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Numerical calculation of optical phonon decay rate in InN/GaN MQW

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Abstract. Anharmonic decay of high frequency phonons into low frequency vibrations is a significant energy loss mechanism in semiconductors. In the field of Hot Carrier Solar Cells (HCSC), preventing this decay is of great importance as it helps increase hot carriers lifetime. Phonon decay in nitride compounds as well as their consisting nano-crystals like quantum dots and multiple quantum wells (MQWs) has not been extensively studied in the literature. In this work, the decay channels of the A1 and the high-lying E2 optical phonons in an InN/GaN MQW are analysed. We find that the no Klemens decay is present in A1 mode whereas on the contrary E2 is dominated by this process. We also observe that the linewidth for A1 is enlarged a few times than the bulk counterpart while that for E2 remains similar, which is attributed to different vibration nature.

1. Introduction

Nano-crystals or nano-materials have been a hot topic since the 1990s when people started to investigate physical phenomenon in such a small scale. Since the debut of nano-crystal prototypes, interpreting the physics behind the findings became inevitable. Due to the confinement effects, transit from bulk to nano-scale properties has attracted researchers' interests, not only because the physics is not clear but also because the differences nano and bulk materials post a wide range of potential applications. A great number of models have been applied to the field of nano-crystals, such as nano-tubes, nano-rods, quantum dots and quantum wells, so that novel properties can be determined even before materials and devices are fabricated. In this paper, a numerical model is built to investigate the optical phonon decay rate in both bulk nitride compounds and their counterpart MQW structure. This study aims to unravel whether MQWs are suitable for HCSCs in terms of phonon lifetime.

Long hot carrier lifetime is the most essential property of HCSC, under which HCSC can reach a very high energy conversion efficiency [1] far beyond the Shockley-Queisser limit [2]. The requirement under which a HCSC can differ from a conventional solar cell is actually quite strict. Optical phonon lifetime of around a few hundred pico-seconds is required but normal bulk materials only have lifetime of a few to some tens pico-seconds or less. One potential mechanism of blocking phonon decay was proposed in the prototype HCSC model. It states that if the phononic band-gap between the optical and the acoustic phonon branch is larger than the energy of the highest acoustic phonon, the optical phonon lifetime could be increased [3]. This



is based on an assumption that the Klemens decay [4] is the dominant channel in materials. In this research, this assumption is examined in an InN/GaN MQW consisting of two atomic layers of both InN and GaN, which is also hot carrier absorber candidate, owing to its multi-layer structure and large mass atomic ratio [5].

2. Anharmonic theory

$$\Gamma = \frac{2\pi}{\hbar} |\langle \psi_i | H | \psi_f \rangle|^2 \quad (1)$$

In an anharmonic crystal, phonon decay is inevitable and in most circumstances three-phonon process is the dominant mechanism because the probability for four-phonon or even higher processes drops extremely fast. The Klemens mechanism states that a phonon can decay into two acoustic phonons of equal energy and opposite momentum. In general, as long as energy and momentum are conserved the decay process can occur. Fortunately, the Fermi's golden rule allows one to evaluate the transition rate using a simple and elegant formula [EQ 1]. Different transitions happen at different rates depending on the coupling between the initial state and the final state characterised by the Hamiltonian. In the case of phonon decay, the Hamiltonian contains phonon creation and annihilation operators as well as the perturbed crystal energy. The initial state is usually the state of interest, i.e. optical phonons, while the final states in this situation are the lower-branch phonons. In this study, we only consider zone-centre optical phonon decay processes including both Klemens' and non-Klemens Channel as this kind of phonon is most likely to interact with photons.

$$E_{tot}(\mathbf{u}) = E_{tot}^{(0)} + \frac{1}{2} \sum_{ll',ss',\alpha\beta} \frac{\partial^2 E_{tot}}{\partial u_{s,\alpha}^l \partial u_{s',\beta}^{l'}} u_{s,\alpha}^l u_{s',\beta}^{l'} \quad (2)$$

$$+ \frac{1}{6} \sum_{ll''',ss's'',\alpha\beta\gamma} \frac{\partial^3 E_{tot}}{\partial u_{s,\alpha}^l \partial u_{s',\beta}^{l'} \partial u_{s'',\gamma}^{l''}} u_{s,\alpha}^l u_{s',\beta}^{l'} u_{s'',\gamma}^{l''} + \mathcal{O}(\mathbf{u}^3)$$

The total energy of the crystal can be expanded into Fourier series in terms of atomic displacements or phonons. In EQ 2, \mathbf{u} represents the atomic displacement while the indices s, l and $(\alpha \beta \gamma)$ are the atomic species, number of unit cell and the three Cartesian coordinates respectively. The third term in this equation gives the perturbed crystal energy due to the anharmonic three phonon process. In the second quantisation theory, atomic displacement can be translated into reciprocal space and same for the third order crystal energy. After all the algebra, the Hamiltonian operator for the three phonons decay process reads:

$$H^{anh} = \frac{1}{6} \sum_{\mathbf{q}, \mathbf{q}', \mathbf{q}''} \sum_{j_0, j_1, j_2, j_3} U(\mathbf{q}, \mathbf{q}', \mathbf{q}''; j_0, j_1, j_2) \left(a_{j_0}^\dagger(-\mathbf{q}) + a_{j_0}(\mathbf{q}) \right) \quad (3)$$

$$\left(a_{j_1}^\dagger(-\mathbf{q}) + a_{j_1}(\mathbf{q}) \right) \left(a_{j_2}^\dagger(-\mathbf{q}) + a_{j_2}(\mathbf{q}) \right)$$

a^\dagger and a are simply the phonon creation and annihilation operators while $U(\mathbf{q}, \mathbf{q}', \mathbf{q}''; j_0, j_1, j_2)$ in EQ 3 denotes the Fourier-transformed anharmonic crystal energy which is evaluated within the Density Functional Theorem (DFT) framework in this study. U is the most important factor in the calculation. Without a well established method, any result from the Fermi's golden rule is nothing more than a rough estimation. In the past, a single macro coupling constant was usually used for all possible transitions, which we are still able to meet in many recent studies. Evaluating the constants by DFT was first attempted by Debernardi [6] in the 1990s. This has proven to be a powerful and accessible method as modern computing capacity continues its rapid

growth. Computing the coupling constants is made possible by the so-called $2n + 1$ theorem [7] which allows one to go to the n^{th} order crystal energy at minimum cost. In this study, all physical quantities including lattice parameters, phonon dispersions and the anharmonic terms were extracted from the results produced by the package “Quantum Espresso” [8].

We then apply the Fermi’s golden rule, simplifying the expression by using the properties from phonon creation and annihilation operators: $a^\dagger |n\rangle = \sqrt{n+1} |n+1\rangle$ and $a |n\rangle = \sqrt{n} |n-1\rangle$, where n is the Bose-Einstein occupation. Eventually one reaches the final formula of zone centre optical phonon decay rate via all possible channels:

$$\Gamma_{LTO} = \frac{\pi}{2\hbar} \sum_{\mathbf{q}, j_1, j_2} |U(\mathbf{0}, \mathbf{q}', \mathbf{q}''; LTO, j_1, j_2)|^2 \{ [n_{j_1}(\mathbf{q}) + n_{j_2}(\mathbf{q}) + 1] \delta[\omega_{LTO}(\mathbf{0}) - \omega_{j_1}(\mathbf{q}) - \omega_{j_2}(-\mathbf{q})] + [n_{j_1}(\mathbf{q}) - n_{j_2}(\mathbf{q})] \delta[\omega_{LTO}(\mathbf{0}) \pm \omega_{j_1}(\mathbf{q}) \mp \omega_{j_2}(-\mathbf{q})] \} \quad (4)$$

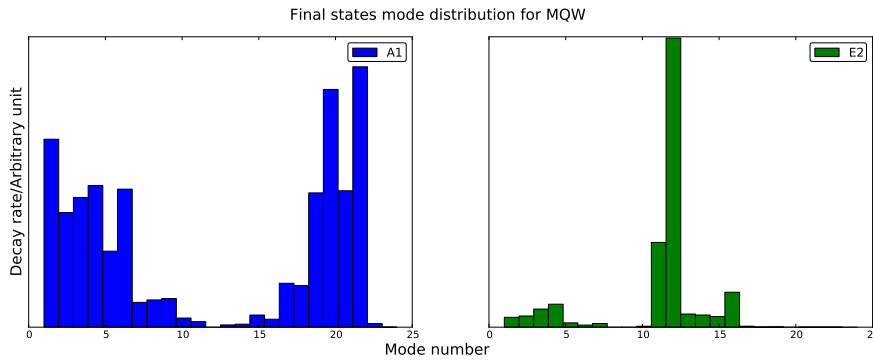


Figure 1. Mode-resolved final state distribution for the MQW. Mode number is simply the number of the eigenstate. A1 is at mode number 24 while the E2 is at 20. In this particular structure, we have 24 modes in total for one reciprocal point.

Table 1. Decay channel comparison and predicted linewidths for A1 and E2 modes. The two numbers under the channel columns are the channel occupation and the actual contribution to the decay rate respectively. The linewidth and the lifetime are predicted at $T = 10$ K.

	Klemens[%]	Ridley[%]	Others[%]	linewidth [cm^{-1}]	lifetime [ps]
A1	0.00, 0.00	29.5, 48.9	70.5, 51.1	55.30	0.10
E2	10.4, 55.2	55.9, 15.1	33.8, 29.8	1.59	3.34

3. Decay channel analysis and conclusion

We observe that the decay channels in the MQW studied in this work are rather complicated consisting of not only Klemens and Ridley [9] channels, but also other pathways (Figure 1 and Table 1). The Ridley channel represents decay into one lower energy optical phonon and one acoustic phonon, while the other channels contain all non-Klemens and non-Ridley pathways. The contributions to the final decay rates for different decay channels are different, of which the

assumed dominant Klemens channel appears to be comparable with Ridley channel and other channels. In Table 1, one can see that there is no Klemens channel for A1 mode however the Ridley and other channels make up the whole decay rate. The case in E1 mode is quite different. The Klemens channel only occupies 10.4% of possible paths but is responsible for 55.2% of the total decay rate. In contrast, Ridley channel consists of more than half of the possible paths but only contributes 15.1% to the total rate. Even the other channels contribute more than the Ridley channel does.

Consistency is demonstrated between the computed decay rates and Raman experiments [10, 11, 12]. In the latter, the authors suggested that in general, A1 mode transits to lower energy phonons at a much faster rate than E2 mode. They reported that the linewidths for A1 and E2 mode in GaN are >11.0 and $1.67\sim 3.9$ cm^{-1} while the widths in InN are 15.0 and $2.0\sim 5.7$ cm^{-1} . This is due to the fact that the phonon energy for A1 mode is some tens to hundred cm^{-1} higher than that for E2 mode, which naturally allows more decaying paths. Compared with the bulk materials, the decay rate for the A1 mode in this MQW is a few times faster than that in the bulk. The E2 mode seems to be comparable. This is reasonable because the A1 mode is the vibration along the confined direction in which the MQW is grown. As the MQW structure reshapes the crystal structure along that direction and more permitted modes exist, A1 can easily decay into modes that conserve both energy and momentum. Contrarily, for E2 mode vibrating in a transverse way, the difference between in bulk and in MQW is limited.

In conclusion, we have examined and analysed possible decay channels in an InN/GaN MQW. Our results show that Ridley and other possible channels can be competitive with the Klemens channel. For phonons vibrating along the growing direction, their lifetime may be suppressed due to more allowed decay paths introduced by the quantum well structure. It seems that our findings do not favour the MQW structure in terms of a longer optical phonon lifetime. However, before drawing this conclusion, it is very important to analyse all other optical modes in the MQW, which could be the subject of future work.

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