Ab Initio Calculation of Dielectric Function in Wurtzite GaN Based on Walter's Model *

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The wavelength-dependent and frequency-dependent dielectric function of wurtzite-GaN is calculated totally from fundamental parameters such as the lattice constant using Walter's ab initio model. The errors occurring in the calculation are carefully reduced by linear interpolation of energy data. The Kramers–Krönig transform of the real part of greater range is obtained by extrapolation of the real part. The calculation is time-consuming but meaningful. The long-wave results are similar to the experimental data of the photon and are useful for related investigation of properties of wide-gap semiconductors such as electron scattering like the Auger recombination and impact ionization.

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Electrical and optical properties of semiconductor materials have been studied for a long time, among which dielectric function reveals the electron scattering inside materials such as the Auger recombination (AR) and impact ionization (II).[1–7] Many models of calculating dielectric function have been proposed, such as Lindhard,[8] Penn,[9] Forouhi *et al.*[10] and Djurisic *et al.*,[11] among which Walter's model[12] is one of the most widely used *ab initio* ones.

Walter's model, which is accurate in both relatively lower and higher frequencies, and effective for both optical and electrical processes, was first given by Ehrenreich *et al.*,[13] then developed by Walter *et al.*[12] to calculate the dielectric function of silicon. The calculations were also expanded to Ge, GaAs and ZnSe.[14,15] Despite its wide applications, the full Brillouin zone calculation of wavelength- and frequency-dependent dielectric function in GaN, one important widegap semiconductor material, has not been reported.

This study describes the calculation of the dielectric function in wurtzite-GaN for the first Brillouin zone in detail with frequency ranging from 0 to 20 eV. Also, linear interpolation (LIP) is used to eliminate errors and extrapolation (EP) to obtain the Kramers–Krönig transform (KKT) of the real part. Furthermore, an analysis of the electric elasticity is also given.

First, the empirical pseudopotential method[17,18] is used to calculate the first 168 Fourier coefficients of wave function and the relation between energy and wave vector because of its efficient and correct performance. Given the Bloch wave function

$$\begin{aligned}\psi_{n,\boldsymbol{k}} &= u(n,\boldsymbol{k})\exp(-i\boldsymbol{k}\cdot\boldsymbol{r}) \\ &= \sum_{\boldsymbol{G}} A(n,\boldsymbol{G},\boldsymbol{k})\exp[-i(\boldsymbol{k}-\boldsymbol{G})\cdot\boldsymbol{r}], \end{aligned} \quad (1)$$

where n refers to a certain conduction band (CB) c or valence band (VB) v, and the dispersion relation $E_n(\boldsymbol{k})$ among the first Brillouin zone, the real part of wavelength- and frequency-dependent dielectric function can be calculated by[12]

$$\begin{aligned}\varepsilon_1(\boldsymbol{q},\omega) =& 1+\frac{e^2}{q^2\pi^2}\sum_c\sum_v\int_{\boldsymbol{k}}\|(u(c,\boldsymbol{k}),u(v,\boldsymbol{k}+\boldsymbol{q}))\|_2^2 \\ &\times\{[E_c(\boldsymbol{k})-E_v(\boldsymbol{k}+\boldsymbol{q})-\hbar\omega]^{-1} \\ &+[E_c(\boldsymbol{k})-E_v(\boldsymbol{k}+\boldsymbol{q})+\hbar\omega]^{-1}\}d^3\boldsymbol{k}, \end{aligned} \quad (2)$$

where $\boldsymbol{q}$ represents the difference between two Bloch wavelengths, and $(u(c,\boldsymbol{k}),u(v,\boldsymbol{k}+\boldsymbol{q}))$ means the inner product. The above equation uses Gaussian units. Here the previous Dirac notation is changed to inner product notation because the former usually stands for the whole wave function. For real calculation we use the following equation

$$\begin{aligned}\varepsilon_1(\boldsymbol{q},\omega) = \frac{1}{q^2}(q^2 &+ 2.8660\times10^{11} \\ &\cdot\sum_{4\text{CBs}}\sum_{6\text{VBs}}\text{WFD}\cdot\text{EF}^{\text{T}}), \end{aligned} \quad (3)$$

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where wave function dot product term (WFD) and energy function term (EF) both are 1×276480 matrices. Each element of WFD and EF which represents one $\boldsymbol{k}$ is

$$\begin{aligned} WFD_{\boldsymbol{k}} &= \| \sum_{\boldsymbol{G}} A(n,\boldsymbol{G},\boldsymbol{k}) A^*(n,\boldsymbol{G},\boldsymbol{k}+\boldsymbol{q}) \|_2^2, \\ EF_{\boldsymbol{k}} &= EF_{\boldsymbol{k},-} + EF_{\boldsymbol{k},+} \\ &= [E_c(\boldsymbol{k}) - E_v(\boldsymbol{k}+\boldsymbol{q}) - \hbar\omega]^{-1} \\ &\quad + [E_c(\boldsymbol{k}) - E_v(\boldsymbol{k}+\boldsymbol{q}) + \hbar\omega]^{-1}. \end{aligned} \tag{4}$$

The imaginary part can be given by KKT from the real part

$$\varepsilon_2(\boldsymbol{q},\omega) = -\frac{2\omega}{\pi}\int_0^\infty \frac{\varepsilon_1(\boldsymbol{q},\omega')}{\omega'^2-\omega^2}d\omega'. \tag{5}$$

To obtain accurate results, the calculation covered the first four conduction bands and six valence bands of wurtzite GaN. The process was a time-consuming work due to the model's inherent defect that though precise in both high and low frequency, there are numerous singularities between them.

The first calculation adopted 276480 ($3\times48\times48\times40$, 3 refers to the 3 parts of the first Brillouin zone. For the division of each part please refer to Fig. 2(a)) large cubes when integrating it by $\boldsymbol{k}$ in the first Brillouin zone for each of the ten energy bands. However, it is found that the result does not converge as shown in Fig. 1. After carefully checking, it is noticed that the singularities will appear when the term $EF_{\boldsymbol{k},-} = [E_c(\boldsymbol{k}) - E_v(\boldsymbol{k}+\boldsymbol{q}) - \hbar\omega]^{-1}$ is calculated. The error in $EF_{\boldsymbol{k},-}$ could be calculated by

$$\delta EF_{\boldsymbol{k},-} = \frac{\sqrt{(\delta E_c(\boldsymbol{k}))^2 + (\delta E_v(\boldsymbol{k}+\boldsymbol{q}))^2}}{(E_c(\boldsymbol{k}) - E_v(\boldsymbol{k}+\boldsymbol{q}) - \hbar\omega)^2}. \tag{6}$$

To reduce the error, the error in $E_c(\boldsymbol{k})$ and $E_v(\boldsymbol{k}+\boldsymbol{q})$ should be reduced. In the second calculation each large cube in the first calculation is divided into 1000 ($10\times10\times10$, in three dimensions) small cubes. In every small cube the energy is obtained by LIP. However, the Bloch wave functions and other terms could simply adopt large cubes' data as averages. Figure 1 shows the difference between the first and the second results. A comparison with the experimental data of a photon[19] is also given in the figure. Because of the long wave approximation, the photon's data appears very similar to the calculation result for a relatively small wave vector $\boldsymbol{q} = \frac{1}{24}\boldsymbol{u} - \frac{1}{24}\boldsymbol{v}$, where $\boldsymbol{u}$ and $\boldsymbol{v}$ are the reciprocal lattice vectors of GaN in units of cm^{-1}, and their positions are shown in Fig. 2(a).

Because of the symmetry of the hexagonal first Brillouin zone as shown in Fig. 2(a), the calculation of $\varepsilon_1(\boldsymbol{q},\omega)$ could be reduced to a much smaller number of $\boldsymbol{q}$ points, such as 184 (see more details in Fig. 2(a). In fact, only 180 points were calculated in this study, 36 points for each constant layer shown in Fig. 2(a), and also 4 points for the line $\boldsymbol{w}$ which is perpendicular to this work and points out, for each $\boldsymbol{q}$, 101 frequencies from 0 to 20 eV with step 0.2 eV were calculated). Considering that simple listing of all 180 curves is meaningless, the real part of dielectric functions for some special points are shown in Fig. 2. It can be found that as the wave vector approaches the edge of the first Brillouin zone, dielectric constants' fluctuation with frequency becomes weaker and weaker. Seriously speaking, there is still noise for the near-center dielectric function. A small peak appears when $\hbar\omega$ varies from 10 to 12 eV, which remains unexplained.

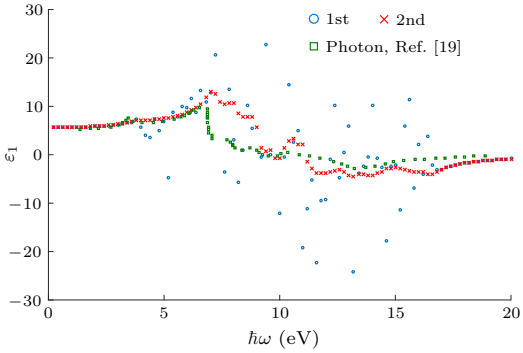

Fig. 1. Comparison of the first calculation, the second calculation and the experimental results of photon (where $\boldsymbol{q} = \frac{1}{24}\boldsymbol{u} - \frac{1}{24}\boldsymbol{v}$). The photon's data is obtained from the first figure of Ref. [19].

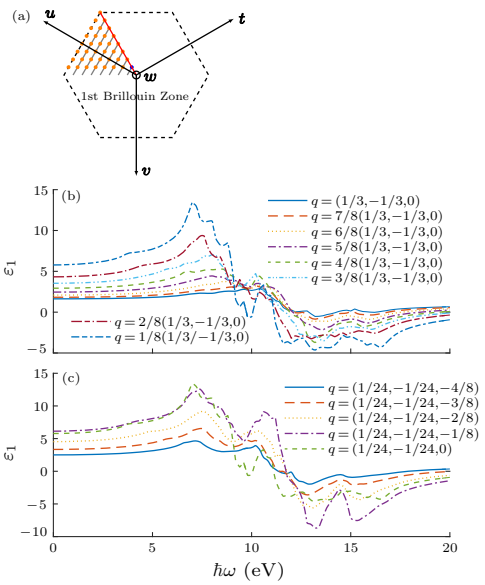

Fig. 2. The calculation results. (a) The diagram of sampling points (in plane $q_w = 0, -1/8, -2/8, -3/8$ or $-4/8$). Here $\boldsymbol{q}$ is represented with the vectors $\boldsymbol{u}$, $\boldsymbol{v}$, $\boldsymbol{w}$, which are all reciprocal lattice vectors of GaN in units of cm^{-1} and $\boldsymbol{w}$ points out of the study. (b) The real part of the dielectric function of GaN on the red line in (a) while $q_w = 0$. (c) The real part of the dielectric function of GaN at the blue point in (a) while $q_w = 0, -1/8, -2/8, -3/8$ or $-4/8$.

We wonder if the real part of the dielectric function has been acquired and the imaginary part could be obtained naturally following KKT. Since the calculation of the real part merely ranges from 0 to 20 eV, to calculate the infinite integral for higher frequencies, we need to carry out EP, which are based on the observation that the Walter-model-based dielectric func-

tion converges to 1 in a way like $1/x$ when frequency approaches infinity. One of the results is shown in Fig. 3(a).

Figure 3(b) shows that the peaks of the dielectric function vanish when $1/\varepsilon$ for the real calculation of matrix element is considered, which implies that the result is convincible to further explore other properties of GaN because in many situations such as AR and II, the dielectric function always appears alone in the denominator.

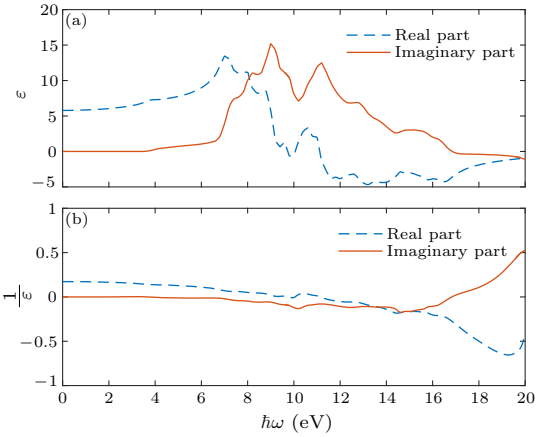

Fig. 3. The complex dielectric function of GaN ($\boldsymbol{q} = \frac{1}{24}\boldsymbol{u} - \frac{1}{24}\boldsymbol{v}$). (a) The imaginary part is given by KKT. (b) The real and imaginary parts of $1/\varepsilon$ for the same point $\boldsymbol{q}$.

In summary, it is successful to remove the errors appearing in the calculation of GaN's wavelength- and frequency-dependent dielectric function using LIP and EP. The long-wave results are similar to the experimental data of a photon.[12] Although there are still slight peaks in the frequency spectrum, the analysis of the electric elasticity shows that these results can be used for other calculations. Because the dielectric function is a very basic property of GaN, these results are very useful in its related investigation.

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