Lasing in nanowires: *Ab initio* semiclassical model

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The semiclassical equations which describe lasing in nanowires are derived from first principles. Both the lasing threshold condition and the steady-state regime of operation are discussed. It is shown that the lasing is governed by the Fourier coefficients of the field susceptibility averaged over the nanowire cross section. The general theory is illustrated by the case where a nanowire supports a single Fabry-Pérot mode originated from a transverse magnetic waveguide mode. Basing on the developed theory, the possibility of lasing without inversion is predicted and the corresponding nanolaser cavity is designed.

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I. INTRODUCTION

Recent advances in nanotechnology have led to the possibility to fabricate nanowires which can lase under pumping conditions [1]. Currently they can be grown from both inorganic and organic materials and demonstrate lasing at both optical and electrical pumping. This progress paves the way to diverse applications of nanowire lasers in nanoscale optical and optoelectronic devices.

To be able to design nanolasers and optimize their operation, one needs to have a deep understanding of the elementary optical processes which lead to lasing. So far, to describe nanolaser action, one utilizes the conventional laser equations which have been obtained for the laser cavities of dimensions much larger than the laser field wavelength [2]. One of the key approximations which has been used to derive such equations implies that both the field and polarization in the laser cavity are described by plane waves which are slowly modulated in both time and space [3]. The latter assumption is obviously not fulfilled in nanocavities where the field changes essentially on the scale of the wavelength. Moreover, such equations involve some semiphenomenological parameters, in particular the photon decay rate and the modal gain, which cannot be well defined for a nanocavity [4]. Another aspect related to small sizes of a nanocavity is the fact that the atoms or molecules of the active medium are in close proximity to the cavity walls, which implies a significant role of the cavity-quantum-electrodynamical effects [5]. One partly takes such effects into account by means of introducing the population decay rate modified by the Purcell factor in the rate equations. The rate equations do not contain, however, information about the phase of the light field and thus are incomplete. All of these arguments reveal that the laser equations should be revisited from the beginning in order to provide a basis for a proper nanolaser design.

The drawbacks highlighted above can be overcome only in the framework of a self-consistent lasing theory based on a Green's function of a system. Such a theory has been developed for rather general lasing media in Ref. [6]. It leads to a set of self-consistent integral equations that gives the laser threshold, frequency, and modal interaction. The laser equations were derived for a two-dimensional (2D) scalar problem, which corresponds to lasing in an infinitely long dielectric cylinder of arbitrary cross section. The finite longitudinal extension of a Fabry-Pérot cavity was considered in a simplified 1D model. It was also pointed out that proper treatment of the openness in the transverse plane of the cylindrical geometry is equally important [7,8].

Recently, we developed an *ab initio* analytical model which is capable of describing rigorously a 3D vector electromagnetic field in a cylindrical nanocavity of arbitrary diameter and length [9–12]. This model allows one to calculate Fabry-Pérot modes of a cylindrical nanowire [10], as well as the dyadic Green's function and the Purcell factor for a cylindrical nanocavity [12]. In this paper, we apply that approach to derive the laser equations for a nanowire (NW) from first principles and thus to fill this gap in the nanolaser theory. We use the developed theory to design a nanowire laser which can operate without a population inversion.

The paper is organized as follows. In Sec. II, we introduce a general theoretical formalism and derive the lasing condition and formulas for the steady-state regime of operation of a NW laser. In Sec. III, we apply the general theory for the case when a NW supports a single transverse waveguide mode. The obtained results are discussed in Sec. IV, which is followed by the conclusion in Sec. V. Some details of the laser condition derivation as well as the consideration of the classical analogy of lasing without inversion are given in the appendices.

II. THEORETICAL FORMALISM

Let us consider a cylindrical NW of radius a and length L which is embedded in the medium with the dielectric function ϵ_1. Let us assume that the material of the NW is described by the dielectric function ϵ_2 and the NW contains active centers (ACs) (impurity atoms, ions, or molecules, quantum dots, etc.). To consider lasing in the NW, we adopt a semiclassical description, namely, we treat the ACs as quantum-mechanical systems, while the electromagnetic field in the NW is described classically as a solution of Maxwell's equations. We model an AC by a two-level system with the ground state $|1\rangle$, excited state $|2\rangle$, transition frequency ω_{21}, and transition dipole moment μ_{12}. We assume that the transition $|2\rangle \rightarrow |1\rangle$ is homogeneously broadened and has the linewidth $\gamma_\perp$.

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We seek the solution of Maxwell's equations in a cylindrical NW in the form of the Fourier transform in time,

$$\mathbf{E}(t) = \int_0^\infty [\mathbf{E}^*(\omega)e^{i\omega t} + \mathbf{E}(\omega)e^{-i\omega t}]d\omega = \mathbf{E}^{(-)}(t)e^{i\omega_m t} + \mathbf{E}^{(+)}(t)e^{-i\omega_m t}, \quad (1)$$

where we assume that the Fourier transform $\mathbf{E}(\omega)$ has a narrow distribution around one of the NW normal-mode (Fabry-Pérot) frequencies, ω_m, so that $\mathbf{E}^{(\pm)}(t)$ are slowly varying functions of time. The amplitudes $\mathbf{E}^{(\pm)}(t)$, in turn, can be expressed as Fourier integrals over the propagation constant β,

$$\mathbf{E}^{(\pm)}(r,\theta,z,t) = \frac{1}{2\pi}\int_C \tilde{\mathbf{E}}^{(\pm)}(r,\theta,t;\beta)e^{i\beta z}d\beta, \quad (2)$$

with r, θ, and z being the cylindrical coordinates of a point, the z axis being directed along the NW axis, and C being a path in the complex plane of β running along the real axis. The Fourier transforms, $\tilde{\mathbf{E}}^{(\pm)}(\beta)$, have poles on the real axis which correspond to the waveguide modes of an infinitely long NW of the same radius, a. We assume in the following that only a single mode labeled by a and having the propagation constant β_a is relevant to lasing that can be achieved by an appropriate choice of the NW radius and length. The contribution of this mode to the total field can be written as

$$\mathbf{E}_a^{(\pm)}(r,\theta,z,t) = \left[\mathbf{E}_-^{(\pm)}(r,t)e^{-i\beta_a z} + \mathbf{E}_+^{(\pm)}(r,t)e^{i\beta_a z}\right]e^{-in\theta}, \quad (3)$$

where the integer n specifies the angular symmetry of the waveguide mode [14] and the quantities $\mathbf{E}_-^{(\pm)}(r,t)$ and $\mathbf{E}_+^{(\pm)}(r,t)$ are determined by the residues of $\tilde{\mathbf{E}}^{(\pm)}(\beta)$ at the poles $-\beta_a(\omega_m)$ and $\beta_a(\omega_m)$, respectively. Let us note that the modes specified by n and $-n$ are degenerate and the further results are identical for them. We assume a cylindrical symmetry of the system under consideration so that these modes do not interact with each other.

The field $\mathbf{E}_a(\mathbf{R},t)$ originating from the contribution (3) induces the dipole moments in the ACs which satisfy the optical Bloch equations. By assuming the smallness of the frequency detuning, $\Delta = \omega_m - \omega_{21}$, and applying the rotating wave approximation, one comes to the equations for the macroscopic polarization, $\mathbf{P}(t) = \mathbf{P}^{(-)}(t)\exp(i\omega_m t) + \mathbf{P}^{(+)}(t)\exp(-i\omega_m t)$, and population difference density D of the AC ensemble:

$$\dot{\mathbf{P}}^{(+)}(\mathbf{R}) = -(\gamma_\perp - i\Delta)\mathbf{P}^{(+)}(\mathbf{R}) - \frac{i}{\hbar}|\mu_{12}|^2 D(\mathbf{R})\mathbf{E}_a^{(+)}(\mathbf{R},t), \quad \mathbf{P}^{(-)} = \mathbf{P}^{(+)*}, \quad (4)$$

$$\dot{D}(\mathbf{R}) = -\gamma_\parallel(\mathbf{R})[D(\mathbf{R}) - D_0] + \frac{2i}{\hbar}[\mathbf{P}^{(-)}(\mathbf{R})\mathbf{E}_a^{(+)}(\mathbf{R},t) - \mathbf{P}^{(+)}(\mathbf{R})\mathbf{E}_a^{(-)*}(\mathbf{R},t)]. \quad (5)$$

Here the radius vector $\mathbf{R} = (r,\theta,z)$ specifies a point inside the NW, $\gamma_\parallel$ is the spontaneous relaxation rate which takes into account the Purcell effect, $D = \rho(n_2 - n_1)$ with ρ being the number density of the ACs, n_1 and n_2 are the populations of the ground and excited states, respectively, and D_0 is its equilibrium value.

Up to this point, the derivation follows in general the conventional laser theory [3]. For the next step, one needs to obtain a relation between the polarization $\mathbf{P}$ and the electric field $\mathbf{E}_a$ to have a closed set of equations. This task is solved in the semiclassical laser theory by using an assumption that both the field and polarization amplitudes are slowly varying functions of both time and coordinates, which is not applicable to a nanocavity. Instead, we use a rigorous relation between $\mathbf{E}$ and $\mathbf{P}$ expressed in terms of the field susceptibility tensor $\bar{\mathbf{F}}$ [12]:

$$\mathbf{E}^{(+)}(\mathbf{R},t) = \int \bar{\mathbf{F}}(\mathbf{R},\mathbf{R}_0;\omega_m)\mathbf{P}^{(+)}(\mathbf{R}_0,t)d\mathbf{R}_0, \quad (6)$$

where the integral is taken over the region occupied by ACs and we have neglected the effect of retardation. Let us note that the quantity $\bar{\mathbf{F}}(\mathbf{R},\mathbf{R}_0;\omega)$ can be derived from the dyadic Green's function of the system and Eq. (6) takes place for a nanolaser of arbitrary geometry. The field susceptibility for points inside a NW includes two contributions [12]. One of them originates from the Hertz potential which has the only nonzero component parallel to the NW axis, Ψ. The other one is represented by the Hertz potential perpendicular to the axis, Φ. The latter contribution is essential only at the points close to the ends, at distances of the order of the wavelength in the NW material, $\lambda/\sqrt{\epsilon_2} = 2\pi c/(\omega\sqrt{\epsilon_2})$ with c being the speed of light in vacuum. For long NWs such that $L \gg \lambda/\sqrt{\epsilon_2}$, the contribution of the facet regions to the total field can be neglected. The further consideration concerns such long nanowires, which is the typical case for many experiments.

To find the evolution of the field in the lasing mode, one has to solve Eqs. (4)–(6) simultaneously. In this paper, we shall consider the two key problems of the laser theory: (i) the lasing condition and (ii) the steady-state regime of operation.

In the first case, to investigate the stability of the system, we write Eqs. (4) and (5) for small deviations $\delta\mathbf{E}_a^{(\pm)}$, $\delta\mathbf{P}^{(\pm)}$, and δD, keeping only the linear terms in them. The so-obtained equations can be solved by applying the Laplace transform in time. As a result, one comes to a set of coupled integral equations for the Laplace transformed electric-field deviations. Using the fact that in the considered approximation the kernel in the integral equation (6) is degenerate, one can readily reduce the problem to a set of two linear homogeneous algebraic equations (see Appendix A for details). Equating the determinant of this set to zero, one finds the condition imposed on the Laplace transformation variable, $s = \sigma + i\Omega$, that it has a nontrivial solution which implies nonzero values of $\delta\mathbf{E}_a^{(\pm)}$. The condition that the corresponding $\sigma > 0$ means that the system is unstable and any initial fluctuations in the field will lead to its exponential growth in time, i.e., to lasing.

This criterion is simplified in the case where the transition dipole moments in all ACs are aligned along the NW axis, z [13]. Then the condition of existence of a nontrivial solution for $\delta\mathbf{E}_a^{(\pm)}$ is reduced to

$$\chi(s)\mathcal{F}_0 = 1, \quad (7)$$

with

$$\mathcal{F}_0 = 4\pi\int_{-L/2}^{L/2}\int_0^a F_{a,zz}^+(r;r,z;\omega_m,\beta_a)u(z)rdrdz \equiv \int_{-L/2}^{L/2} K(z)dz, \quad (8)$$

where $F_{a,zz}^+$ is the contribution of the lasing mode in the field susceptibility tensor, $u(z) = \cos\beta_a z$ for even and

$u(z) = i \sin \beta_a z$ for odd Fabry-Pérot modes, and

$$\chi(s) = \frac{|\mu_{12}|^2}{\hbar} \frac{D_0}{\Delta + i(s + \gamma_\perp)}. \tag{9}$$

Let us note that $\chi(0)$ is the linear optical susceptibility of the gain medium.

Taking into account that, for the Fabry-Pérot modes of certain parity with numbers $m \gg 1$, $\beta_{am} \approx 2\pi m/L$ [10], one concludes from Eq. (8) that the nanolaser operation in the mth mode is governed by the mth Fourier coefficient of the field susceptibility averaged over the NW cross section.

Separating the real and imaginary parts in Eq. (7), one finds that σ is positive if

$$\eta \equiv (n_2 - n_1)\frac{\Gamma_a}{\gamma_\perp} > 1, \tag{10}$$

with $\Gamma_a = (1/\hbar)|\mu_{12}|^2 \rho \mathrm{Im}\mathcal{F}_0$, which gives the condition for the lasing threshold [15]. The quantity $(n_2 - n_1)\Gamma_a$ in this inequality describes the rate of the field amplification. Being divided by the mode group velocity, it can be considered as the modal gain. The other equation which follows from here describes the frequency pulling effect for the lasing frequency, $\omega_l = \omega_m - \Omega$:

$$\omega_l - \omega_{21} = \frac{1}{\hbar}|\mu_{12}|^2 D_0 \mathrm{Re}\mathcal{F}_0. \tag{11}$$

To obtain the laser equations in the steady-state regime, we assume that the inversion D acquires a constant value and perform the Laplace transform of Eqs. (4) and (6) in time. Then, Eq. (7) still holds if one makes the substitutions $\sigma = 0$ and $\mathcal{F}_0 \to \mathcal{F}$ with

$$\mathcal{F} = 4\pi \int_{-L/2}^{L/2} \int_0^a F^+_{a,zz}(r,r,z) D_r(r,z) u(z) r\,dr\,dz \tag{12}$$

and

$$D_r(r,z) = \left[1 + \frac{E_0^2}{E_s^2}\frac{\gamma_\perp^2}{\Delta^2 + \gamma_\perp^2} J_n^2(q_{20}r)|u(z)|^2\right]^{-1}, \tag{13}$$

where $E_0 \equiv E^{(+)}_{+z}(r = 0)$, $E_s = \hbar(\gamma_\perp \gamma_\parallel)^{1/2}/(4|\mu_{12}|)$ is the saturation field, and $q_{20} = \sqrt{(\omega/c)^2 - \beta_a^2}$. The intensity of the generated field is determined by the condition

$$\frac{1}{\hbar}|\mu_{12}|^2 D_0 \mathrm{Im}\mathcal{F} = \gamma_\perp. \tag{14}$$

The frequency pulling effect in the steady-state regime is given by Eq. (11), with $\mathcal{F}_0$ replaced by $\mathcal{F}$.

Let us consider the lasing threshold condition, given by Eq. (10), in some more detail. In the same manner as in the conventional laser theory, it determines the population inversion, $n_2 - n_1$, at which the system starts to lase. The factor at the inversion, Γ_a, is the averaged contribution of the lasing mode to the field susceptibility. For even modes, it is related to the Purcell factor [15]. As follows from Ref. [12], the latter quantity can be negative for certain positions of the emitter in the NW. This gives a hint that if the distribution of ACs is such that Γ_a is negative, then the lasing condition can be realized when $n_1 > n_2$, i.e., without a population inversion.

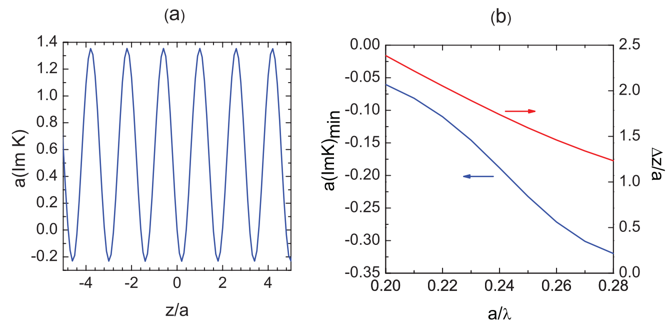

FIG. 1. (Color online) (a) Dependence of ImK, given by Eq. (8), for the TM_{01} mode on the dimensionless z coordinate in the range $-5 \leqslant z/a \leqslant 5$ for the odd Fabry-Pérot mode which exists for $L/a \approx 499$, $a/\lambda = 0.25$. (b) Dependence of the minimum value of ImK and the separation between the adjacent minima, Δz, on the dimensionless wavelength. L is chosen in the range $498 \leqslant L/a \leqslant 501$ so that there exists an odd Fabry-Pérot mode.

III. NUMERICAL RESULTS

We illustrate the general theory developed above by the consideration of a NW which supports a single Fabry-Pérot mode associated with the waveguide transverse magnetic TM_{01} mode ($n = 0$) and contains ACs with the transition dipole moments parallel to the z axis. In such a case, the field inside the NW is described by a single coefficient, $a^e_z(\beta)$. The further calculations have been carried out for $\epsilon_1 = 1$ and $\epsilon_2 = 6$ which is close to the dielectric function of GaN, ZnO, and CdS at the lasing frequencies. To find a proper design of the NW for lasing without inversion (LWI), we plot the imaginary part of the integrand in Eq. (8), Im$K(z)$. This quantity is shown in Fig. 1 for one of the odd Fabry-Pérot modes. It is a sinusoidal function of z with the period given by π/β_a, which is negative for certain intervals of z.

Let us assume now that the ACs are distributed only within thin layers at $z = z_i$ ($i = 1, \ldots, N$) perpendicular to the NW axis and that $K(z)$ does not change essentially within a layer. Then the quantity $\mathcal{F}_0$ can be calculated as $\mathcal{F}_0 \approx d \sum_{i=1}^N K(z_i)$, with d being the layer thickness. Choosing the positions of the layers in the regions where Im$K(z) < 0$, one ensures that Im$\mathcal{F}_0 < 0$ and hence the quantity Γ_a is negative. In the following, we assume that all the quantities $K(z_i)$ are equal to each other.

Let us estimate whether the lasing condition, given by Eq. (10), can be fulfilled without a population inversion. Taking the realistic parameters for a gain medium [16]: $\gamma_\perp = 10^{13}\ \mathrm{s}^{-1}$, $|\mu_{12}| = 1.5 \times 10^{-17}$ esu, $\rho = 2.4 \times 10^{20}\ \mathrm{cm}^{-3}$, and assuming $n_2 \approx 0$, $a = 100$ nm, $d = 20$ nm, one finds for $a\mathrm{Im}K(z_i) = -0.10$ that this criterion is met if $N \geqslant 10$. If the doping concentration is increased ten times, then LWI is possible even when there is only a single active layer.

The field E_0 generated in the steady-state regime in such a nanolaser is found from Eq. (14). The corresponding number of the mode quanta, N_q, can be calculated from the mode electromagnetic field energy. This quantity, which determines the mode intensity, is shown in Fig. 2 for different values of the excess above the threshold in Eq. (10), η.

The frequency pulling effect near the threshold, given by Eq. (11), is illustrated in Fig. 3. The frequency shift

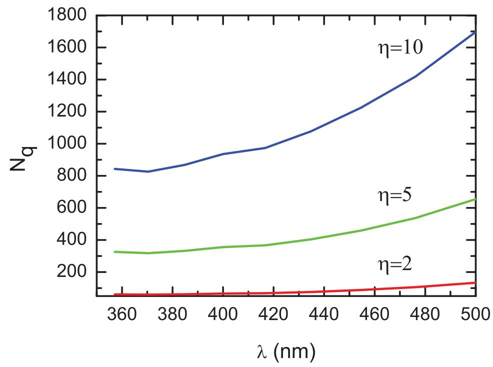

FIG. 2. (Color online) Number of the lasing mode quanta generated in the steady-state regime vs the mode wavelength for different parameters η, given by Eq. (10). $\Delta = 0$, $a = 100$ nm; L is chosen as in Fig. 1(b).

is normalized to the quantity $(1/\hbar)|\mu_{12}|^2 D_0$, which equals 5.4×10^{13} s^{-1} for the parameters given above, which is comparable with $\gamma_\perp$. This quantity is a sinusoidal function of the active layer position, z. The regions where ImK is positive correspond to usual lasing with inversion, whereas those where ImK is negative correspond to LWI. One can see that at a certain position, $z/a \approx 1.2$, the frequency pulling is absent [the other zeros do not satisfy the lasing condition, given by Eq. (10)]. In the steady-state regime, the frequency pulling is even smaller due to the saturation effect.

IV. DISCUSSION

The criterion (10) can be understood as the requirement that, in order to achieve lasing, the rate of the field amplification should exceed the rate of the polarization decay. The amplification rate, in turn, is a result of integration over the whole NW volume. As follows from Eq. (8), the areas where ImK is positive contribute to the quantity Γ_a with a plus sign, whereas those where ImK is negative contribute with a minus sign. Let us assume now that there is no population inversion, i.e., $n_2 < n_1$. Then the areas of positive ImK contribute to the field attenuation, while those where ImK is negative contribute to the field amplification. When the active medium occupies the whole NW volume, there are alternating layers of attenuation and amplification, with the contribution of the first ones being predominant. If, however, one fabricated active centers only within the amplification areas, the net effect would be the field amplification. The magnitude of this effect decreases when the NW radius increases. For large a such that $a \gg \lambda/\sqrt{\epsilon_2}$, the function Im$K$ takes only non-negative values (not shown).

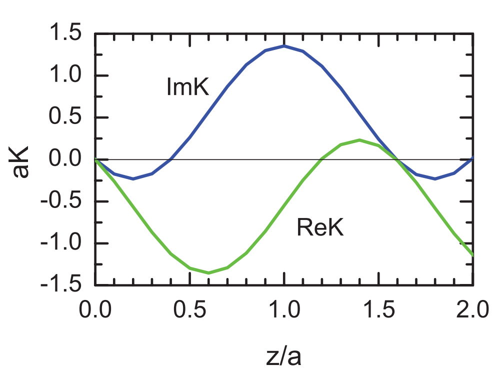

FIG. 3. (Color online) The lasing frequency pulling, given by Eq. (11), normalized to the quantity $(1/\hbar)|\mu_{12}|^2 D_0$ as a function of the dimensionless z coordinate (green line). For comparison, the quantity ImK is also shown (blue line). $a/\lambda = 0.25$; the other conditions are as in Fig. 1.

This feature stems from an effect that is well known in cavity quantum electrodynamics. When a radiating atom is located nearby a reflective surface, its radiative decay rate demonstrates oscillating behavior with the atom-surface distance, which is essential at distances $\lesssim \lambda$ [17,18]. A similar effect occurs when an atom is placed between two parallel surfaces [19,20], inside or nearby a cylindrical waveguide [21], or inside a cylindrical cavity [12]. It originates from the interference between the field emitted by the atom and the field reflected from the surface. The result of this interference depends on the optical path difference and hence on the location of the atom relative to the surface. As a consequence, the range of distances where the decay rate γ is larger than its value far from the surface, γ_0, alternate with the ranges where $\gamma < \gamma_0$. The total decay rate in the vicinity of the surface can be represented as $\gamma = \gamma_0 + \gamma_s$, where γ_s is the contribution due to the reflected field. Thus, the ranges where $\gamma > \gamma_0$ correspond to positive γ_s, whereas those where $\gamma < \gamma_0$ correspond to negative γ_s.

The quantity γ_s, in turn, is determined by the imaginary part of the field susceptibility associated with the influence of the surface, F_s [18]. On the other hand, positive and negative values of Im(F_s) correspond to the dipole moment attenuation and amplification, respectively (see Appendix B). One can conclude, therefore, that in the ranges where $\gamma < \gamma_0$, the atom dipole moment is amplified by the field reflected by the surface. The amplified dipole moment leads to a larger reflected field, and so on. In other words, an atom located in the amplification range experiences a positive feedback for its dipole moment provided by the reflective surface, i.e., the atom and the surface form loop gain. This effect has numerous analogues in different fields of science and engineering [22]. Since usually $|\gamma_s| < \gamma_0$, this gain does not lead to instability.

Turning to the case of a cavity, one has to take into account the cavity mode structure given by the function $u(z)$ and the distribution of ACs over the NW volume. Therefore, the above arguments concerning the sign of Im(F_s) should be replaced by those on the sign of ImK. Similarly to an atom located in an amplification range, ACs within an amplification area in a NW experience loop gain. The magnitude of this effect is proportional to the number of the active centers involved in the loop and, under certain conditions, this gain can lead to instability (see Appendix B).

The considered gain is, however, different from the one which takes place in conventional lasers. The loop gain which occurs under the population inversion condition originates from the light intensity reflected back to the cavity interior and causes the stimulated emission (in this sense, it can be called the *intensity* gain). The gain which is predicted in this paper for the LWI effect stems from the polarization amplification (it can be called the *polarization* gain) and is a classical effect.

In the discussion above, we have implied, for the sake of simplicity, that the active medium is represented by ACs which do not interact with each other. The further increase of the doping level will lead to the formation of quantum wells comprised of the active material. This should ensure even larger excess over the lasing threshold for LWI. Recently, the possibility to grow such structures in a nanowire has been demonstrated for multi-quantum-well nanowire lasers based on InGaN/GaN quantum wells [23].

LWI in nanowires predicted in this paper has its analog for atomic systems in gas media [24]. However, there is a principal difference. In atomic systems, it can be achieved for specially prepared atomic states due to a quantum coherence between two atomic transitions. This effect can be interpreted as the parametric instability in the system of two coupled oscillators associated with *two atomic transitions of the same atom* [25]. The laser cavity does not play any role in that. In NWs, lasing without inversion can take place in specially designed nanocavities. Such an effect can be understood as the collective parametric instability in a system of oscillators associated with *a single transition* in the active centers where the coupling is *mediated by the cavity* due to the reflected field (see Appendix B). As it is known, such a system can exhibit instabilities which are different from those of a single oscillator [26].

Let us note that the ACs located within a thin active layer contribute to the lasing mode with the same phase. Besides that, the NW design which provides the LWI effect ensures that the contributions of different active layers to the lasing field are determined by the same quantity, $K(z_i)$, and hence they are also in phase with each other. In other words, the distribution of ACs over the NW volume forms a coherent ensemble of oscillating dipoles. In such a case, even a weak incoherent pumping will lead to lasing.

V. CONCLUSION

In conclusion, we have derived the semiclassical nanowire laser equations from first principles. Basing on the developed theory, we have predicted the effect of lasing without inversion in nanowires. We have also designed a nanolaser cavity in which LWI is possible. The estimates have shown that this effect can be readily observed in experiment. Its implementation in nanowires will open up new prospects for the development of nanolasers which do not require powerful pumping. This will allow one to avoid the parasitic effect of amplified spontaneous emission. Let us also note that a similar theory can be developed for surface plasmon amplification in a nanowire-based spaser [27].

APPENDIX A: DERIVATION OF THE LASING CONDITION

To derive the laser equations, we assume that only a single mode labeled by a and having the propagation constant β_a is relevant to lasing. Its contribution to the total field can be written as

$$\mathbf{E}_a(\mathbf{R},t) = \mathbf{E}_a^{(-)}(\mathbf{R},t)e^{i\omega_m t} + \mathbf{E}_a^{(+)}(\mathbf{R},t)e^{-i\omega_m t}, \tag{A1}$$

where the quantities $\mathbf{E}_a^{(\pm)}$ are given by Eq. (3).

The integral in Eq. (6) contains the contribution of the lasing mode in the form of Eq. (3) that gives the relation

$$\mathbf{E}_\pm^{(+)}(r,t)e^{-in\theta} = \int_{-L/2}^{L/2}\int_0^{2\pi}\int_0^a \bar{\mathbf{F}}_a^\pm(r,\theta,z;r_0,\theta_0,z_0;\omega_m) \times \mathbf{P}^{(+)}(r_0,\theta_0,z_0,t)r_0 dr_0 d\theta_0 dz_0, \tag{A2}$$

where

$$\bar{\mathbf{F}}_a(z) = \bar{\mathbf{F}}_a^- e^{-i\beta_a z} + \bar{\mathbf{F}}_a^+ e^{i\beta_a z} \tag{A3}$$

with

$$\bar{\mathbf{F}}_a^\pm(r,\theta;r_0,\theta_0,z_0;\omega_m) = \pm\frac{i}{2}\mathrm{Res}\tilde{\bar{\mathbf{F}}}(r,\theta;r_0,\theta_0,z_0;\omega_m,\pm\beta_a) \tag{A4}$$

being the contribution of the lasing mode a into the field susceptibility tensor. Here, $\tilde{\bar{\mathbf{F}}}(\beta)$ is the Fourier transform of $\bar{\mathbf{F}}(z)$, and $\mathrm{Res}\tilde{\bar{\mathbf{F}}}(\pm\beta_a)$ notates the tensor composed of the residues of $\tilde{\bar{F}}_{ij}(\beta)$ $(i,j = r,\theta,z)$ at the specified poles.

The Fourier transform of the field susceptibility tensor originating from the potential Ψ, $\tilde{F}_{ij}^\psi$, can be written as follows [12]:

$$\tilde{F}_{ij}^\psi(r,\theta;r_0,\theta_0,z_0;\beta) = \frac{4\pi}{\epsilon_2}e^{-in\theta}\sum_{\sigma=e,m} H_i^\sigma(r;\beta) \times a_j^\sigma(r_0,\theta_0,z_0;\beta), \tag{A5}$$

where the coefficients a_j^σ describe transverse magnetic (TM, $\sigma = e$) and transverse electric (TE, $\sigma = m$) field components associated with a point dipole oriented along the ort $\mathbf{e}_j$ $(j = r,\theta,z)$, and the coefficients $H_i^\sigma(r;\beta)$ have the form

$$H_r^e(r;\beta) = \frac{i\beta}{q_2}J_n'(q_2 r), \tag{A6}$$

$$H_\theta^e(r;\beta) = \frac{\beta n}{q_2^2 r}J_n(q_2 r), \tag{A7}$$

$$H_z^e(r;\beta) = J_n(q_2 r), \tag{A8}$$

$$H_r^m(r;\beta) = \frac{\omega n}{c q_2^2 r}J_n(q_2 r), \tag{A9}$$

$$H_\theta^m(r;\beta) = -\frac{i\omega}{c q_2}J_n'(q_2 r), \tag{A10}$$

$$H_z^m(r;\beta) = 0, \tag{A11}$$

where J_n is the Bessel function of the first kind and $q_2 = \sqrt{(\omega/c)^2\epsilon_2 - \beta^2}$. The coefficients a_j^e and a_j^m satisfy the integral equation

$$\hat{M}(\beta)\vec{A}_j(\beta) - \frac{1}{2\pi}\int_C [e^{-i(\beta-\beta')(L/2-z_0)} - e^{i(\beta-\beta')(L/2+z_0)}] \times \hat{N}(\beta,\beta')\vec{A}_j(\beta')d\beta' = -\vec{B}_j(\beta), \tag{A12}$$

where the explicit form of the matrices $\hat{M}$ and $\hat{N}$ and the column vector $\vec{B}_j$ can be found in Refs. [10] and [12], respectively, and

$$\vec{A}_j(\beta) = \exp(i\beta z_0)\begin{pmatrix} a_j^e(\beta) \\ a_j^m(\beta) \\ b_j^e(\beta) \\ b_j^m(\beta) \end{pmatrix}, \tag{A13}$$

with b_j^e and b_j^m being the coefficients which determine the TM and TE field components, respectively, outside the nanowire. The integral equation (A12) is exact and, in principle, can be solved numerically. Its iterative solution corresponds to taking into account successive reflections from the nanowire ends. In this work, we assume that the nanowire aspect ratio $L/(2a)$ is large so that Eq. (A12) allows an approximate solution (see Refs. [9,10,12] for details).

To investigate the stability of the system, we write Eqs. (4) and (5) for small deviations $\delta\mathbf{E}_a^{(\pm)}$, $\delta\mathbf{P}^{(\pm)}$, and δD, keeping only the linear terms in them. The so-obtained equations can be solved by applying the Laplace transform in time. Assuming the zero initial conditions, one comes to a set of integral equations,

$$\delta\mathcal{E}_{+,i}(r,s)e^{-in\theta} = i\chi(s)\sum_{j=r,\theta,z}\int_{-L/2}^{L/2}\int_0^{2\pi}\int_0^a \mathrm{Res}[\tilde{F}^+_{a,ij}(r,\theta;r_0,z_0;\omega_m,\beta_a)]\delta\mathcal{E}_{+,j}(r_0,s)u(z_0)r_0dr_0d\theta_0dz_0, \tag{A14}$$

where $\delta\vec{\mathcal{E}}_+(s)$ is the Laplace transform of $\delta\mathbf{E}_+^{(+)}(t)$, $s = \sigma + i\Omega$, $u(z) = \cos\beta_a z$ for even and $u(z) = i\sin\beta_a z$ for odd Fabry-Pérot modes, and $\chi(s)$ is given by Eq. (9).

Taking into account Eq. (A5) and introducing the quantity

$$Y_\mu = \sum_{i=r,\theta,z}\int_{-L/2}^{L/2}\int_0^{2\pi}\int_0^a \mathrm{Res}\big[a_i^\mu(r,\theta,z;\beta_a)\big] \times \delta\mathcal{E}_{+,i}(r,s)e^{-in\theta}u(z)rdrd\theta dz, \tag{A15}$$

one obtains a set of two linear homogeneous algebraic equations relative to Y_μ,

$$Y_\mu = \frac{4\pi i}{\epsilon_2}\chi(s)\sum_\sigma R_{\mu\sigma}Y_\sigma, \quad \mu,\sigma = e,m, \tag{A16}$$

where

$$R_{\mu\sigma} = \sum_i\int_{-L/2}^{L/2}\int_0^{2\pi}\int_0^a H_i^\sigma(r;\beta_a)\mathrm{Res}\big[a_i^\mu(r,\theta,z;\beta_a)\big] \times e^{-in\theta}u(z)rdrd\theta dz. \tag{A17}$$

Equating the determinant of this set to zero, one finds the condition imposed on s that it has a nontrivial solution, which implies nonzero values of $\delta\mathcal{E}_{+,i}(r,s)$. The condition that the corresponding $\sigma > 0$ means that the system is unstable and any initial fluctuations in the field will lead to its exponential growth in time, i.e., to lasing.

The set of equations (A16) is simplified if the transition dipole moments in all active centers are aligned along the nanowire axis z. Then, only the coefficients $a_z^\sigma(\beta)$ are nonzero, which leads to the equalities $R_{\mathrm{em}} = R_{\mathrm{mm}} = 0$. As a result, the condition of existence of a nontrivial solution is reduced to

$$\chi(s)\mathcal{F}_0 = 1, \tag{A18}$$

where

$$\mathcal{F}_0 = 4\pi\int_{-L/2}^{L/2}\int_0^a F^+_{a,zz}(r;r,z;\omega_m,\beta_a)u(z)rdrdz = 4\pi\int_0^a J_n^2(q_{20}r)rdr\int_{-L/2}^{L/2}F^+_{a,zz}(0;0,z;\omega_m,\beta_a)u(z)dz, \tag{A19}$$

$q_{20} = \sqrt{(\omega/c)^2 - \beta_a^2}$, and we have taken into account that the quantity $F^+_{a,zz}$ does not depend on θ. Equation (A18) determines the values of $s = \sigma + i\Omega$ at which nonzero deviations $\delta\mathbf{E}_a^{(\pm)}(t)$ can exist. If the corresponding σ is positive, then these deviations grow exponentially in time.

APPENDIX B: CLASSICAL ANALOGY OF LASING WITHOUT INVERSION

One can follow an analogy between the lasing without inversion in a system of active centers located in a cavity and a collective instability in a system of coupled parametric oscillators. The behavior of an atomic system in a cavity can be modeled by an oscillating dipole which interacts with its own field reflected from the cavity walls. Such an approach has been proven to be an efficient model which describes the quantum electrodynamics of an atom in the vicinity of a reflecting surface [17]. The equation of motion of the dipole μ reads as

$$\ddot{\mu} + \gamma_0\dot{\mu} + \omega_0^2\mu = \frac{e^2}{m}E_R, \tag{B1}$$

where ω_0 is the frequency of free dipole oscillations, γ_0 is the damping constant which has the sense of inverse lifetime, e and m are the charge and the effective mass of the dipole, respectively, and E_R is the reflected electric field at the dipole position. The reflected field can be expressed in terms of the field susceptibility part related to the surface, F_s, as

$$E_R = F_s\mu, \tag{B2}$$

which leads to the equation

$$\ddot{\mu} + \gamma_0\dot{\mu} + \omega^2\mu = 0, \tag{B3}$$

where we have introduced the renormalized frequency

$$\omega^2 = \omega_0^2 - \frac{e^2}{m}F_s = \omega_0^2 - \frac{e^2}{m}\mathrm{Re}(F_s) - i\frac{e^2}{m}\mathrm{Im}(F_s). \tag{B4}$$

Applying the Laplace transform to Eq. (B3) with a zero initial condition, one obtains that the transformed equation has a nontrivial solution, $\mu(s)$, if

$$s = \frac{-\gamma_0 - \sqrt{\gamma_0^2 - 4\omega^2}}{2}, \tag{B5}$$

where the minus sign in front of the square root is chosen to ensure the time behavior at $F_s = 0$ $\mu(t) \sim \exp(-i\omega_0 t)$. Assuming a reasonable approximation that $\gamma_0^2, (e^2/m)|F_s| \ll \omega_0^2$, one finds

$$\mathrm{Re}(s) \approx -\frac{\gamma_0}{2} - \frac{e^2}{2m\omega_0}\mathrm{Im}(F_s). \tag{B6}$$

Here, the quantity $(e^2/m\omega_0)\mathrm{Im}(F_s)$ can be identified with the surface contribution to the relaxation rate, γ_s. Equation (B6) implies that if $\mathrm{Im}(F_s) < 0$, then the field reflected from the surface introduces the dipole amplification. If, besides that, $-(e^2/m\omega_0)\mathrm{Im}(F_s) > \gamma_0$, then the dipole is parametrically unstable and any nonzero initial value of the dipole amplitude will lead to its exponential increase in time.

For a system of M oscillating dipoles interacting with the cavity, the equations of motion are given by

$$\ddot{\mu}_i + \gamma_0\dot{\mu}_i + \omega_0^2\mu_i = \frac{e^2}{m}\sum_{j=1}^{M} F_{ij}\mu_j, \quad i = 1,2,\ldots,M, \tag{B7}$$

where the quantities F_{ij} describe the reflected field of the jth dipole at the position of the ith dipole.

Let us assume that the quantities F_{ij} are factorized as

$$F_{ij} = g_i h_j \tag{B8}$$

[this corresponds to a degenerate kernel in the integral equation (6)]. Then one comes to the equation

$$\ddot{\mathcal{M}} + \gamma_0\dot{\mathcal{M}} + {\omega_0}^2\mathcal{M} = \frac{e^2}{m}\mathcal{F}\mathcal{M}, \tag{B9}$$

where

$$\mathcal{M} = \sum_{i=1}^{M} h_i\mu_i \tag{B10}$$

and

$$\mathcal{F} = \sum_{i=1}^{M} F_{ii}, \tag{B11}$$

which is equivalent to Eq. (B1). One obtains from here that the system described by Eqs. (B7) is parametrically unstable if $\mathrm{Im}(\mathcal{F}) < 0$ and $-(e^2/m\omega_0)\mathrm{Im}(\mathcal{F}) > \gamma_0$. Let us note that in this case, the quantity $\mathcal{F}$ is proportional to the number of dipoles in the system, M.

[1] C. Z. Ning, in *Advances in Semiconductor Lasers*, edited by J. J. Coleman, A. C. Bryce, and C. Jagadish, Semiconductors and Semimetals, Vol. 86 (Academic, Burlington, 2012).

[2] C. Z. Ning, Phys. Status Solidi B **247**, 774 (2010).

[3] H. Haken, *Light*, Vol. 2 (North-Holland, Amsterdam, 1985).

[4] A. Mock, J. Opt. Soc. Am. B **27**, 2262 (2010).

[5] *Cavity Quantum Electrodynamics*, edited by P. R. Berman (Academic, Boston, 1994).

[6] H. E. Türeci, A. D. Stone, and B. Collier, Phys. Rev. A **74**, 043822 (2006).

[7] H. E. Türeci, L. Ge, S. Rotter, and A. D. Stone, Science **320**, 643 (2008).

[8] J. Andreasen, A. A. Asatryan, L. C. Botten, M. A. Byrne, H. Cao, L. Ge, L. Labonté, P. Sebbah, A. D. Stone, H. E. Türeci, and C. Vanneste, Adv. Opt. Photon. **3**, 88 (2011).

[9] V. G. Bordo, Phys. Rev. B **78**, 085318 (2008); **79**, 039901(E) (2009).

[10] V. G. Bordo, Phys. Rev. B **81**, 035420 (2010).

[11] V. G. Bordo, Phys. Rev. B **84**, 075465 (2011).

[12] V. Bordo, J. Opt. Soc. Am. B **29**, 1799 (2012).

[13] The results obtained below are also valid when the projections of the transition dipole moments onto the transverse plane are distributed randomly.

[14] Different NWs for which lasing has been demonstrated, e.g., those grown from GaN, ZnS, CdS, and ZnO, have a shape which can be well described by a circular cylinder model (see Ref. [1]). This justifies the usage of a single angular momentum wave function.

[15] Let us note that for even Fabry-Pérot modes, the quantity Γ_a is proportional to the Fourier coefficient of the Purcell factor averaged over the NW cross section.

[16] M. I. Stockman, J. Opt. **12**, 024004 (2010).

[17] R. R. Chance, A. Prock, and R. Silbey, J. Chem. Phys. **60**, 2744 (1974).

[18] J. M. Wylie and J. E. Sipe, Phys. Rev. A **30**, 1185 (1984).

[19] S. M. Dutra and P. L. Knight, Phys. Rev. A **53**, 3587 (1996).

[20] H. Nha and W. Jhe, Phys. Rev. A **54**, 3505 (1996).

[21] H. Nha and W. Jhe, Phys. Rev. A **56**, 2213 (1997).

[22] See, e.g., M. A. Copeland, *Electrical Instability*, AccessScience (McGraw-Hill, New York, 2012), AccessScience.com.

[23] F. Qian, Y. Li, S. Gradečak, H.-G. Park, Y. Dong, Y. Ding, Z. L. Wang, and C. M. Lieber, Nat. Mater. **7**, 701 (2008).

[24] J. Mompart and R. Corbalán, J. Opt. B: Quantum Semiclass. Opt. **2**, R7 (2000).

[25] O. Kocharovskaya and P. Mandel, Phys. Rev. A **42**, 523 (1990).

[26] I. Bena, C. Van den Broeck, R. Kawai, M. Copelli, and K. Lindenberg, Phys. Rev. E **65**, 036611 (2002).

[27] S.-Y. Liu, J. Li, F. Zhou, L. Gan, and Z.-Y. Li, Opt. Lett. **36**, 1296 (2011).