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A proposal for Coulomb assisted laser cooling of piezoelectric semiconductors

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Anti-Stokes laser cooling of semiconductors as a compact and vibration-free method is very attractive. While it has achieved significant milestones, increasing its efficiency is highly desirable. The main limitation is the lack of the pristine material quality with high luminescence efficiency. Here, we theoretically demonstrate that the Coulomb interaction among electrons and holes in piezoelectric heterostructures could lead to coherent damping of acoustic phonons; rendering a significantly higher efficiency that leads to the possibility of cooling a broad range of semiconductors. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4891763]

Laser cooling of solids has been an important topic in recent decades.[1,2] It is a compact, vibration-free, and robust technique for cooling to cryogenic temperatures[3] and is quite favorable to be used in spacecrafts and for future all-integrated electronics operating at low temperatures. Apart from the laser trapping of atoms, the first observation of laser cooling of a solid material dates back to 1995, when Epstein *et al.*[4] demonstrated the first optical refrigeration of a rare-earth doped glass by anti-Stokes process. In this method, the electrons undergo phonon absorption after photoexcitation and re-emit photons with higher energy. As a consequence, the phonon density is suppressed and the material cools down. Another important method of laser cooling of solids uses the coupling of the cavity optical modes to the mechanical vibration modes of a mirror in an optomechanical setup.[5] Both methods have been implemented in semiconductors.[6,7]

The problems associated with anti-Stokes laser cooling of semiconductors are primarily parasitic absorption, photoluminescence trapping, and the insufficient quantum efficiency. All of these have been addressed for a CdS nano-ribbon recently due to its outstanding material quality and its sub-wavelength thickness which yielded a high photon extraction efficiency.[7] However, for bulk III-V semiconductors, anti-Stokes cooling is predicted to be impossible.[7] On the other hand, the implementation of traditional optomechanical cooling of thin membranes of a semiconductor cools (damps) only one isolated mechanical mode. The decoupling of this mechanical mode stems from high quality factor of the mechanical cavity, and prevents effective laser cooling of the phonon bath.

In this Letter, we demonstrate that Coulomb interaction can be exploited to suppress phonons through piezoelectric effect with much higher efficiency than anti-Stokes cooling. Here, the term "Coulomb interaction" refers to the interaction between the phonon and the electric field in the piezoelectric material under consideration rather than only magnitude of electric field between electrons and holes. Inspired by optomechanical cooling and amplification,[5,8] we investigated the possibility of a similar process by coherent light-induced piezo strain on the atoms. Although each phonon mode can be regarded as a low-Q cavity mode, however if numerous number of phonon modes get involved in this cooling process, then observation of a macroscopic temperature drop is expected. It is shown that the proposed mechanism under specific conditions can lead to optical refrigeration of highly piezoelectric GaN/InGaN quantum well structures at room temperature with less strict material quality requirements.

The underlying mechanism is shown in Fig. 1 and can be explained as follows: The strain associated with the phonons leads to an effective piezoelectric field, which coherently changes the bandgap through Quantum Confined Stark Effect (QCSE). The resulted change of bandgap leads to instantaneous change of the laser detuning and the density of photogenerated carriers. Consequently, the electrostatic field due to Coulomb interaction between the electron-hole pairs and the associated piezoelectric stress exerted on the atoms is changed. This produces a backaction mechanism between the average displacement of atoms (Δu_{avg}) caused by the propagating phonons and the resulting piezoelectric stress (σ_{piezo}) on atoms due to the change of carrier density. Similar to cavity optomechanical backaction, this process can lead to either cooling or parametric amplification depending on the direction of the piezoelectric force upon the change of carrier density caused by phonons. In the case of cooling, the photogenerated electrons gain energy from phonons; the amount of energy for each phonon mode is equal to the area enclosed by the stress-displacement curve as shown in Fig. 1. The excess energy is then extracted from the material by radiative recombination, leading to an effective net cooling.

The dynamics of the average phonon displacement within the well ($u_{avg} = (1/N)\sum u_i$, N being the total number of mono-layers of the well and u_i is the displacement of ith monolayer) can be expressed as follows:

$$M\frac{d^2u_{avg}}{dt^2} = -(C_{couloumb}(t) + C)u_{avg}, \qquad (1)$$

where M is the mass of a monolayer and C is the effective elasticity of the crystal. $C_{Coulomb}$ is an effective elasticity which is equal to the ratio of the backaction piezo-stress to

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[a)] Electronic mail: hmohseni@ece.northwestern.edu the induced average strain of the phonon. Based on the phase of $C_{Coulomb}(t)$, it can be either an amplifying or a dissipative term. In the proposed structure, the change of carrier density and the corresponding change in electrostatic field results in a change of piezoelectric stress which retards atomic motion, and hence, it is dissipative. In what follows, we describe our semi-classical theory to find the cooling efficiency of this dissipative term. Our approach is based on small signal analysis of the aforementioned physical observables in the first order approximation. This assumption holds at room temperature and for a bulk material where each phonon mode affects atoms' positions infinitesimally leading to very small changes in transition energies, photogenerated carrier density and the backaction piezo strain. Furthermore, the damping piezo-stress should be coherent with the corresponding phonon mode in order to result in a nonzero cooling power over time. Therefore, the correlation of Coulomb cooling of different phonon modes can be neglected, and the net cooling power is obtained by summing over the cooling powers of individual phonon modes. In order to simplify the analysis, it is assumed that the system is in thermal equilibrium with a reservoir at room temperature. In this regard, Bose-Einstein statistics, in conjunction with phonon density of states of the material, can be used to find the relative contribution of different phonon modes.

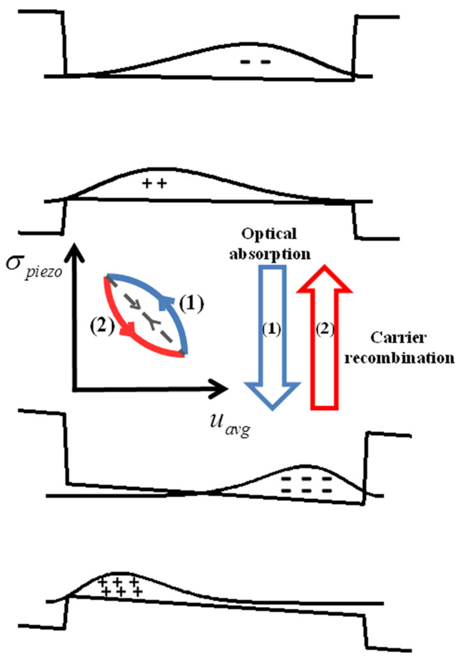

FIG. 1. The schematics of the Coulomb assisted cooling process: The propagation of an acoustic phonon results in a change of piezoelectric field and the bandgap. Therefore, the resulting change of photogenerated carrier density leads to the change of piezo-stress on the atoms. The diagram of piezo-stress and the average displacement of the atoms by a phonon mode within the well is also shown. The nonzero area encircled by curves (1) and (2) implies a net work done on the atoms. While route (1) corresponds to the gradual increase of photogenerated carrier density, route (2) pertains to the gradual decrease of carrier density by various recombination processes. For high frequency phonons, the piezo-stress is almost in phase with the average displacement. This linear behavior suppresses the cooling power as shown by the dashed line in the stress-displacement diagram.

By solving the equation of the carrier density dynamics[9] (i.e., $\frac{d\Delta n}{dt} = \Delta G - R_n \Delta n$, where Δn is the change in carrier density, R_n is the recombination rate, and G is the generation rate per unit volume), the amplitude of carrier density modulation by the phonon mode q can be found

$$\Delta n_q(t) = \frac{R_n}{R_n^2+\omega_q^2}\Delta G_q - \frac{1}{R_n^2+\omega_q^2}\left(\frac{d\Delta G_q}{dt}\right), \tag{2}$$

where ω_q is the frequency of the phonon which modulates the bandgap ($\Delta E_{g,q}$) and, consequently, the generation rate (ΔG_q) with the same frequency. Due to the charge separation within the QW, the change of carrier density leads to the change of the internal electric field, i.e., $\lambda_{21}\Delta n$, where λ_{21} is the coulomb strength parameter. This leads to coherent modulation of piezoelectric stress ($\Delta\sigma_{piezo,q}$) which is proportional to the change in the internal electric field.[10,11] The cooling power per unit area of the phonon mode q can be calculated as:[8]

$$I_q = \sum\left\langle \Delta\sigma_{piezo}\left(\frac{du_z}{dt}\right)\right\rangle, \tag{3}$$

where $\langle\ \rangle$ denotes average of the quantity of interest over one period of phonon oscillation and the summation has to be taken over all of the atomic layers perpendicular to the growth direction.

It should be noted that the generation rate (G) is related to the optical absorption (α). The optical absorption is a Lorentzian function of laser detuning with an effective absorption broadening[12] (Γ). Therefore, it is straightforward to show that ΔG_q, which is proportional to $\frac{d\alpha}{dE_g}\Delta E_{g,q}$, is inversely proportional to Γ^2. This relationship is independent of the phonon mode; therefore, the maximum Coulomb cooling efficiency is also inversely proportional to Γ^2. It should be noted that the absorption broadening arises from various sources such as carrier-carrier scattering, scattering by the impurities and defects, composition fluctuation, and electron-phonon interaction.[13] The absorption broadening is lower for materials with higher quality and lower crystal defects.

$\Delta E_{g,q}$ is proportional to the induced piezoelectric field of the phonon across the quantum well, and hence its average induced strain. On the other hand, the average strain is directly related to the velocity of the phonon mode. Therefore, $\frac{d\Delta G_q}{dt} \propto \frac{d\Delta E_{g,q}}{dt}$ is 90° out of phase with the average velocity of the phonon mode. As a result, the contribution of the second term on the right hand side of Eq. (2) to I_q is negligible as confirmed by numerical calculations.

We can also conclude from Eqs. (2) and (3) that I_q is proportional to $R_n/(R_n^2+\omega_q^2)$ which compares the phonon oscillation frequency and the carrier recombination rate. It is evident that the carrier recombination rate plays a key role in both the magnitude and phase of $\Delta\sigma_{piezo}$ and, consequently, the cooling power of the phonon mode. In this regard, Coulomb cooling efficiency of low frequency phonons is larger, since the delay in carrier recombination compared to the phonon oscillation period is negligible and does not lead to a significant phase difference between the backaction stress and the phonon velocity.

To demonstrate this concept in a real structure, we considered a wurtzite c-GaN/InGaN multi quantum well (MQW) system to study the Coulomb assisted laser cooling because of its high piezoelectricity. Furthermore, the strong piezoelectric field results in spatial separation of electrons and holes leading to a strong Coulomb interaction to suppress the phonons. The bandstructure of the MQW system was obtained by a six band $k.p$ model similar to Ref. 14. The piezoelectric field, effective masses, and band line-ups in presence of strain have been calculated based on data of Ref. 15. The quantum well structure consists of 1 nm of $In_{0.15}Ga_{0.85}N$ sandwiched between 6 nm GaN barriers. The self-consistent solution of Poisson and Schrödinger equations was performed in order to find the change of bandgap by applying a constant electric field over the quantum well region to find the QSCE coefficient. The change of carrier density is calculated by solving the rate equation considering all generation and recombination processes into account. We obtained the Coulomb strength parameter defined as the ratio of the change in internal electric field to the change in carrier density.

The total Coulomb cooling power per unit area ($I_{Coulomb}$) is calculated by summing over I_q considering the phonon density of states (DOS) within the first Brillion-zone ($I_{Coulomb} = \int I_q DOS(q) d^3q$). The Coulomb cooling efficiency is consequently obtained by taking the ratio of the total cooling power to the absorbed laser intensity. This is a parameter that characterizes the energy extracted from the phonon bath by the Coulomb interaction with the photogenerated carriers. However, all of the excess energy gained by the carriers cannot be extracted from the material through the radiative recombination. Non-radiative recombination processes produce heat and recycle the energy back to the phonon bath. The implementation of Coulomb cooling mechanism is independent of anti-Stokes cooling. Therefore, the excess energy of the carriers gained by this process can be added to the anti-stokes excess energy (kT) to find the total excess energy of the carriers. As a result, the internal cooling efficiency (η_{ct}) is a summation of both the anti-Stokes cooling efficiency ($\sim kT/E_g$) and the Coulomb cooling efficiency. The net cooling efficiency is calculated as follows:[16]

$$\eta_{c,net} = \eta_{ext}(1 + \eta_{ct}) - 1, \tag{4}$$

where η_{ext} is the external quantum efficiency (EQE) and is defined as[16]

$$\eta_{ext} = \frac{\eta_e B n^2}{\eta_e B n^2 + An + Cn^3}, \tag{5}$$

where η_e is the light extraction efficiency (the ratio of the luminescent photons coupled outside of the material to those generated) and n denotes the carrier density. A is Shockley-Reed-Hall, B is radiative, and C is the Auger recombination rate. Fig. 2 shows the net cooling efficiency with and without the presence of Coulomb cooling versus detuning for both an ideal case of 100% luminescence efficiency and a practical set of recombination parameters (100% extraction efficiency has been considered for both cases). For the ideal case, the presence of Coulomb cooling mechanism increases the net cooling efficiency from 0.9% (with only anti-Stokes cooling) to 10.5%. For the latter case, the maximum net cooling efficiency with only anti-Stokes process is about −4.5% (no net cooling), and it can reach to 2% with the presence of the Coulomb process. We notice from Fig. 2 that for the detunings far from the band-edge, the net cooling efficiencies for both cases (with and without the Coulomb effect) become essentially equal. This is due to the fact that: (1) Most of the internal power goes into nonradiative Shockley-Reed-Hall (negative detunings) or Auger process (positive detunings) and (2) the Coulomb cooling efficiency is only significant near the band-edge where the magnitude of dn/dE_g is high. Therefore, for large detunings, the term $\eta_{ext}\eta_{ct}$ becomes negligible in Eq. (4), and the results for both cases converge.

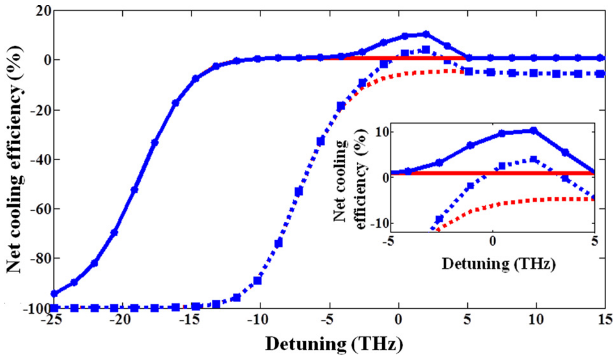

FIG. 2. The net cooling efficiency versus detuning for two cases: The solid lines (both with and without markers) pertain to the ideal case of 100% luminescence efficiency, whereas the dotted lines are for the case of practical quantum efficiency. The curves with markers correspond to the cases where both the anti-Stokes and Coulomb cooling are present. It is assumed that the extraction efficiency is 100%, the intensity of the laser is 5 $mW/\mu m^2$, the absorption broadening is 19 meV and T = 300 K. The inset shows a magnified view of the net cooling efficiency for small detunings.

According to Eqs. (4) and (5), net cooling happens when the nonradiative recombination lifetime (defined as $\tau_{nr} = 1/A$) is longer than a certain value named the "break-even" non-radiative lifetime ($\tau_{nr} > \tau_{nrb}$).[16] The presence of the internal Coulomb cooling leads to a shorter "break-even" non-radiative lifetime as demonstrated in Fig. 3. We also notice that the lower the broadening, the shorter the break-even non radiative lifetime will be. This is due to the fact that the change of carrier density versus detuning increases by the reduction of absorption broadening. The red line is the measured value of non-radiative lifetime in a similar

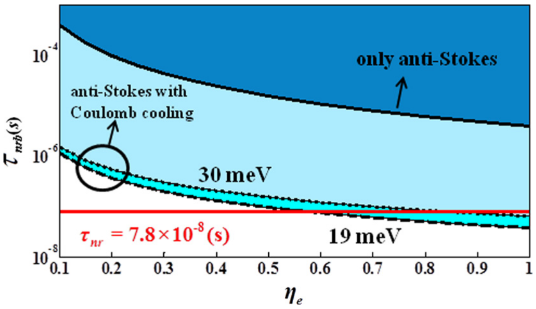

FIG. 3. The break-even nonradiative recombination lifetime (τ_{nrb}) as a function of extraction efficiency for anti-Stokes cooling (solid line with no marker) and the combination of anti-Stokes and Coulomb cooling for different values of absorption broadening. While 30 meV, absorption broadening has been experimentally demonstrated;[18] we predict that with material improvement, better linewidths close to 19 meV (the value for GaN epilayers[19]) might be achieved in the future. The red line is a reported non radiative recombination lifetime for a similar structure.[17] All of the results are for room temperature.

InGaN/GaN QW structure,[17] which shows that a net laser cooling is possible with the presence of Coulomb cooling, when the broadening is at or below 30 meV.[18] We have also calculated the "break-even" non radiative lifetime for the lowest reported value of the broadening (∼19 meV) as reported for GaN epilayers in Ref. 19. For a high quality epitaxial structure with 19 meV absorption broadening, net cooling can be achieved with a light extraction efficiency of only 55% at room temperature. Note that the anti-Stokes process cannot produce a net cooling even with 100% extraction efficiency. We also point out that a record of 73% extraction efficiency has been experimentally demonstrated for InGaN/GaN MQW structures.[20]

It should be noted that both EQE and the Coulomb cooling are dependent on laser intensity. At very high intensities, two-photon absorption dominates. Two-photon absorption is almost independent of detuning near the band-edge;[21] therefore, the change of carrier density and the Coulomb cooling efficiency is suppressed. On the other hand, small change of bandgap by a phonon has a much more significant effect on photogeneration than thermal generation when laser wavelength is tuned close to the bandgap. This is due to the fact that the optical absorption varies very quickly in the vicinity of the band-edge. Therefore, as the thermal generation rate becomes dominant (for example at very low laser intensities), the change in carrier density with bandgap modulation ($\Delta E_{g,q}$) is decreased, and the Coulomb cooling efficiency is suppressed. However, before the thermal generation starts to degrade the Coulomb cooling efficiency, the reduction of the EQE due to low carrier density suppresses the net cooling.

In conclusion, we demonstrated that Coulomb interaction among spatially separated electron hole pairs can be used for cooling of a piezoelectric material. This is due to the coherent piezoelectric force on the atoms caused by the change of carrier density. Our investigations show that the cooling efficiency of this mechanism can be significantly higher than anti-Stokes cooling. However, the cooling efficiency is strictly dependent on the laser intensity and wavelength. It is demonstrated that this mechanism can lead to laser cooling at room temperature with existing material quality that is achievable in a broad range of semiconductor compositions. The next step is to explore the same mechanism for coherent phonon amplification in piezoelectric materials.

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