Theoretical Study of Unconventional Plasmon Generation by Solving Finite-Size 3D Hubbard Model within Mean Field Approximation

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Abstract. Recent experimental study on Strontium Niobate Oxide system has revealed unconventional plasmons generated due to confinement by oxygen planes. A phenomenological model accompanied the experimental data on that report has suggested that the confined electrons behave as harmonic oscillators. These motivate us to further study the effect of space confinement to the electrons on the formation of unconventional plasmons theoretically. For this purpose, we propose a method of modeling the generation of the unconventional plasmons in a finite system. The dynamics of correlated electrons in this confined system is described using finite-size 3D Hubbard model that is solved within mean field approximation. We study a hypothetical cubic system that consists of 5x5x5 single-orbital atoms. The calculation is done with and without incorporating the on-site Coulomb repulsion. We also consider both restricted and unrestricted mean field treatments to the calculation. Our dielectric function results show that for restricted mean field with $U = 0 \, \text{eV}$ and unrestricted mean field with $U \leq 2 \, \text{eV}$, the system is metal and only two and four unconventional plasmons found respectively. While, for $U > 2 \, \text{eV}$, it becomes insulator and shows more emergence of unconventional plasmons.

1. Introduction

Plasmonics is a research field that studies the interaction between electromagnetic field and free electrons in metals. This field of research is promising due to the fact that subwavelength light confinement in metals lead to creation of novel devices [1]. The term plasmon refers to the collective oscillation of plasma, which is an electrically neutral medium with at least one type of charge is mobile [2]. According to Drude model, the plasma frequency can be defined as

$$\omega_p^2 = \frac{4\pi ne^2}{m\varepsilon_{\text{core}}}$$

(1)

where $n$ is the total carrier density, $m$ is the mass of electron and $\varepsilon_{\text{core}}$ is the core dielectric constant.

A recent experimental study on strontium niobate oxide by Asmara et al. [3] shows that there exists plasmons which have different characteristics of real part $\varepsilon_1(\omega)$, imaginary part of complex dielectric function $\varepsilon_2(\omega)$, and loss function. We call these plasmons as unconventional plasmons which are generated due to oxygen planes confinement. Theoretical study of this
confined system suggests that the electrons behave as harmonic oscillator. The results of that research motivates us to study the effect of confinement and electron-electron interaction to the generation of the unconventional plasmons. We develop a model of a hypothetical system that consists of 5x5x5 single-orbital atoms. Our goal of this research is to calculate the density of states, complex dielectric function, and loss function of the system.

2. Model
We demonstrate a system of 5x5x5 single-orbital atoms where the mobility of the electrons is limited due to the finite amount of sites.

![Figure 1. Illustration of the system where each dot represents a single-orbital atom.](image)

Each site has index corresponds to the coordinate of the site. Following are the list of indices and their corresponding coordinate (x,y,z):

- (0, 0, 0) → 1 (1, 0, 0) → 26 (2, 0, 0) → 51 (3, 0, 0) → 76 (4, 0, 0) → 101
- (0, 0, 1) → 2 (1, 0, 1) → 27 (2, 0, 1) → 52 (3, 0, 1) → 77 (4, 0, 1) → 102
- (0, 0, 2) → 3 (1, 0, 2) → 28 (2, 0, 2) → 53 (3, 0, 2) → 78 (4, 0, 2) → 103
- (0, 0, 3) → 4 (1, 0, 3) → 29 (2, 0, 3) → 54 (3, 0, 3) → 79 (4, 0, 3) → 104
- (0, 0, 4) → 5 (1, 0, 4) → 30 (2, 0, 4) → 55 (3, 0, 4) → 80 (4, 0, 4) → 105
- (0, 1, 0) → 6 (1, 1, 0) → 31 (2, 1, 0) → 56 (3, 1, 0) → 81 (4, 1, 0) → 106
- (0, 1, 1) → 7 (1, 1, 1) → 32 (2, 1, 1) → 57 (3, 1, 1) → 82 (4, 1, 1) → 107
- (0, 1, 2) → 8 (1, 1, 2) → 33 (2, 1, 2) → 58 (3, 1, 2) → 83 (4, 1, 2) → 108
- (0, 1, 3) → 9 (1, 1, 3) → 34 (2, 1, 3) → 59 (3, 1, 3) → 84 (4, 1, 3) → 109
- (0, 1, 4) → 10 (1, 1, 4) → 35 (2, 1, 4) → 60 (3, 1, 4) → 85 (4, 1, 4) → 110
- (0, 2, 0) → 11 (1, 2, 0) → 36 (2, 2, 0) → 61 (3, 2, 0) → 86 (4, 2, 0) → 111
- (0, 2, 1) → 12 (1, 2, 1) → 37 (2, 2, 1) → 62 (3, 2, 1) → 87 (4, 2, 1) → 112
- (0, 2, 2) → 13 (1, 2, 2) → 38 (2, 2, 2) → 63 (3, 2, 2) → 88 (4, 2, 2) → 113
- (0, 2, 3) → 14 (1, 2, 3) → 39 (2, 2, 3) → 64 (3, 2, 3) → 89 (4, 2, 3) → 114
We propose a hamiltonian which is based on tight binding model as follows

\[ H = \sum_i a_{i,\sigma}^\dagger [H_0] a_{j,\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} \]

The first term of the hamiltonian corresponds to the kinetic term. \( a_{i,\sigma}^\dagger \) and \( a_{j,\sigma} \) are row vector of creation and column vector of annihilation operator respectively. \( H[0] \) is a 250x250 that can be grouped into four submatrices as follows

\[ [H_0] = \begin{bmatrix} [H_0]_{\uparrow \downarrow} & 0 \\ 0 & [H_0]_{\downarrow \uparrow} \end{bmatrix} \]

where \([H_0]_{\uparrow \downarrow}\) diagonal elements are the on-site energy \( \varepsilon \). Other elements that have index representing two neighbouring site where the difference of the coordinates \((x - x', y - y', z - z')\) equals to \( (0, 0, 1)\), \((0, 1, 0)\), or \((1, 0, 0)\) such as \( H_{1,2}, H_{1,6}, H_{1,26}, H_{2,1}, H_{2,3}, H_{2,7}, H_{2,27}, ..., H_{125,100}, H_{125,120}, H_{125,124} \) have value of \(-t\).

The second term is on-site Coulomb repulsion which represents the interaction between two electrons in the same orbital with \( U \) is the repulsion energy. This term is treated using mean-field approximation and can be written as

\[ U \sum_i n_{i\uparrow} n_{i\downarrow} = U \sum_i \left( \langle n_{i\uparrow} \rangle n_{i\downarrow} + \langle n_{i\downarrow} \rangle n_{i\uparrow} - \langle n_{i\uparrow} \rangle \langle n_{i\downarrow} \rangle \right) \]

where the occupation number \( \langle n_{i\uparrow} \rangle \) and \( \langle n_{i\downarrow} \rangle \) are calculated using self-consistent method.

3. Methods of Calculation

We define Green’s function to calculate the density of states of the system. In matrix form, the Green’s function can be written as

\[ [G(z)] = \left[ z[I] - [H_0] - [\Sigma(z)] \right]^{-1} \]

where \( z \) is complex frequency variable defined as \( \omega + i0^+ \) and \( [I] \) is a 125 x 125 identity matrix. The self-energy matrix, \([\Sigma(\omega)]\) contains interaction terms of the Hamiltonian and it will be solved using self-consistent method. Elements of \([\Sigma(\omega)]\) are all zero except for the diagonal elements \( \Sigma_i \) which has value of

\[ \Sigma_1 = \Sigma_2 = \Sigma_3 = \ldots = \Sigma_{125} = U \langle n_{i\downarrow} \rangle \]

\[ \Sigma_{126} = \Sigma_{127} = \Sigma_{128} = \ldots = \Sigma_{250} = U \langle n_{i\uparrow} \rangle \]
where the value of $\langle n_{i\uparrow}(\downarrow) \rangle$ is determined by certain initially guessed value. From the Green’s function, we can calculate the density of states (DOS) of the system through

$$DOS(\omega) = -\frac{1}{\pi} Im Tr[G(\omega + i0^+)]$$

(8)

Average electron occupancy at site $i$ with spin $\sigma$ can be calculated through

$$\langle n_{i\sigma} \rangle = \int d\omega PDOS_{i\sigma}(\omega) f(\omega, \mu, T)$$

(9)

where $PDOS_{i\sigma}$ is defined as

$$PDOS_{i\sigma} = -\frac{1}{\pi} Im [G_{i\sigma}(\omega + i0^+)]$$

(10)

The calculated $\langle n_{i\sigma} \rangle$ are then used to be the new self-energy matrix elements. This updated self-energy matrix is fed back into Green’s function as we defined in equation (5). We iterate this process until convergence is achieved.

We calculate the optical conductivity for $T = 0$ K through

$$Re\sigma_{\alpha\beta} = \frac{\pi e^2}{\hbar V} \sum_k Tr[\nu_{\alpha\beta}[A_{\alpha\beta}(\mu)][\nu_{\alpha\beta}][A_{\alpha\beta}(\mu)]$$

(11)

Real and imaginary part of complex dielectric function $\varepsilon_1(\omega)$ and $\varepsilon_2(\omega)$ can be calculated using Kramers-Kronig formula

$$\varepsilon_1(\omega) - 1 = \frac{2}{\pi} \int_0^\infty \frac{\omega' \varepsilon_2(\omega')}{\omega'^2 - \omega^2} d\omega'$$

(12)

$$\varepsilon_2(\omega) = -\frac{2}{\pi} \int_0^\infty \frac{\omega' \varepsilon_1(\omega')}{\omega'^2 - \omega^2} d\omega'$$

(13)

From the $\varepsilon_1$ and $\varepsilon_2$, we can define loss function as

$$-Im[\varepsilon^{-1}(\omega)] = \frac{\varepsilon_2}{\varepsilon_1^2 + \varepsilon_2^2}$$

(14)

4. Results and Discussion

We set the value of the parameters as follows: $\varepsilon = 0$ eV, $t = 1$ eV, $T = 0$ K, and vary the value of $U = 0$ eV, 0.5 eV, 1 eV, 2 eV, 3 eV, and 4 eV. Mean field approximation where $U = 0$ eV is said to be unrestricted, while for $U > 0$ eV is restricted due to the initial guess given for the occupation number. We work with half-filled electron system by setting $n_{filling}$ to be 125 electrons.

By ignoring the on-site Coulomb interaction between electrons, we aim to see only the effect of confinement to the formation of unconventional plasmons. We can see from dielectric function curve that it has the characteristic of multi-oscillator model [4] and interband transition due to bound charges [5, 6, 7] as at 0.75 and 1 eV. Intense peak arises from loss function curve shows the characteristic of conventional plasmons due to its correspondence to the point where $\varepsilon_1 = 0$. Other conventional plasmons can also be found at low frequency (0.1 eV and 0.8 eV) as shown in Figure 3. Setting $U = 0.5$ and 1 eV, resulting in lowering the magnitude of the peaks and slightly shifting the loss function. Therefore, conventional plasmons are no longer found at low frequency.
Figure 2. Complex dielectric function and loss function for $U = 0$ eV, 0.5 eV, and 1 eV. Upper subplot of subfigure 2(a) shows the shift of conventional plasmon. While middle subplot of figure 2(a), 2(b), and 2(c) shows the peaks of conventional and unconventional plasmons.

Figure 3. A more careful inspection of Figure 2 on lower frequency shows the emergence of both conventional and unconventional plasmons.
Figure 4. Real and imaginary part of complex dielectric function and loss function for $U = 2$, 3, and 4 eV. Unconventional plasmons can bee seen from peaks at subfigure 4(b) which aligned with dashed line.

Figure 5. More conventional and unconventional shown in region between 4.5 eV to 6 eV. Black dashed lines indicate conventional plasmons, while red and blue for unconventional plasmons which weakly interact with electrons and photons respectively.
Here we identify unconventional plasmon from \( \varepsilon_2 \) or loss function peak which does not correspond to zero crossing point of \( \varepsilon_1 \). Each of them indicated by \( \varepsilon_2 \) and loss function peaks, is said to be inactive to interaction with electrons and photon respectively. From Figure 2, we can see that unconventional plasmons are found at 2.7 eV and 2.74 eV for \( U = 0 \) eV. Two more unconventional plasmons arise at 0.2 eV and 0.85 eV for \( U = 0.5 \) eV and 1 eV as can be seen from Figure 3.

For \( U = 2, 3, \) and 4 eV, the \( \varepsilon_1 \) and \( \varepsilon_2 \) curves show the characteristic of complex dielectric function for insulator [6] as shown in Figure 4. If we focus in region between 4.5 - 6 eV as in Figure 5, we see that there exist several additional peaks of \( \varepsilon_2 \) and loss function curve, compared to Figure 2. Those peaks indicate unconventional plasmons as we defined previously. Black dashed lines indicate conventional plasmons, while red and blue for unconventional plasmons which weakly interact with electrons and photons respectively.

5. Conclusion
In conclusion, the emergence of unconventional plasmons in our model using Hubbard model for system of 5x5x5 single-orbital atoms and treated using mean-field approximation, can be seen from peaks arise from \( \varepsilon_2 \) and loss function curve. We define two types of unconventional plasmons, the first one is identified by peak from \( \varepsilon_2 \) and the other one from loss function curve which does not correspond to zero-crossing point of \( \varepsilon_1 \). Each type is inactive to interaction with electrons and photons respectively.

Calculation of unrestricted mean field in our model shows characteristic of metal and Unconventional plasmons are found at 2.7 and 2.74 eV. While, for restricted mean field with \( U = 0.5 \) eV and 1 eV, results additional unconventional plasmons at 0.2 eV and 0.85 eV. For higher Coulomb repulsion, \( U = 2 \) eV, 3 eV, and 4 eV, the system turns into insulator and several unconventional plasmons are identified in frequency range of 4.5 eV to 6 eV. Therefore, confinement of electrons in our model is able to generate unconventional plasmons.

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