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Effective Numerical Calculation of Second Harmonic Generation Using the Linear Analytic Tetrahedron Method and Piezoelectric Coefficients Using Berry Phase Theory

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1. Theory

The Brillouin integrals in question are of the form of (1), where the integrand consists of matrix elements and energy eigenvalues. These can be acquired by various means, e.g. ab initio calculations or through the empirical pseudopotential method.

$$I(\omega) = \int A(\boldsymbol{k})\delta\left(\tilde{E} - \alpha\hbar\omega\right)d^3k \quad (1)$$

By linearly interpolating the integrand between the tetrahedron vertices, the volume integral can be restated as an integral over energy surfaces of constant value within each tetrahedron.

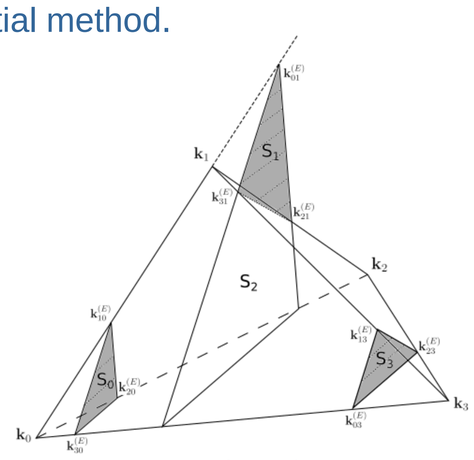

Figure 1

$$I(\omega) = \sum_T \int A(\boldsymbol{k})\delta\left(\tilde{E} - \alpha\hbar\omega\right)\frac{1}{|\nabla_{\boldsymbol{k}}\tilde{E}|}d\tilde{E}(\boldsymbol{k})dS \quad (2)$$

Solving the integral would then yield analytic expressions, only dependent on the function A(**k**) and the energy differences in the corners of the microcell. Hence the overall integral may be approximated as the sum of the analytic results over each microcell, hence yielding an easily implementable approach for reducing the number of **k**-points. This method is also known as the Linear Analytic Tetrahedron Method (LATM), and its applicability to nonlinear optical responses was tested.

2. LATM applied to SHG

To test the LATM the Second Harmonic Generation (SHG) was calculated for zinc blende crystals. This was then compared with a calculation using simple point sampling. The SHG response can be calculated as shown in eq. (3) to (5).

$$Im\left\{\chi_{\alpha,\beta,\gamma}^{(2i)}\right\} = -\frac{\pi e^3}{(2\pi)^3\varepsilon_0 m^3\hbar^2\omega^3} \times \int \sum_{v,c} \{\Pi_{\alpha,\beta,\gamma}^{(\omega)}\delta(\omega_{cv} - \omega) + \Pi_{\alpha,\beta,\gamma}^{(2\omega)}\delta(\omega_{cv} - 2\omega)\} d^3\boldsymbol{k} \quad (3)$$

$$\Pi_{\alpha,\beta,\gamma}^{(\omega)} = \sum_l Im\left\{\left\langle p_{vc}^{(\alpha)} p_{cl}^{(\beta)} p_{lv}^{(\gamma)}\right\rangle \times \frac{\omega_{cl} - \omega_{lv}}{(\omega_{cv} + \omega_{cl})(\omega_{cv} + \omega_{lv})}\right\} \quad (4)$$

$$\Pi_{\alpha,\beta,\gamma}^{(2\omega)} = \sum_l Im\left\{\left\langle p_{vc}^{(\alpha)} p_{cl}^{(\beta)} p_{lv}^{(\gamma)}\right\rangle \times \frac{2}{(\omega_{cl} + \omega_{lv})}\right\} \quad (5)$$

In figure 2 it can be seen that there is a high level of agreement between the two methods for a large number of points. Figure 3 shows the convergence. It was proposed that the poor convergence is explained due to double resonances and that for a few eV good convergence should be seen. This is confirmed in Figure 4.

Figure 2

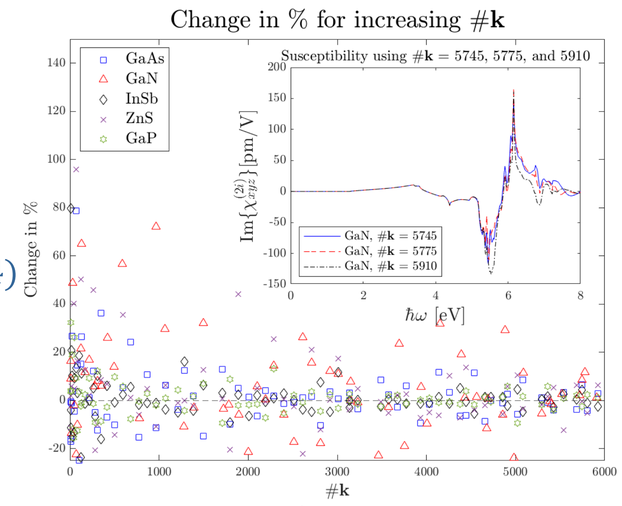

Figure 3

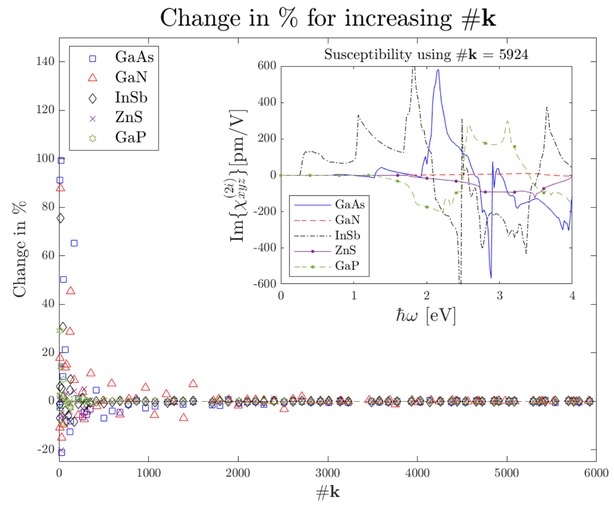

Figure 4

3. Calculation of Piezoelectric coefficients

The piezoelectric coefficients describe how a polarisation is induced when a strain is applied can be described by eq. (6)

$$\gamma_{i,j} = \frac{\partial P_i}{\partial \epsilon_j}. \quad (6)$$

To find these coefficients, one can consider a small applied strain along the c-axis which would in turn induce a polarisation along the c-axis described by (7).

$$\delta P_3 = \gamma_{3,3}\epsilon_3 \quad (7)$$

$$\gamma_{3,3} = \frac{\partial P_3}{\partial \epsilon_3} + \frac{\partial P_3}{\partial u}\frac{\partial u}{\partial \epsilon_3}\Big|_{u_0} \quad (8)$$

By some different expansions, the piezoelectric coefficient is obtained as (8), where similar expressions can be found for the other coefficients. To calculate the polarisation, Berry phase theory is employed. In this framework the polarisation is expressed as

$$\boldsymbol{P}_{tot} = \frac{e}{V_{cell}}\left[\sum_\mu Z_\mu \boldsymbol{\tau}_\mu - \sum_j \frac{\bar{\phi}_j}{2\pi}\boldsymbol{a}_j\right] \quad (8)$$

$$\phi = -Im\left\{\ln\left[\prod_{i=0}^{N-1}\det\left(\begin{bmatrix}\langle u_{1,\boldsymbol{k}_i}|u_{1,\boldsymbol{k}_{i+1}}\rangle & \cdots & \langle u_{1,\boldsymbol{k}_i}|u_{M,\boldsymbol{k}_{i+1}}\rangle \\ \vdots & \ddots & \vdots \\ \langle u_{M,\boldsymbol{k}_i}|u_{1,\boldsymbol{k}_{i+1}}\rangle & \cdots & \langle u_{M,\boldsymbol{k}_i}|u_{M,\boldsymbol{k}_{i+1}}\rangle\end{bmatrix}\right)\right]\right\} \quad (9)$$

XC	$\frac{\partial P_3}{\partial u}$	$\gamma_{3,1}^{(0)}$	$\gamma_{3,3}^{(0)}$	$\gamma_{1,5}^{(0)}$	$\frac{\partial u}{\partial \epsilon_1}$	$\frac{\partial u}{\partial \epsilon_3}$	$\frac{\partial u}{\partial \epsilon_5}$	$\gamma_{3,1}$	$\gamma_{3,3}$	$\gamma_{1,5}$
LDA	-8.95	0.43	-0.87	0.48	0.09	-0.18	0.00	-0.40	0.73	0.48
PBE	-9.03	0.43	-0.88	0.48	0.10	-0.19	0.00	-0.48	0.88	0.48
LDA[23]	-	0.45	-0.84	-	-	-0.16	-	-0.49	0.73	-
LDA[22]	-	-	-	-	-	-	-	-0.44	0.86	0.30†

4. Conclusion

The LATM was shown to be able to calculate the SHG response of zinc blende crystals. The convergence of the method in the high frequency range was questionable as numerical noise seemed to persist even for a dense grid. It did however converge much faster for the low frequencies. It was shown that DFT calculations could successfully calculate the piezoelectric response of GaN.

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