

Photon-phonon laser on crystalline silicon: a feasibility study

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Abstract. We discuss a feasibility of photon-phonon laser action in bulk silicon with electron population inversion. It is well known, that only direct gap semiconductors are used as an active medium in optical lasers. In indirect gap semiconductors, such as crystalline silicon, the near-to-gap radiative electron transitions must be assisted by emission or absorption of phonons to conserve the momentum. The rate of such two-quantum transitions is much less than in direct gap semiconductors, where the similar radiative transitions are single-quantum. As a result, the quantum efficiency of luminescence in silicon is too small to get it as a laser material. Numerous proposals to overcome this problem are aimed at increasing the rate of radiative recombination. We suggest enhancing the quantum efficiency of luminescence in silicon by stimulating the photon part of the two-quantum transitions by light from an appropriate external laser source. This allows us to obtain initially an external-source-assisted lasing in silicon and then a true photon-phonon lasing without any external source of radiation. Performed analysis revealed a number of requirements to the silicon laser medium (temperature, purity and perfection of crystals) and to the intensity of stimulating radiation. We discuss different mechanisms that may hinder the implementation of photon-phonon lasing in silicon.

1. Introduction

The possibility to exploit indirect band gap semiconductors as an active medium for lasers is discussed for many years. Already in the first publication on this subject [1], which appeared shortly after demonstration of the first semiconductor laser, it has been concluded that indirect band gap semiconductors are not suitable for light emitting devices and realizing a laser. In such semiconductors the bottom of the conduction band is not located directly above the top of the valence band in the momentum space of a crystal. This means that an electron near the conduction band edge can not to have the same momentum as a hole near the valence band edge. As a result, the near-to-gap radiative electron-hole recombination can only occur through two-quantum transitions with emission of a photon and simultaneous emission or absorption of a phonon to satisfy the laws of energy and momentum conservation. The rate of such two-quantum transitions is very low and therefore, indirect band gap semiconductors are highly inefficient as a host material for light sources.

Despite this disappointing conclusion researchers do not leave the efforts to find a solution to this problem [2–5]. The reason for such persistence is silicon – one of the most famous and attractive indirect band gap semiconductors. Silicon is the fundamental material used ubiquitously in the electronics industry. It is exploited as a substrate for most integrated circuits. Therefore, creation of silicon-based light sources (either light emitting diodes or lasers) would pave the way for closer integration of photonics and electronics.



The advent of nanotechnology has opened up entirely new possibilities for researchers. Optical properties of a tiny crystal with the size that are comparable to the electron de Broglie wavelength (for instance, less than 5 nm at room temperature) is fundamentally different from its bulk counterpart. When the dimensions are so small, quantum effects start to play an important role and can fully alter the properties of the original material.

The quantum confinement effects may offer a solution to the problem of silicon. According to the Heisenberg uncertainty principle, when a particle is spatially localized, its momentum becomes uncertain. This means that in low-dimensional indirect band gap semiconductors (nanocrystals, thin layered structures, porous materials), the tails of the wavefunctions of an electron and a hole can now partially overlap, allowing the quasi-direct transitions to occur and thus increasing the probability of radiative recombination.

A number of groups have presented the experimental data on the observation of an enhanced luminescence and even an optical gain in nanostructured silicon, namely, in nanocrystals [6, 7], in silicon on insulator superlattices [8] and in nano-porous silicon [9–11]. Soon afterwards, the efficient silicon-based light emitting diodes [12, 13] have been demonstrated.

Silicon laser is the most challenging goal for researchers and a number of important breakthroughs have been made in the past decade [14]. Among the impressive achievements are the first demonstration of a pulsed silicon Raman laser [15] and the first successful demonstration of a continuous-wave silicon Raman laser shortly thereafter [16]. It should be noted, however, that Raman laser is a specific type of laser where the light-amplification mechanism is stimulated Raman scattering. The optical pumping in Raman lasers does not produce a population inversion. The pumping radiation is rather converted into stimulated radiation in nonlinear laser medium. In contrast, most conventional lasers rely on stimulated electronic transitions between inversely populated quantum states. Silicon laser with electron population inversion remains still elusive despite the efforts of many researchers [17–22].

In this paper we discuss a new approach to the implementation of laser action in crystalline silicon – two-quantum photon-phonon laser. Dynamics of the photon-phonon laser generation is investigated with aid of the rate equations. We formulate a set of requirements to the silicon laser medium (temperature, purity and perfection of crystals) and discuss different mechanisms that may hinder the development of photon-phonon lasing in silicon.

2. Dynamics of photon-phonon laser generation

Dynamics of the photon-phonon laser generation in a silicon crystal can be described by the rate equations for the electron concentration N_e in the conduction band, the photon concentration N_k in an optical mode, and for the phonon concentration N_q in an acoustic mode [23]. We restrict ourselves to the steady state solutions of the rate equations. One can see that there are two types of these solutions: below the phonon lasing threshold with $n_q = 0$ and above the phonon lasing threshold with $n_q \neq 0$. These solutions are shown in the figure 1 (curves of type I and type II, respectively) in the form of dependence of the normalized photon concentration n_k in the optical mode versus the normalized pumping rate W_p/W_0 with the normalized photon concentration n_0 of external stimulating radiation taken as a parameter of a family of curves.

It can be shown with the help of Routh-Hurwitz criterion that only increasing parts of the curves II in figure 1 correspond to the stable steady state solutions. Intersection in the point A between the stable branch of a curve II and a curve I with the same n_0 determines the phonon lasing pumping threshold $(W_p/W_0)_{th}$ and the threshold value for the normalized photon concentration $(n_k)_{th}$ inside the crystal. If the pumping is further increased the system goes on the stable branch of the curve II and the phonon laser action occurs. This leads to a growth of the phonon and the photon concentrations in the modes. If the pumping rate W_p/W_0 exceeds some threshold value W_s/W_0 , the external source of stimulating radiation can be switched off and the system goes from the point C₁ to the working point C

on the stable branch of the curve II with $n_0=0$. Thus, the system continues to operate with simultaneous lasing of both photons and phonons.

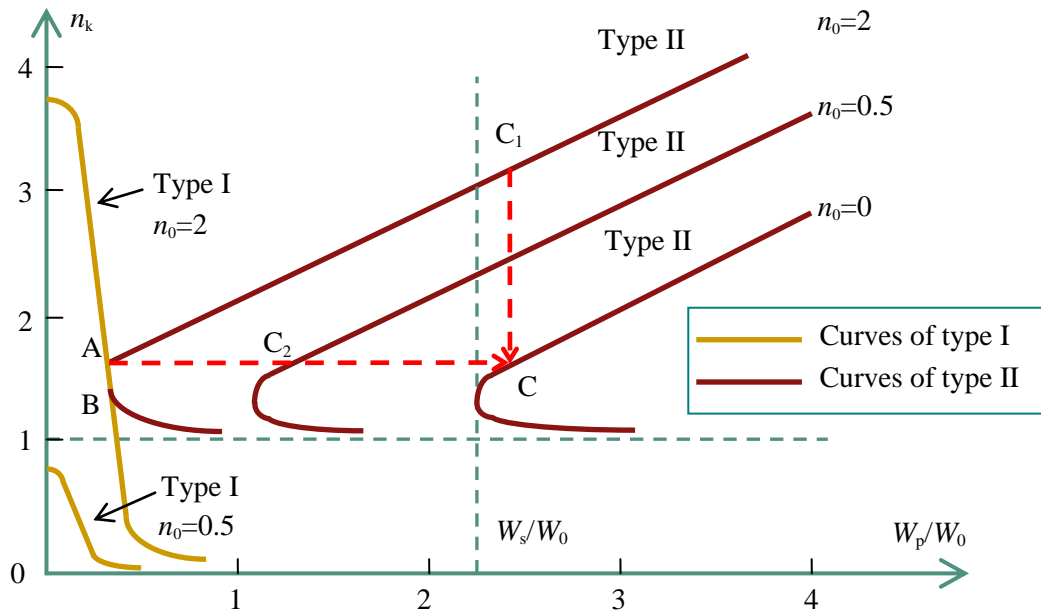


Figure 1. The steady state solutions of the rate equations.

The path $A \rightarrow C_1 \rightarrow C$ is not the only way to come to the working point C. Alternatively, we can use the path $A \rightarrow C_2 \rightarrow C$ shown in the figure 1. For this we should gradually reduce the intensity of external radiation according as the pumping rate increases so that the photon concentration $(n_k)_{th}$ in the optical mode remains constant.

Numerical estimates are collected in the table 1.

Table 1. Numerical estimates

Semicon- ductor	N , 10^{14} cm^{-3}	W_0 , $10^{25} \text{ cm}^{-3} \text{ s}^{-1}$	$(W_p/W_0)_{th}$, 10^{-3}	$(n_k)_{th}$	W_s/W_0	n_{ks}	n_{qs}	n_0
Si	1.6×10^2	5.6×10^2	4.2	1.6	2.4	3.0	2.4	2.0

N	the normalizing particle concentration
W_0	the normalizing pumping rate
$(W_p/W_0)_{th}$	the normalized threshold pumping rate for starting the phonon laser action
$(n_k)_{th}$	the normalized threshold photon concentration inside the crystal for starting the phonon laser action
W_s/W_0	the normalized threshold pumping rate for starting the simultaneous laser action of both photons and phonons
n_{ks}	the normalized photon concentration inside the crystal for starting the simultaneous laser action of both photons and phonons
n_{qs}	the normalized phonon concentration for starting the simultaneous laser action of both photons and phonons
n_0	the normalized photon concentration of external stimulating radiation

Returning from normalized to the physical variables, we obtain at the optimum photon energy of stimulating radiation, $\hbar\omega_{\mathbf{k}_0}=1.165$ eV, the following estimate for the threshold light intensity (at the point A in figure 1) $I = c\hbar\omega_{\mathbf{k}_0}N(n_k)_{\text{th}} = 2.7 \times 10^7$ W/cm², where $c = 0.88 \times 10^{10}$ cm s⁻¹ is the speed of light in silicon.

3. Feasibility discussion of the silicon photon-phonon laser

In this section we discuss different mechanisms that may hinder the implementation of photon-phonon lasing in silicon. Performed analysis revealed a number of requirements to the active laser medium (temperature, purity and perfection of crystals) and to the intensity of stimulating radiation. First of all, we need cryogenically cooled silicon crystals in order to reduce the anharmonic phonon-phonon scattering of TA phonons down to an acceptable level. The TA phonons are long-lived at low temperatures, since the selection rules forbid the phonon decay into two other phonons with lower energies [24]. Under these conditions, the principal mechanism of the TA phonon losses from an acoustic mode becomes Rayleigh scattering by isotope impurities. For silicon with natural isotopic abundances the inverse lifetime τ_i^{-1} of the TA phonon is estimated by [25] $\tau_i^{-1} = 7.2 \times 10^9$ s⁻¹. Further reduction of the phonon losses requires isotopically pure and perfect silicon crystals. The above obtained estimates presented in table 1 are based on the assumption that the natural concentration of isotopic impurities in silicon can be reduced by 4 orders of magnitude.

Even under such special conditions, the intensity of stimulating radiation, required to attain the photon-phonon lasing in bulk silicon, remains a rather high and may approach to the optical damage threshold. A study of optical resistance of silicon was made in connection with search for an upper limit for the light intensity restricting the performance of silicon Raman laser. It was found [26] that the optical damage threshold of crystalline silicon ranges from 1 GW/cm² to 4 GW/cm² depending on the radiation wavelength. In particular, at the operating wavelength of the silicon photon-phonon laser, $\lambda = 1.06$ μm , it is estimated to be 1 GW/cm². One can see that the above obtained threshold intensity of stimulating radiation, $I = 2.7 \times 10^7$ W/cm², is approximately two orders of magnitude lower and therefore cannot damage the silicon crystal. Furthermore, in an isotopically pure and perfect silicon crystal, where the phonon losses from an acoustic mode is mainly due to lattice absorption and due to phonon absorption in the anti-Stokes vibronic sideband, the intensity of stimulating radiation may be additionally reduced by an order of magnitude.

Another potentially detrimental process, that should be considered, is the two-photon absorption of stimulating radiation. This process which was not taken into account in the rate equations may have a significant impact on the dynamics of lasing, especially at a high optical intensity. Since the total energy of two photons of stimulated radiation is greater than the band gap energy of silicon, they can be simultaneously absorbed in indirect transitions with excitation of electrons from the valence band to the conduction band. The rate of the two-photon generation of free carriers in silicon was experimentally and theoretically investigated in numerous papers. To evaluate this rate, the coefficient β was introduced through the relation

$$\frac{dN_e}{dt} = \frac{\beta I^2}{2\hbar\omega_{\mathbf{k}_0}}. \quad (1)$$

The corresponding values of β were experimentally determined for silicon at different wavelengths and can be found in the review [27]. In particular, at the operating wavelength of the silicon photon-phonon laser, $\lambda = 1.06$ μm , the coefficient β is estimated to be 1.5 cm/GW. This leads to the following estimate $dN_e/dt = 2.9 \times 10^{24}$ cm⁻³s⁻¹ for the rate (1) at the threshold intensity of stimulating radiation $I = 2.7 \times 10^7$ W/cm². One can see that this value is an order of magnitude lower than the threshold rate of pumping $W_p = 2.3 \times 10^{25}$ cm⁻³s⁻¹ for starting the phonon lasing (point A in figure 1) and almost four orders of magnitude lower than the threshold rate of pumping $W_s = 1.3 \times 10^{28}$ cm⁻³s⁻¹.

for starting the simultaneous lasing of photons and phonons (point C in figure 1). Therefore, the contribution of (1) to the rate equation for the electron concentration N_e is negligible.

As far as the rate equation for the photon concentration N_k is concerned, the photon losses due to the two-photon absorption should be compared with other channels of photon losses. It is well known that bulk photon losses in semiconductors are dominated by the free carrier absorption with the rate

$$W_e = \sigma N_k N_e, \quad (2)$$

where σ is the photon capture cross section ($\sigma = 6 \times 10^{-18} \text{ cm}^2$ for silicon). The rate of photon losses due to radiation through the end crystal surfaces of reflectivity R ($R = 0.30$ for silicon) is equal to N_k/τ , where

$$\tau^{-1} = \frac{c}{L} \ln(1/R) \quad (3)$$

and L is the crystal length. Assuming $L = 250 \text{ } \mu\text{m}$, we obtain $\tau^{-1} = 3.5 \times 10^{11} \text{ s}^{-1}$.

Since the generation of one electron-hole pair is accompanied by simultaneous absorption of two photons, the rate of the two-photon absorption, dN_k/dt , is two times greater than (1) and can be estimated by the value of $5.8 \times 10^{24} \text{ cm}^{-3} \text{ s}^{-1}$. The rate of photon absorption by free carriers W_e (2) at the electron concentration $N_e = 8.9 \times 10^{18} \text{ cm}^{-3}$ and the rate of photon losses through the crystal facets N_k/τ (3) are $1.2 \times 10^{28} \text{ cm}^{-3} \text{ s}^{-1}$ and $9.1 \times 10^{27} \text{ cm}^{-3} \text{ s}^{-1}$, correspondingly. One can see that the contribution of the two-photon absorption in the rate equation can be neglected.

Summarizing the above estimates, we come to a conclusion that the process of two-photon absorption does not affect the dynamics of the photon-phonon lasing in silicon.

Among the different mechanisms hindering the laser action in silicon, the parasitic non-radiative Auger recombination is of particular concern. This is a three-particle process where an excited electron recombines with a hole and the excess energy is transferred to another free carrier rather than emitted in the form of a photon. It occurs even in pure and perfect semiconductor crystals. The rate of competitive Auger recombination in bulk silicon is generally much higher than the rate of radiative recombination. The internal quantum efficiency of luminescence, which is defined as the ratio of the probability of radiative electron-hole recombination to the total probability of electron-hole recombination, is limited in silicon by the value of 10^{-5} – 10^{-6} [14]. Although the efficiency can be improved to up to 10^{-3} by employing ultrapure silicon [28], it is still too small to consider silicon as a good light-emitting material.

The above obtained estimates for the rate of photon-phonon transitions in silicon confirm this conclusion. Indeed, the rate of Auger recombination is proportional to the third degree of concentration of excited carriers, that is $W_{\text{Auger}} = C_n N_e^3$ with $C_n \sim 10^{-31} \text{ cm}^6 \text{ s}^{-1}$ [29] for intrinsic silicon. Taken into account the electron concentration at the gain maximum, $N_e = 8.9 \times 10^{18} \text{ cm}^{-3}$, we obtain for the rate of Auger recombination the value of $W_{\text{Auger}} = 7.1 \times 10^{25} \text{ cm}^{-3} \text{ s}^{-1}$, whereas the rate of spontaneous-spontaneous photon-phonon transitions W_{00} is estimated to be only $1.6 \times 10^{23} \text{ cm}^{-3} \text{ s}^{-1}$. Hence, most of the excited electron-hole pairs recombine non-radiatively and the corresponding quantum efficiency is

$$\eta = \frac{W_{00}}{W_{00} + W_{\text{Auger}}} = 2.2 \times 10^{-3}. \quad (4)$$

All proposals to circumvent this problem and to get silicon as a laser material are aimed at increasing the rate of radiative recombination and thus at raising the quantum efficiency. In the approach considered in this paper we suggest to stimulate the photon part of the two-quantum transitions in silicon by the light from an appropriate external laser source. This leads to enhancement the rate of radiative recombination. The rate of stimulated-spontaneous transitions W_{k0} at the threshold intensity of stimulating radiation, $I = 2.7 \times 10^7 \text{ W/cm}^2$, is estimated to be $5.5 \times 10^{25} \text{ cm}^{-3} \text{ s}^{-1}$. Accordingly, the associated quantum efficiency,

$$\eta = \frac{W_{k0}}{W_{k0} + W_{\text{Auger}}}, \quad (5)$$

increases by more than two orders of magnitude and reaches the value of 0.44. This allows us to obtain initially an external-source-assisted lasing in silicon (point A in figure 1) and then a true photon-phonon lasing without any external source of radiation (point C in figure 1).

4. Conclusions

Most proposals for creation a silicon laser are based on overcoming or circumventing the problem of low rate of radiative recombination in indirect band gap semiconductors. Among them are [30]: 1) overcoming the indirect band structure by using spatial confinement of electrons, 2) introducing rare-earth impurities as optically active dopants, 3) applying epitaxy and hybrid integration with III-V-semiconductor lasers and 4) using Raman scattering to achieve optical gain. Our paper supplements this list with one more proposal. We suggest to make use of stimulated photon-phonon interband electron transitions in crystalline silicon to obtain phonon lasing and simultaneous lasing of both photons and phonons.

To drive the process, we start with stimulating the photon part of the two-quantum transitions with simultaneous emission of phonons and photons by the light from an appropriate laser source. This leads to a growth of the rate of phonon emission. At a high enough intensity of the external stimulating radiation and at a high enough level of pumping, the phonon gain turns out to be equal to the phonon losses in a particular acoustic mode and the phonon laser action appears. It is then possible to sustain the phonon laser action by gradual increasing the pumping rate with simultaneous decreasing the intensity of the external stimulating radiation. Finally, the source of stimulating radiation can be switch off and the system continues to operate with simultaneous lasing of both photons and phonons.

Dynamics of the photon-phonon laser generation has been investigated with aid of the rate equations. Numerical estimates of the threshold intensity of stimulating radiation, the threshold rate of pumping for starting the phonon lasing and the threshold rate of pumping for starting the simultaneous lasing of both photons and phonons have been made for pure (including isotopic purity) and perfect crystals of silicon. The feasibility discussion of the silicon photon-phonon laser allows us to make a conclusion about the self-consistency of our proposal.

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