Lasing in single-crystal silicon through phonon pumping: A feasibility study

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Abstract

We evaluate the possibility of facilitating fast electron-hole recombination in silicon, including lasing, by phonon pumping–that is, inducing stimulated emission of phonons. However, it is concluded that the large number of phonon modes and their short lifetime makes this idea impractical.

INTRODUCTION

Optoelectronics, the integration of microelectronics with optics, holds great technological importance: While electrons are best for logic and processing, photons are intrinsically superior for communication, due to their higher bandwidth, greater speed, and near lack of heat dissipation [1]. However, the coupling of electrical signals to light is a technological challenge, which is why optical signals are presently used primarily for long-haul fiber-optic communication, and not, say, within a computer chip.

A primary reason for the difficulty of optoelectronics is that the free carriers in silicon are relatively optically inactive, because of the indirect bandgap. Therefore, current optoelectronic devices typically integrate silicon with other, more optically-active materials (typically III-V semiconductors), presenting a great challenge in achieving inexpensive, defect-free, and mechanically-stable epitaxy [1]. An all-silicon design would be greatly preferable.

The indirect bandgap of silicon means that thermally-relaxed electrons and holes have different wavevectors. Therefore a one-step process (an electron and hole recombine and release a photon) would not conserve crystal momentum, which means it is forbidden in a perfect, translationally-symmetric crystal. Instead, a two-step process occurs, where a photon is emitted and a phonon is absorbed or emitted. This is a second-order process, and hence less likely to occur. This explains why the radiative lifetime of minority carriers in single-crystal silicon is of order milliseconds, compared to nanoseconds in direct-gap III-V materials [1]. Instead, recombination is dominated by nonradiative modes associated with defects, surfaces, or the Auger effect [2]. One way around this is to break the translational invariance, through using nanocrystals, superlattices, or porous silicon [1]. Another approach is to not use free electrons and holes at all, but instead stimulate other types of transitions, such as at localized defect levels (often erbium), intersubband transitions in superlattices, or stimulated Raman emission [1, 3–5]. Yet another recent approach is to suppress other recombination mechanisms so successfully that radiative recombination occurs with high likelihood, despite its slowness. This was recently shown to give internal quantum efficiencies of 20% at room temperature [6], and despite early doubts [7], it is now understood that this approach can in theory eventually lead to a laser, at least at cryogenic temperatures ($\lesssim$ 23K) [8, 9]. However, the necessarily-long carrier lifetime means that fast modulation of light output will not likely be achievable this way [1].

Instead, we propose increasing the radiative lifetime by making the phonon emission or absorption occur more readily. More specifically, we evaluate the possibility of creating a large, non-thermal population of an appropriate phonon mode through optical pumping, thereby inducing higher rates of phonon stimulated emission and absorption.

We note that the inverse process, light absorption, could be similarly facilitated by the proposals discussed here. This would be particularly important for silicon photovoltaics, which would benefit immensely from a shorter light-penetration depth. However, for simplicity, we will frame this discussion in terms of light emission.

In this paper, we will first discuss some background on the indirect transition in silicon, followed by methods that might be able to pump phonons. Finally, we discuss how much pumping is necessary and whether this is realistic.

THE INDIRECT TRANSITION IN SILICON

We briefly review some relevant aspects of the indirect optical transition in silicon. While the valence band maximum (VBM) is at the Γ-point ($k = 0$), there are six equivalent conduction band minima (CBM) near the X points of the Brillouin zone, on the along the $\langle 100 \rangle$ ("Δ") lines connecting X to Γ [10]. We introduce the notation $\mathbf{k}_{\mathrm{CBM}}$ as the wavevector of one of those CBM. The VBM and CBM are separated in energy by 1.1eV, the silicon bandgap.

In the indirect radiative transition, a photon near 1.1eV is emitted, and a phonon is emitted or absorbed. The phonon is usually in the transverse-optical (TO) mode, where it has energy 57.8 meV (14.0 THz) [11].

From second-order perturbation theory, the relevant coefficient for any indirect transition is:

$$\left| \frac{\langle \mathbf{k}_f, v | \, H'_{photon} \, | \mathbf{k}_f, c \rangle \, \langle \mathbf{k}_f, c | \, H'_{phonon} \, | \mathbf{k}_i, c \rangle}{E_{\mathbf{k}_i, c} - E_{\mathbf{k}_f, c}} + \frac{\langle \mathbf{k}_f, v | \, H'_{phonon} \, | \mathbf{k}_i, v \rangle \, \langle \mathbf{k}_i, v | \, H'_{photon} \, | \mathbf{k}_i, c \rangle}{E_{\mathbf{k}_f, v} - E_{\mathbf{k}_i, v}} \right|^2 \tag{1}$$

where $|\mathbf{k}, c\rangle, |\mathbf{k}, v\rangle$ denote electronic states in the conduction and valence band respectively, and E is the energy of an electronic state. In this expression, we have ignored the small photon momentum and phonon energy. We have also not explicitly included the phonon-state in the kets, but we note that the phonon-absorbing indirect transitions and phonon-emitting indirect transitions are each calculated independently. Note that there are only two

"intermediate states", included in the sum: All other states give zero contribution, since the photon matrix element must conserve momentum.

This indirect-bandgap expression can be compared to the matrix element controlling the direct-bandgap process:

$$\left|\langle \mathbf{k}, c| \, H'_{photon} \, |\mathbf{k}, v\rangle\right|^2. \tag{2}$$

The term (1) is strongly suppressed relative to (2) by the phonon Hamiltonian, which is much smaller than the energy denominator (of order 1eV). In fact, in silicon, a given phonon mode yields a matrix element of only (in the case of phonon absorption)

$$\langle \mathbf{k}_f| \, H'_{phonon} \, |\mathbf{k}_i\rangle \approx (0.2 \text{ eV})\sqrt{n/N}, \tag{3}$$

where n is the occupancy of the phonon mode and N is the number of unit cells in the crystal [12, 13]. (For phonon emission, replace $\sqrt{n/N}$ by $\sqrt{(n+1)/N}$.) Since $n \approx 0.1$ and $N \approx 10^{23}$, each phonon mode gives a tiny contribution to the transition rate. This is partially compensated by the fact that any pair $(|\mathbf{k}_i, c\rangle, |\mathbf{k}_f, v\rangle)$ may give rise to an indirect transition, but only $\mathbf{k}_i = \mathbf{k}_f$ is allowed in the direct transition. (This fact eliminates the N-dependence.)

PHONON PUMPING MECHANISM

As mentioned above, the TO phonon modes near $\mathbf{k}_{\text{CBM}}$ have energy 58 meV, and therefore by Bose-Einstein statistics the room-temperature equilibrium phonon density is $n \approx 0.1$ phonons per mode. However, this number may be dramatically increased by some process which we will call "phonon pumping". Here we discuss possible pumping mechanisms.

First, we note that it does not matter whether the generated phonons are truly single-mode or fill a distribution of modes of similar wavevector. Indeed, as seen from (1), each phonon contributes linearly to the transition rate. Moreover, the photon energy and phonon wavevector have no exact relationship: A finite range of different phonon wavevectors will all be compatible with electron-hole recombinations at a given photon energy. Therefore, even if the photon is single-mode, like in many lasers, this still does not yield a benefit to creating single-mode phonons.

The simplest pumping mechanism one might think to try is to shine laser light at 58 meV, exciting the phonon mode at the same energy. However, by wavevector conservation, a one

photon-one phonon transition is forbidden. On the other hand, one-photon two-phonon optical absorption may be allowed, if the two phonons have opposite wavevector. While a selection rule forbids a photon creating two TO phonons at $\pm\mathbf{k}_{\mathrm{CBM}}$, a photon of the right energy can create a TO phonon at $+\mathbf{k}_{\mathrm{CBM}}$ and an LO, LA, or TA phonon at $-\mathbf{k}_{\mathrm{CBM}}$ [14].

A second possibility would be to use coherent stimulated two-phonon Raman emission [15]. This process differs from ordinary two-phonon Raman emission in that stimulated emission is used to greatly enhance the Stokes-shifted beam, transferring energy both into the beam and into the phonon system.

A third possibility would be to create a "phonon cavity", allowing positive feedback and amplification of emitted phonons. "Phonon mirrors" can be created by appropriately-chosen material interfaces, or superlattices [16], perhaps allowing standing waves to build up.

Yet another possibility would be to use defect vibrational modes to mediate between photons and bulk phonons. An impurity introduced into an otherwise-perfect crystal will create new vibrational modes. In the case of a substitutional impurity that is lighter than the surrounding atoms, this often leads to well-localized vibrational modes with a higher oscillation frequency than any of the unperturbed bulk modes. In the case of a heavier impurity, the newly-introduced modes are often in the continuum of bulk modes, and are called "resonant", "quasilocal", or "pseudolocal" modes [17–19], reflecting the fact that the localized vibration mixes with the bulk phonon modes of the same frequency. One would search for an impurity atom that is electronically benign but which happens to have pseudolocal modes in silicon very close to the $\mathbf{k}_{\mathrm{CBM}}$ TO phonon frequency of 14.0 THz. After introducing this impurity into silicon, light could excite the pseudolocal mode, which would quickly exchange energy with the desired bulk mode.

REQUIRED PUMPING

First we note that the TO phonon lifetime is unfortunately quite small: Around 30 picoseconds, after which the phonons decay anharmonically into other modes [20].

More importantly, the number of relevant phonon modes is quite large. It is an informative comparison to consider the number of phonons in thermal equilibrium that contribute to the indirect transition (in the absence of pumping). Consider a transition in which an electron and hole recombine, emitting a a photon of energy E, and a phonon. If we ignore

the small phonon dispersion, we can assume the emitted phonon always has energy E_Q; then let $\Delta = E + E_Q - E_{gap}$ be the excess energy over the bandgap. Let m_e^* and m_h^* be the electron and hole effective masses, ignoring anisotropy. The recombination could involve an electron at the CBM and a hole at $|\mathbf{k}| = \sqrt{2m_h^*\Delta}/\hbar$, or a hole at the VBM and an electron at $|\mathbf{k} - \mathbf{k}_{\rm CBM}| = \sqrt{2m_e^*\Delta}/\hbar$, or something in between. All told, taking into account the six conduction-band valleys and the electron and hole effective masses in silicon, the involved phonons take up a volume of k-space of roughly $8\pi(2m\Delta)^{3/2}/\hbar^3$, where m is the true electron mass. For $\Delta = 0.1$ eV, and with 0.1 phonons per mode, we find that in thermal equilibrium there are about 4×10^{19} cm^{-3} phonons helping assist the transition (one per 20 nm^3).

If we wanted to speed the transition up by a factor of 100 (which would still leave it several orders of magnitude slower than direct-bandgap III-V materials), we would need to create 4×10^{21} cm^{-3} phonons. These newly-created phonons would have a collective energy of 40 J/cm^3, and if they dissipated they would heat the silicon by 20K. Such strong pumping is unrealistic even using a pulsed laser, particularly since the pumping power must be delivered within the 30ps phonon lifetime, and since the required laser wavelength would be in the mid- to far-infrared regime.

CONCLUSION

We have argued that phonon-pumping might in principle be a way to facilitate radiative transitions in silicon, but in practice the large number of phonon modes and weakness of the electron-phonon interaction makes this impractical. Nevertheless, we remain hopeful that there may be some other technique to enhance the electron-phonon interaction in silicon, and we hope that researchers will continue to work on this interesting and important challenge.

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