients for absorption and stimulated emission under conditions when the homogeneous broadening due to interaction with the buffer gas significantly prevails over the natural one (at a high pressure of the buffer gas). Using the balance equations, we have obtained a formula for the radiation gain of a two-level system in the 'red' wing of its spectral line. It has been found that the higher the buffer gas pressure and the higher the concentration of active particles, the greater the gain. However, the concentration of active particles cannot be too high; otherwise, the pump radiation will be completely absorbed at the entrance to the active medium.

Calculations of the radiation gain in the 'red' wing of the D_1 line of caesium atoms have shown that the maximum value of the gain is 0.011 cm^{-1} (at the radiation intensity of the pump diodes I_p = 5 kW cm^{-2}, the half-width of the pump radiation spectrum $\Delta\omega/(2\pi c)$ = 2 cm^{-1} and the pressure of the buffer gas helium p_{He} = 10 atm). Since the radiation gain turns out to be rather small, a long path through the gain medium is required in order to achieve a noticeable gain in the active medium for one round trip. This can be realised using transverse diode pumping of the active medium. When use is made of a resonator, oscillation with frequency tuning can be obtained. For the active medium length L = 50 cm, the gain per round trip is $g_{las}L$ = 0.55, which is quite sufficient to exceed the losses in the cavity.

The calculations performed and the estimates made show that, in a gas of two-level active particles, the problem of converting incoherent broadband radiation into coherent laser radiation is quite solvable when pumping is performed by existing laser diodes. In this case, the energy of the laser photon hardly differs from the energy of the pumping photon. Finally, we note that the effect should be significantly enhanced when using laser diodes operating in a pulsed regime, since in this case it is possible to significantly increase the power density of the pump radiation.

Acknowledgements. The study was carried out at the expense of subsidies for the financial support for fulfilling the State Task (Project No. AAAA-A21-121021800168-4) at the Institute of Automation and Electrometry of the Siberian Branch of the Russian Academy of Sciences.

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